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Research Article

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Effects of Axial Pressure and Pre oxidation Temperature on Coal Spontaneous Combustion Characteristics

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Abstract In order to study the spontaneous combustion characteristics of crushed coal under different stress states at different pre oxidation temperatures, the uniaxial compression equipped with temperature programmed (UCTP) device was used for pretreatment, and the characteristic temperature points and heat absorption and release changes during the oxidation process were analyzed using a thermogravimetric analyzer. The experimental results showed that the pre oxidation temperature and axial pressure have the greatest impact on the two stages of water evaporation, desorption, and oxygen absorption and weight gain in the coal spontaneous combustion oxidation process. When the axial pressure is kept constant, the characteristic temperature point of 4MPa is generally greater than 8MPa, and the characteristic temperature point of 8MPa coal sample is basically stable. The characteristic temperature of pre oxidized 70℃ coal samples varies greatly with axial pressure, which reduces the heat release and absorption of coal. The heat absorption is significantly affected by axial pressure, but the difference in heat release is not significant.

Keywords uniaxial compression, thermogravimetric analysis, characteristic temperature point, heat absorption

Introduction

China is rich in coal resources. According to the statistical data in 2021, coal consumption accounts for 56.0% of the total energy consumption [1]. Narrow coal pillar mining technology has been widely used in mines [2]. However, the problem of spontaneous combustion of coal in goaf is serious [3]. During the mining process, the concentrated stress on the narrow coal pillar is too large, the coal body is fractured and leaks air, and the air flow enters the old goaf through the coal pillar cracks, causing the old goaf to leave coal. Oxidation again, thus affecting the oxidative spontaneous combustion characteristics of the residual coal.

Aiming at the phenomenon of spontaneous combustion of crushed coal in gobs, many scholars have studied the oxidative spontaneous combustion characteristics of crushed coal. Yu Minggao[4] [4] studied the gas generation law and activation energy change of different types of coal bodies, and concluded that the oxygen consumption rate is positively correlated with the spontaneous combustion tendency, and there are significant differences in the activation energy change law; Deng Jun [5] studied the temperature-programmed experiment was carried out on coal samples, and it was found that the activation energy increased with the increase of particle size; Lu Xinxiao [6] studied the re-ignition characteristics of coal samples at different oxidation temperatures, and found that the spontaneous combustion tendency of coal samples oxidized at 80°C Higher; Ren Shuaijing [7] studied the spontaneous combustion characteristics of pre-oxidized coal samples at different temperatures during secondary oxidation; Kong Lingbo [8] studied the difference between oxidation of raw coal and oxidized coal, and found that the ignition point of oxidized coal decreased; Zhang Xinhai [9] compared the gas changes during the heating process of primary and secondary oxidation of crushed coal, and found that secondary oxidized coal is more prone to spontaneous combustion before the cross temperature; Chen Kai [10] used thermogravimetric analysis to analyze the activation energy of the combustion reaction of coal with different oxidation degrees, and found that the activation energy of coal samples treated at 70℃ and 180 min was the lowest; Xu Yongliang [11] analyzed the effect of stress on the oxidation combustion process of bituminous coal and found that the pyrolysis gas concentration, apparent activation energy, and oxygen consumption rate first increase, then decrease, and then increase in a cubic function with the increase of uniaxial stress. Chu Tingxiang [12] carried out oxidation and heating experiments on the broken coal body under different stress loading conditions, and found that the axial pressure is conducive to the low-temperature oxidation and spontaneous combustion of coal; Zhu Hongqing [13] carrying out simulation research on coal spontaneous combustion, it was found that the higher oxygen concentration in broken coal is more prone to spontaneous combustion; Yang Kai [14] analyzed raw coal, primary oxidized coal and secondary oxidized coal through ESR and FTIR, and found that the free radical types and the concentration is higher than that of raw coal.

The above studies have analyzed the relevant factors and reasons affecting the spontaneous combustion process of coal, but lack the oxidative spontaneous combustion characteristics of secondary oxidized coal samples under stress [15]. In this paper, the load pressurized coal spontaneous combustion characteristic parameter measuring device is used for pre-oxidation treatment, and the NETZSCH STA-449C thermogravimetric analyzer is used to analyze the change law of the characteristic temperature point and the heat absorption and heat release characteristics of the broken coal body under the combined action of temperature and stress. Provide theoretical guidance for prevention and control of coal spontaneous combustion in goaf.

2. Experiment

2.1 Coal sample

In the experiment, coal samples from the Queen's Coal Mine of Yangquan Coal Industry were selected, and after being mined from the working face, they were wrapped with plastic film and transported. After peeling off the oxide layer in the laboratory, the coal samples with particle sizes of 0.2-3, 3-6, and 6-10 mm were pulverized and screened, mixed uniformly according to 1:1:1, and 12 coal samples were weighed, each 0.8 kg. The coal samples were placed in a vacuum drying oven, dried at 30°C and -0.1MPa, and put into sealed bags after 48 hours for use. The industrial analysis of the coal samples used this time is shown in Table 1. The coal samples were air-dried based on weight.

Table 1: Industrial analysis results

M_{ad}/%	A_{ad}/%	V_{ad}/%	FC_{ad}/%
1.3	9.5	9.29	79.91

2.2 Experimental conditions

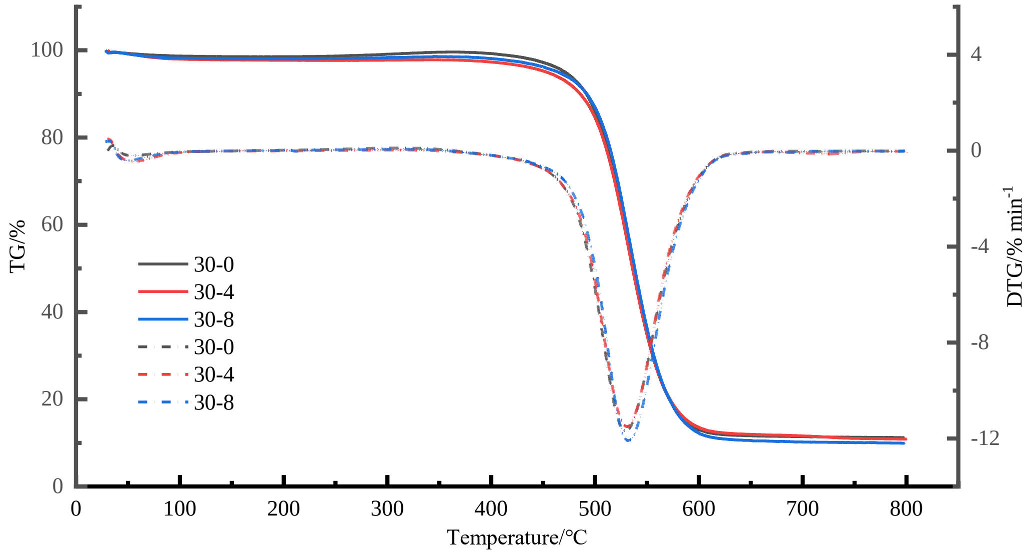
(1) The coal sample is pre-oxidized using a load pressurized coal spontaneous combustion characteristic parameter measuring device. Divide the coal samples from room temperature into three groups with pressures of 0, 4, and 8 MPa, heat them up to pre-oxidation temperatures of 30°C, 70°C, 120°C, and 180°C in an air atmosphere, and then cool them down to room temperature in a nitrogen environment as experimental coal samples. A total of 12 groups of coal samples were pretreated. The first part of the coal sample number is the pre-oxidation temperature, and the latter is the loaded axial pressure. (For example, the 70-0 coal sample is the sample of the preoxidation temperature 70°C coal sample under 0MPa).

(2) Take 15mg of coal samples from 12 groups of coal samples, and use NETZSCH STA-449C thermogravimetric analyzer to conduct thermogravimetric experiments, and the temperature range is 30-800℃.

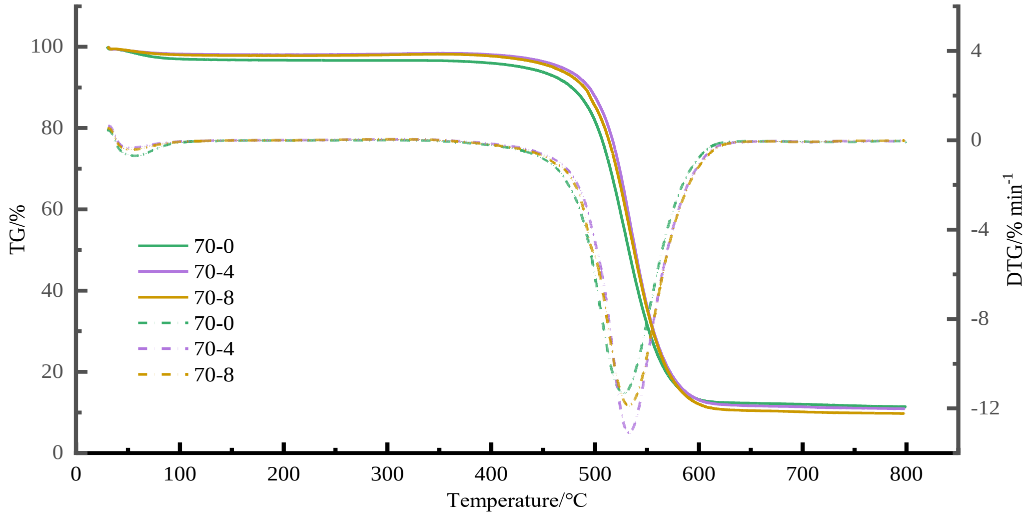
3. Experimental Results and Analysis

3.1 Coal sample characteristic temperature and stage

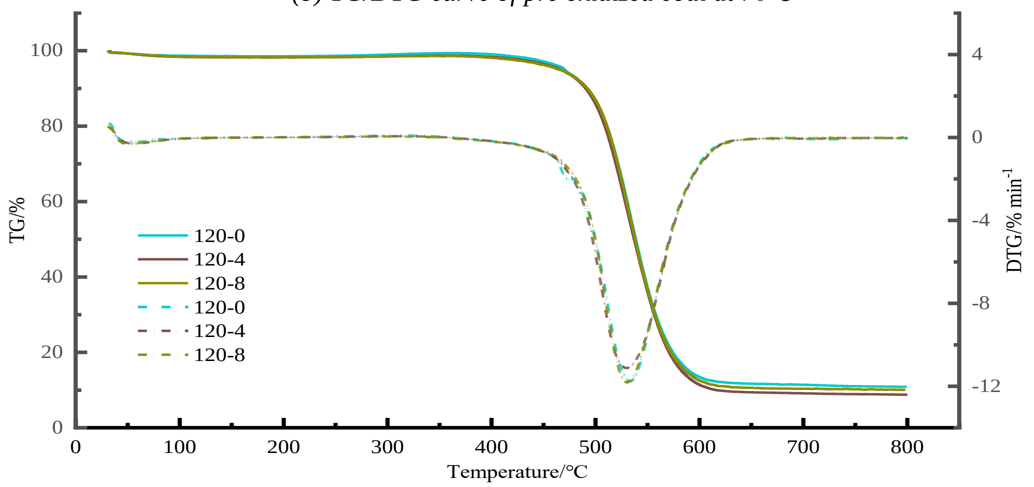
The curve obtained from the thermogravimetric experiment is called the TG curve, and the derivative of the thermogravimetric curve over time indicates that the rate of change in coal sample quality is a DTG curve. As shown in Figure 1 below.



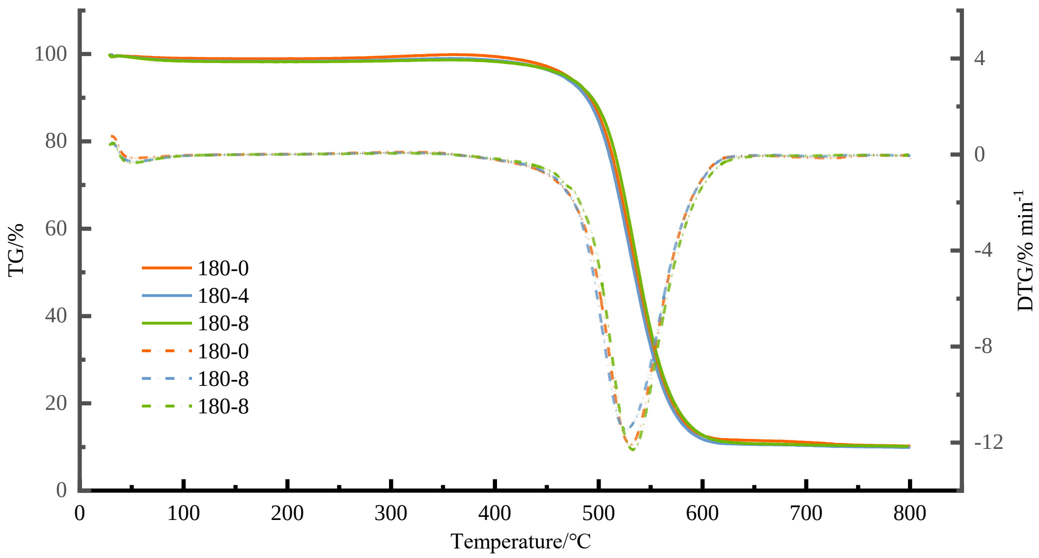
(a) TG/DTG curve of pre oxidized coal at 30℃



(b) TG/DTG curve of pre oxidized coal at 70℃



(c) TG/DTG curve of pre oxidized coal at 120℃



(d) TG/DTG curve of pre oxidized coal at 180℃

Figure 1: TG/DTG curve of coal sample

The process of coal spontaneous combustion is divided into five stages: water evaporation and desorption stage (30℃-T_2), oxygen absorption and weight gain stage (T_2-T_5), thermal decomposition stage (T_5 - T_6), combustion stage (T_6 - T_8), and burnout stage (T_8 - end of experiment). T_1: The temperature at which the maximum rate of weight loss occurs during the water evaporation and gas desorption stages during coal oxidation and combustion, the corresponding temperature at the first lowest point on the DTG curve. Dry cracking temperature T_2: The temperature at which oxygen absorption, weight gain, and physical desorption reach a dynamic equilibrium; Active temperature T_3: the temperature at which the dynamic balance ends; Growth temperature T_4: the temperature corresponding to the maximum point of coal sample mass growth rate; Thermal decomposition temperature T_5: the temperature at which the coal sample mass reaches the maximum value; Ignition point temperature T_6: the temperature at which the coal sample enters the stage of intense oxidation; Maximum weight loss rate temperature T_7: The temperature corresponding to the lowest point of the DTG curve; Burnout temperature T_8: the temperature of the coal sample after complete combustion. The characteristic point temperatures of the experimental coal samples are shown in Table 2 below.

Table 2: Characteristic temperature points of coal samples

Sample	**T_1**	**T_2**	**T_3**	**T_4**	**T_5**	**T_6**	**T_7**	**T_8**
30-0	56.3	180	190	313.1	362.3	497.4	529.6	618
30-4	54	220	255	295.1	342	497.6	530.6	624
30-8	52	188	215	293.1	350.1	501.2	531.9	615
70-0	57	174	195	302	349	503	527.8	614
70-4	53.5	193	233	318.5	353	504.4	532.5	613
70-8	53	210	221	312.6	350	500	532.4	615
120-0	51.5	186	206	314.3	362.5	500.7	533.2	623
120-4	56.7	197	217	313	357.3	497.2	529.8	617
120-8	53	190	210	296.2	350	500.4	530.1	625
180-0	55.2	187	202	316	357.3	498.9	531	612
180-4	55	195	208	312.6	354	495.6	526.1	613
180-8	50.4	190	218	295.4	360	503	532.9	623

3.1.1 Critical temperature

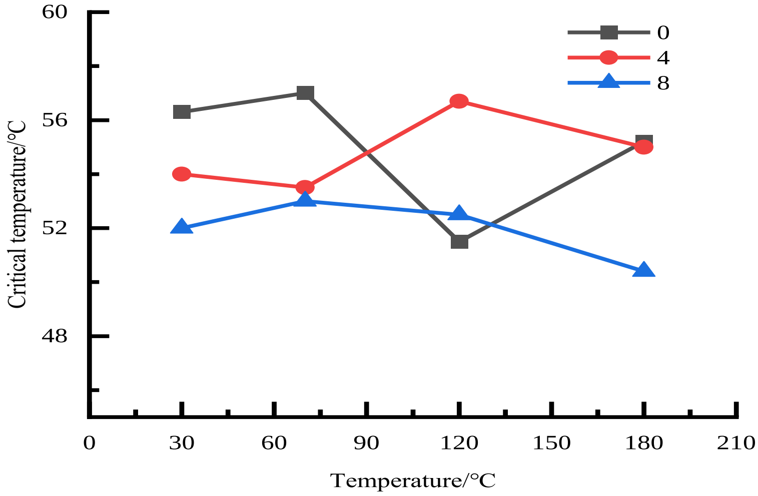


Figure 2: Change law of critical temperature of coal sample

As can be seen from Figure 2, the curve of 0MPa coal sample shows a wave valley shape as the pre oxidation temperature increases, reaching a maximum value of 57℃ at a critical temperature of 70℃, and reaching a minimum value of 51.5℃ at 120℃. The critical temperature of a 4MPa coal sample is generally higher than 8MPa, because the proportion of fine particles broken by the 4MPa coal sample is large and the amount of adsorbed gas is large. Under a certain gas flow rate, the time for water evaporation and gas desorption will be prolonged, and a higher temperature is required to reach the maximum weight loss rate of the coal sample. At a pre oxidation temperature of 70℃, the two are basically close, indicating that when the pre oxidation 70℃ coal sample is subjected to axial pressure, the magnitude of the axial pressure has a small impact on its spontaneous combustion characteristics. This is because at this temperature, there are many types and quantities of active groups in the coal, the chain reaction intensity is large, and the coal oxygen reaction conditions are sufficient, so the critical temperature is less affected by the axial pressure. The critical temperature of a 4MPa coal sample reaches the maximum value of 56.7℃ during pre oxidation at 120℃, and then begins to decrease to 55℃, indicating that a higher pre oxidation temperature will increase the critical temperature. The critical temperature fluctuation of 8MPa coal samples is minimal, and generally maintains a downward trend. Because high axial pressure will reduce the porosity of coal samples, limit the evaporation of coal moisture and gas desorption, and weaken the adsorption of oxygen by coal, the critical temperature fluctuation of 8MPa coal samples is minimal.

3.1.2 Dry cracking temperature

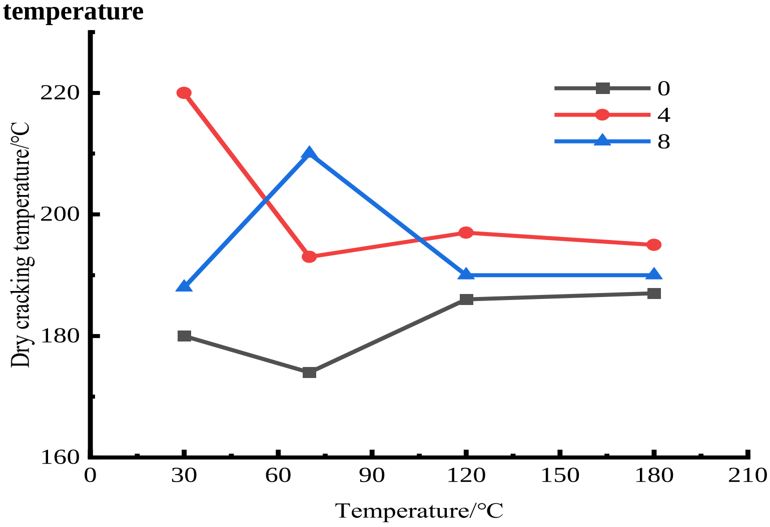


Figure 3: Variation law of dry crack temperature

Overall, the dry cracking temperature of 4MPa coal sample is the highest, followed by 8MPa, and the minimum is 0MPa. The dry cracking temperature of 4MPa and 0MPa coal samples decreased first and then increased with the increase of pre oxidation temperature, and the trend of both changes gradually stabilized. On the contrary, 8MPa coal samples increased first and then decreased. After pre oxidation treatment, the dry cracking temperature of 4MPa coal sample is least affected by the pre oxidation temperature, and the fluctuation range is within 5℃, which is basically consistent. This indicates that under this axial pressure, the internal pore structure of the coal sample does not change much, the side chains in the coal molecular structure are consistent with the dry cracking speed of the bridge, and the temperature at which the oxygen absorption and weight gain of the coal reach an equilibrium state with water evaporation and gas escape is relatively stable.

The dry cracking temperature of the pre oxidized 70℃ coal sample reaches a maximum of 210℃ at 8MPa, due to the high stacking density of the coal sample, the low flow rate of oxygen in the cracks, and the slow reaction speed of coal oxygen, which requires a higher temperature to reach the weight gain stage. The dry cracking temperature of pre oxidized coal samples at 120℃ and 180℃ is mainly affected by axial pressure, and the difference between the three is basically unchanged under different axial pressures. The difference between 4, 8, and 0MPa at 120℃ is 7℃ and 4℃, and the difference between 4, 8, and 0MPa at 180℃ is 5 ℃ and 3 ℃, respectively. When the pre oxidation temperature exceeds 100℃, the free water inside the coal sample is basically evaporated, so the temperature at the end of the water loss and weight loss stage remains basically stable.

3.1.3 Active temperature

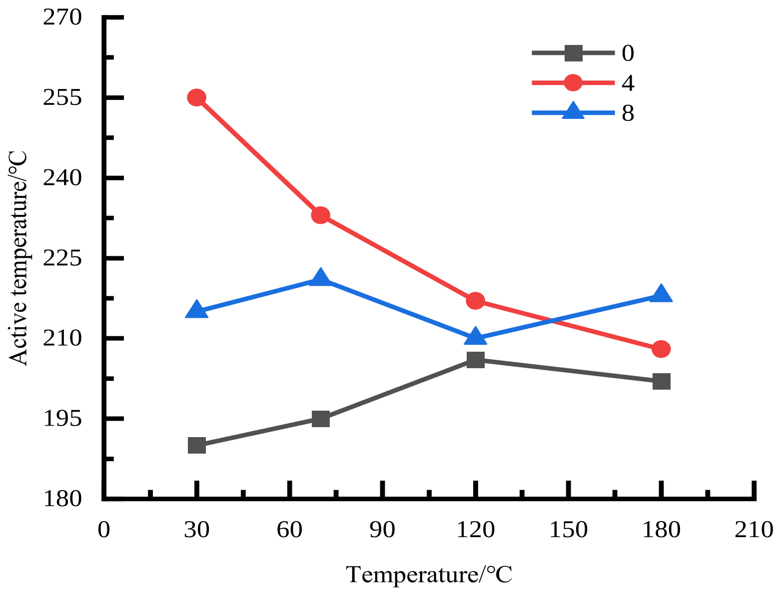


Figure 4: Change rule of active temperature

As can be seen from the above figure, the performance of the active temperature of coal samples varies greatly under different axial pressures. The active temperature of 4MPa coal samples gradually decreases as the pre oxidation temperature increases, with a significant downward trend. The active temperature decreases from 255 ℃ to 208℃, with a decrease of 47℃. The active temperature is most affected by the pre oxidation temperature at 4MPa. The 8MPa coal sample is V-shaped, with a maximum value of 221℃ and a minimum value of 210. The temperature fluctuation range is small. Overall, the active temperature of the 8MPa coal sample is relatively stable. The overall trend of 0MPa coal sample is slowly increasing. As the pre oxidation temperature increases, the three curves gradually approach each other, with the highest degree of approximation at 120℃. Subsequently, the activation temperature of 8MPa coal samples rises and deviates from the trend of convergence, while 4MPa and 0MPa further converge.

The difference in the active temperature of pre oxidized 30℃ coal samples under different axial pressures is the largest, which indicates that the proportion of particles with strong oxygen absorption is large in the coal samples subjected to axial pressure treatment. At the same time, more oxygen molecules are adsorbed on the surface area of the coal molecules, and the coal oxygen reaction rate is fast. It requires a higher temperature to make the oxygen absorption weight increase exceed the gas escape amount, breaking the dynamic balance of coal sample quality. As the pre oxidation temperature increases, the chemical activity of coal samples is higher, and the coal oxygen composite reaction is more likely to occur. The temperature required for the weight increasing stage of 4MPa and 8MPa coal samples gradually approaches. The dry cracking temperature difference between pre oxidized 120℃ and 180℃ coal samples is basically consistent under different axial pressures, while the active temperature is also relatively close. This indicates that the temperature range from the end of the water loss and weight loss stage to the dynamic balance of coal sample quality after high pre oxidation temperature coal samples is mainly affected by axial pressure.

3.1.4 Growth temperature

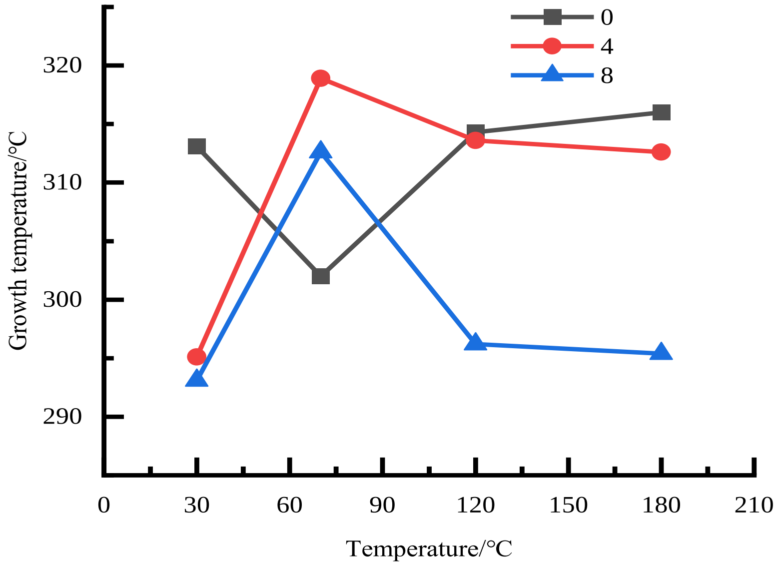


Figure 5: Temperature change law of coal sample growth rate

The growth temperature of 4MPa and 8MPa has a consistent trend with the increase of pre oxidation temperature, increasing first and then decreasing. The growth temperature of 4MPa is greater than 8MPa, but the gap between the two gradually increases and remains stable. Both growth temperatures reach the maximum values of 318.9℃ and 312.6℃ at 70℃ of pre oxidation, and then basically remain around 312℃ and 295℃. From the trend of the first half of 4MPa and 8MPa, it can be seen that under different pressure states, the growth temperature is greatly affected by the pre oxidation temperature. The trend of the second half indicates that the growth temperature is closely related to the axial pressure. The growth temperature of the 0 MPa coal sample reached a minimum of 302℃ at 70℃ for pre oxidation, and then remained approximately 315℃.

The change rules of the active temperature and growth temperature of pre oxidized 70℃ coal samples under axial pressure are consistent, with 4MPa>8MPa>0MPa. This is because after pre oxidation treatment of coal samples at 70℃, the internal active structure and chemical properties are relatively stable under different axial pressures, so the temperature ratio required to reach the active temperature and the growth temperature is basically the same. The growth temperature of pre oxidation at 120℃ and 180℃ is basically stable, as high pre oxidation temperatures will consume excessive active groups, resulting in reduced chemical adsorption of oxygen by coal, with the influence of axial pressure dominating.

3.2 Analysis of Heat Release Characteristics of Coal Samples

In the early stage of oxidation combustion, there is a heat absorption stage for coal samples, which is caused by the fact that the heat absorption by water evaporation is greater than the heat release by coal oxygen reaction during the early stage of oxidation combustion. As the reaction temperature increases, the coal oxygen reaction rate gradually increases, exceeding the water evaporation rate. On the whole, it appears that the heat absorption rate of coal samples decreases until the heat absorption rate reaches the minimum, and the heat absorption and heat release reach a balance. This temperature is called the thermal equilibrium temperature, after that, the overall performance of the coal sample is exothermic. The heat absorption, heat release, and heat balance temperatures of different pre oxidized coal samples under axial pressure are shown in Table 3.

Table 3: Heat absorption, heat release and heat balance temperature of coal sample

Sample	Heat absorption /J	Heat release /J	Thermal equilibrium temperature /℃
30-0	-144.1	13285	235
30-4	-131.3	11469	245
30-8	-120.3	12659	223
70-0	-269.8	11944	289
70-4	-111.7	12492	236
70-8	-238	12764	268
120-0	-142.4	12988	236
120-4	-145.8	12357	247
120-8	-131.4	12077	240
180-0	-134.7	13894	230
180-4	-164.8	13540	245
180-8	-201.5	13454	258

3.2.1 Heat absorption analysis

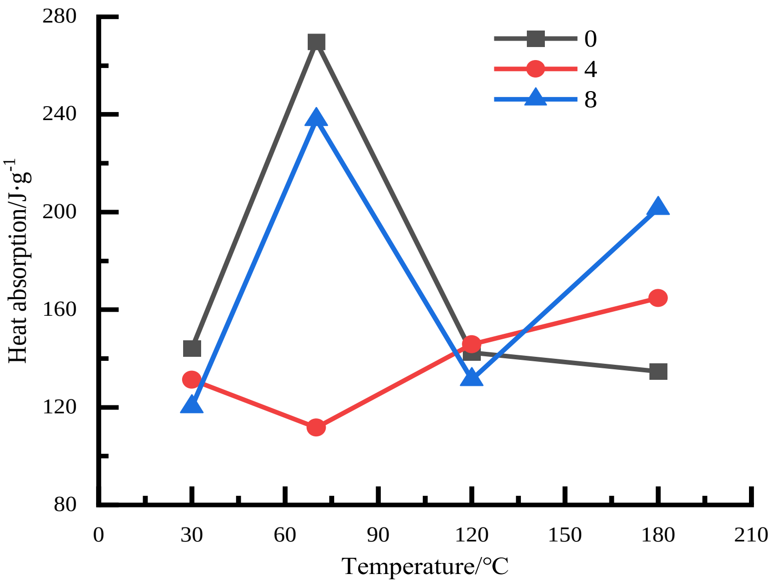


Figure 6: Heat absorption of coal samples with different treatments

Before reaching the thermal equilibrium temperature, there are significant differences in the fluctuation range of the heat absorption capacity of coal samples under different axial pressures, with the maximum fluctuation of 8MPa and 0MPa, and the minimum fluctuation of 4MPa. The heat absorption capacity of 8MPa and 0MPa coal samples reaches the highest values of 238J/g and 269.8J/g at 70℃ of pre oxidation. As the pre oxidation temperature increases, the heat absorption capacity of 0MPa coal samples gradually decreases to the minimum value of 134.7J/g, and 8MPa coal samples first decrease and then increase. The heat absorption capacity of 4MPa coal sample basically maintains an upward trend, mainly related to the internal water content of the coal sample. After 4MPa treatment, the internal water content of the coal sample is relatively low, so the overall heat absorption capacity fluctuates slightly.

Before the pre oxidation temperature of 120℃, the axial pressure will reduce the heat absorption of the coal sample, and the heat required for the coal sample to reach the thermal equilibrium temperature is small. The three axial pressure curves have the smallest difference at pre oxidation temperature of 120℃, with a heat absorption range of 131.4-145.8J/g, indicating that the impact of axial pressure on the heat absorption of coal samples is minimal at this pre oxidation temperature.

Comparing the heat absorption capacity of pre oxidized coal at 70℃ and 180℃, it can be found that the heat absorption capacity of coal under three axial pressures is exactly the opposite. The heat absorption capacity of 4MPa coal sample increases slightly, while that of 8MPa coal sample decreases slightly. The heat absorption capacity of 0MPa coal sample decreases from 269.8J/g to 134.7J/g, a decrease of about 50%. On the one hand, this indicates that the impact of axial pressure on the heat absorption capacity of coal samples at different pre oxidation temperatures is different. On the other hand, it indicates that at high pre oxidation temperatures, Axial pressure will increase the amount of heat required for coal to reach thermal equilibrium.

3.2.2 Heat release analysis

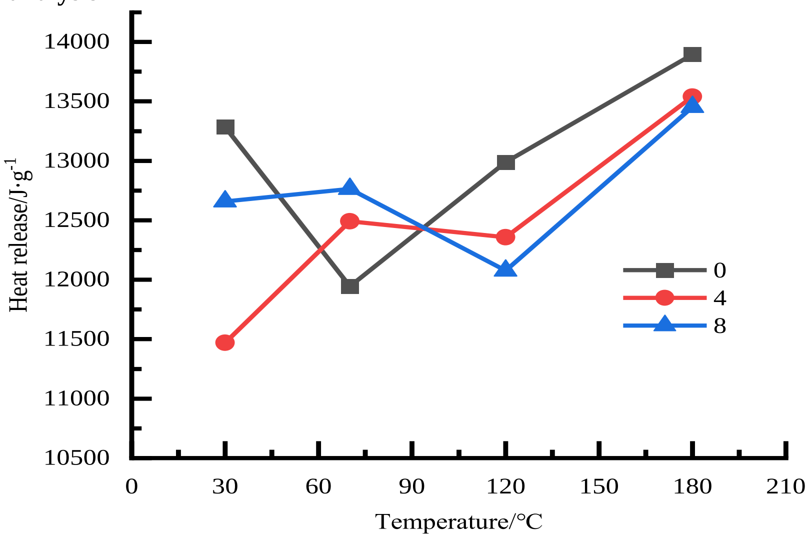


Figure 7: Heat release of different processed coal samples

From the above figure, it can be seen that the change trend of heat release of coal samples with different axial pressures is consistent with the increase of pre oxidation temperature, with an overall fluctuating increase. The highest point of heat release of the three coal sample curves is reached at 180℃ of pre oxidation, with the largest to the smallest being 13894J/g, 13540J/g, and 13454J/g. The curve has a significant upward trend between 120℃ and 180℃ of pre oxidation, and the spacing between the three curves is stable, this indicates that high pre oxidation temperature has a greater impact on coal heat release than axial pressure.

The minimum heat release of a 0 MPa coal sample at 70℃ for pre oxidation is due to the fact that the thermal equilibrium temperature of the 70-0 coal sample has reached 289℃. At this time, the coal-oxygen composite reaction is strong. Based on the oxygen consumption rate of the 70-0 coal sample, it can be seen that the oxygen consumption rate of the coal sample has reached the maximum value, and the oxygen consumption is extremely fast. The inner part of the coal sample is in an oxygen-free condition. Compared to the oxygen-free condition, the number of groups in the coal under the oxygen-free reaction condition is significantly reduced, Therefore, there is a downward trend in heat release.

The heat release of 8MPa coal sample at pre oxidation temperature of 120℃ is the smallest, and the reduction range is smaller than that of 0MPa coal sample because of the impact of axial pressure. The heat release of 4MPa coal samples after pre oxidation at 70℃ and 120℃ is basically similar, and the heat balance temperatures of both are also basically similar. However, 30℃ and 180℃ are the maximum values, indicating that pre oxidation at 70℃ and 120℃ has the same effect on the heat release of coal samples at 4MPa.

4. Conclusion

(1) The effects of pre oxidation temperature and axial pressure on the five stages of coal spontaneous combustion and oxidation process are different. The two stages of water evaporation and desorption, as well as

oxygen absorption and weight gain, are the most affected. The first half of thermal decomposition is less affected, the second half is almost unaffected, and the combustion and burnout stages are not affected.

(2) When maintaining an axial pressure of 0MPa, the variation trend of the characteristic temperature with the increase of the pre oxidation temperature is different, with T_1 decreasing in a fluctuating manner, and T_2, T_3, and T_4 increasing in a fluctuating manner. T_1 and T_2 fluctuate slightly, while T_3 and T_4 fluctuate greatly, with 16℃ and 14℃, respectively. When the axial pressure is maintained at 4MPa and 8MPa, the characteristic temperature point of 4MPa is generally greater than 8MPa. Although the T_1, T_2, and T_3 of 8MPa coal samples fluctuate, they are generally maintained at 51℃, 190℃, and 215℃, respectively. The T_1 of 4MPa coal sample is the highest at pre oxidation temperature of 120℃, and T_2 remains basically unchanged after decreasing to 195℃. T_3 gradually decreases as pre oxidation temperature increases. The variation trend of T_4 in both of them with pre oxidation temperature is consistent, increasing first and then decreasing.

(3) When maintaining the same pre oxidation temperature, T_1 and T_4 generally decrease with increasing axial pressure. T_2 and T_3 of coal samples under pressure are greater than 0MP, but decrease with increasing axial pressure. The characteristic temperature of pre oxidized coal samples at 70℃ varies greatly, while the characteristic temperature of pre oxidized coal samples at 120℃ and 180℃ varies slightly.

(4) The higher the heat balance temperature, the greater the heat absorption capacity of the coal sample. The greater the heat absorption capacity of a 0MPa coal sample, the smaller the corresponding heat release. This rule does not apply to 4MPa and 8MPa coal samples. Overall, coal samples subjected to high-temperature pre oxidation emit more heat during the pre-oxidation stage, so axial pressure will reduce the heat release and absorption of coal. The heat absorption is significantly affected by axial pressure, but the difference in heat release is not significant. The amount of heat absorption increases with the increase of axial pressure at pre oxidation temperature of 180℃, and the amount of heat release also conforms to this rule at pre oxidation temperature of 70℃.

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