

# Study on the Inhibition Characteristics of Coal Spontaneous Combustion by Silica Gel Foam

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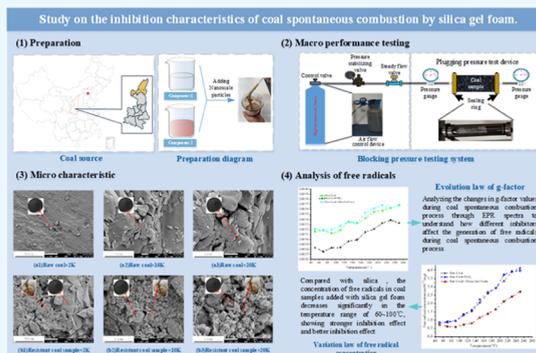
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**ABSTRACT:** The spontaneous combustion of residual coal in abandoned mining areas severely affects the safe and efficient extraction of coal, employee occupational health, and regional environmental ecology. A technical measure for preventing and controlling the spontaneous combustion of residual coal involves injecting antispontaneous combustion materials into abandoned areas. In this study, the composition, preparation, and mechanism of action of silica gel foam, a material used to suppress spontaneous combustion during coal mining, were investigated to improve the performance of materials designed to prevent spontaneous combustion in abandoned areas. The inhibitory efficiency improved, and the mechanical strength and stability of the foam liquid film increased upon adding modified antioxidants and nanosilica particles to the gel foam. Macro performance tests, microstructural characterization, and chemical inhibition mechanism analyses verified the efficacy of silica gel foam for suppressing spontaneous combustion. The air leakage resistance of the silica gel foam effectively increased the air leakage resistance of the coal samples at different pressures. New radicals formed during the spontaneous combustion of coal comprising different inhibitors, as indicated by the  $g$ -factor results of electron paramagnetic resonance (EPR) spectroscopy analysis; the formation of radicals initially decreased and then increased when the inhibitor material changed. The concentration of free radicals decreased markedly during the spontaneous combustion process of both raw and inhibited coal samples at low oxidation temperatures ( $\sim 60$ – $100$  °C), indicating a marked inhibitory effect.



## 1. INTRODUCTION

Spontaneous coal combustion is considered a major disaster in mines and affects the development of coal safety and efficient and intelligent mining technology in various countries while causing safety, economic, ecological, and social problems.<sup>1,2</sup> More than 90% of mine fires are caused by spontaneous combustion.<sup>3,4</sup> Spontaneous coal combustion occurs when coal and oxygen come into contact with each other and produce a large amount of heat via an oxidation reaction; the oxidation reaction causes the temperature of the medium to rise, owing to ineffective heat dissipation, thereby leading to spontaneous coal combustion. The coal–oxygen composite law describes the oxidation reaction that occurs when coal and oxygen come into contact with each other.

The reaction rate of the oxidation increases with increasing concentration of oxygen<sup>5</sup> and can be lowered by plugging. Plugging reduces the contact between the coal and oxygen.<sup>6</sup> In addition, the heat release rate of the coal–oxygen composite can be reduced by lowering the concentration of surface-active groups on coal.<sup>7</sup> Moreover, because the coal–oxygen composite rate increases with temperature, lowering the coal temperature could also reduce the coal–oxygen composite rate.<sup>8</sup> The prevention and control of the spontaneous combustion of residual coal require further in-depth research

because spontaneous coal combustion can have severe ramifications.

The spontaneous combustion of residual coal could increase the occurrence rate of major accidents, such as gas and coal dust explosions, thereby posing hidden dangers to the health and safety of frontline production personnel.<sup>9</sup> Additionally, coal continues to self-ignite, consuming valuable coal resources and producing harmful gases, such as CO, H<sub>2</sub>S, and CO<sub>2</sub>, thereby causing air pollution.<sup>10</sup> The heat generated by coal self-ignition affects the survival of surface crops, causing reduced production or crop failure, affecting the productivity and life of residents.<sup>11</sup> Moreover, with the continuous development of technologies such as comprehensive mining and top coal caving,<sup>12</sup> improvement of coal mining efficacy, increase in coal residue in the goaf, and intensification of air leakage, the risk of coal self-ignition is higher.<sup>13</sup> Many

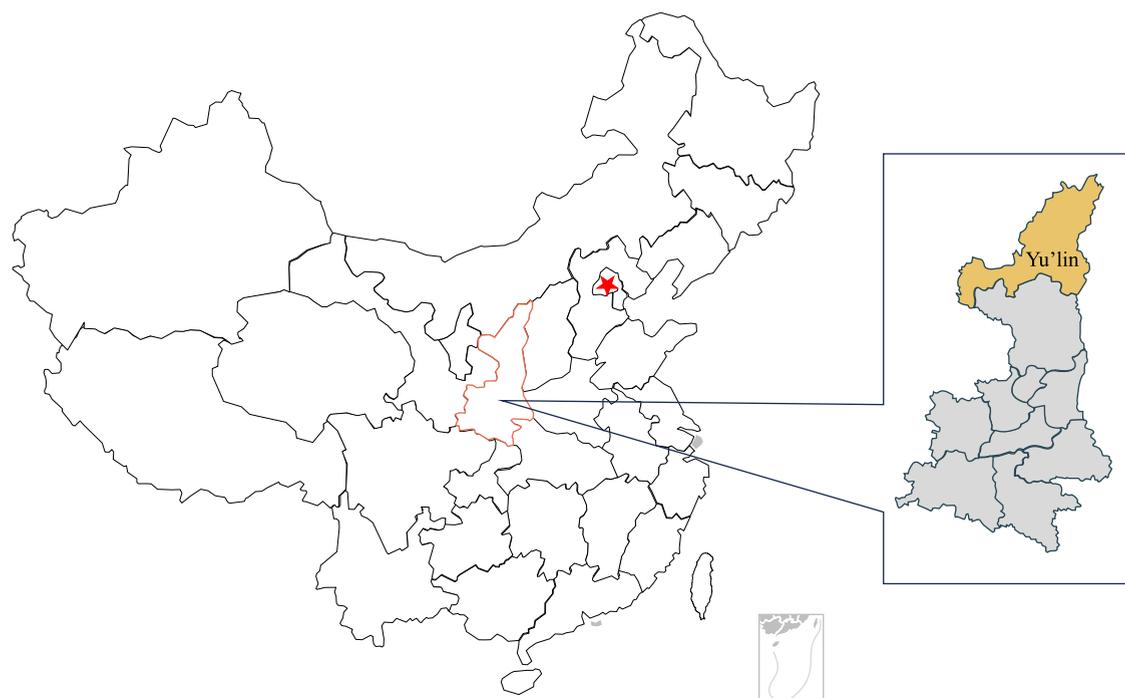
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**Figure 1.** Schematic illustration of the Changyan coal production area.

technologies have been developed to reduce the possibility of spontaneous coal combustion, one of which is gel foam technology.

Gel foam<sup>14</sup> is an advanced form of three-phase foam technology and exhibits superior solid–water retention and fluidity; additionally, it is widely applicable. Gel foam is extensively used for fire prevention and extinguishing fires in coal mines.<sup>15</sup> Many researchers have developed unique materials to suppress coal combustion. Xi et al.<sup>16</sup> developed a novel thermoplastic foam material synthesized from a sodium silicate solution, organic acids, polycaprolactone, and poly(ethylene oxide) to suppress spontaneous coal combustion. Similarly, Wang et al.<sup>17</sup> prepared a novel biomass composite thermoresponsive gel using konjac glucomannan (KGM) and fly ash (FA), which inhibited the spontaneous combustion of coal. Vinogradov et al.<sup>18,19</sup> synthesized ultrafast silica gel foams, which can coat combustible materials in forests. Owing to its strong water absorption, solid–water retention, adhesion, and viscosity, ultrafast silica gel foams can remain longer on combustibles, act swiftly to cool, and enhance the efficiency of gel foam in extinguishing fires.<sup>20</sup> Wu et al.<sup>21</sup> combined the advantages of foam, gel, and inert gases to develop a gel foam that markedly reduced reignition after extinguishing a fire. Tian et al.<sup>22</sup> prepared a new type of gelatin protein foam that physically outperformed the commercial film-forming fluoroprotein foam (FFFP). Tang et al.<sup>23</sup> established the optimal concentration of the foam components, SDBS, guar gum, and  $MgCl_2$  to produce a fire-retardant gel foam that works by inhibiting the reaction of coal–oxygen complexes, thus accomplishing a flame-retardant effect. Thus, using gel foam in fire extinguishing has many advantages, and its potential for use is high. Researchers continue to explore ways to further improve the performance of gel foams, seeking new materials that combine a controllable gelation time, good thermal stability, high extinguishing efficiency, and strong viscoelastic properties.

This study addresses the common problem of spontaneous combustion in the coal mining industry, which affects the safe operation of coal mines and can lead to notable economic losses and environmental issues. We developed a silica gel foam composite material to prevent spontaneous coal combustion by considering the advantages of existing antispontaneous combustion materials. The aim was to effectively inhibit the spontaneous combustion of residual coal using its unique oxygen barrier properties, rheological characteristics, and wide practical applicability, thus ensuring the safe operation of coal mines and reducing economic losses.

## 2. THEORY AND EXPERIMENT

**2.1. Inhibition Mechanism.** Silica gel foam was selected as a multiphase and multicomponent dispersion system to prevent the spontaneous combustion of leftover coal in goaf areas by considering the characteristics of the spontaneous combustion of coal and the necessity for materials for its prevention and control. Silica gel foam comprises five basic components: water, gel, foam, antioxidants, and silica nanoparticles. Combining these components in varying ratios results in silica gel foams with different shapes and coal self-ignition resistance properties. The selection and functions of each component are as follows.

- (1) Water: Water is the solvent and carrier of silica gel foam and has a high specific heat capacity, excellent fluidity, and significant heat absorption and cooling functions. After water absorbs heat and vaporizes, the water vapor expands in space and periodically cuts off the oxygen supply.<sup>24</sup> Therefore, the fire prevention function of water mainly involves heat absorption and cooling, slowing down heat accumulation, and cutting off the oxygen supply.
- (2) Gel component: The gel component can improve the water retention and cooling characteristics of the foam, that is, increase the water content in the foam, enhance

the heat absorption capacity of the foam, and reduce the temperature of the coal seam.<sup>25</sup> The gel component can also improve the spatial retention time of the foam, that is, increase the viscosity and adhesion of the foam, so the foam can better adhere to the surface and cracks of the coal seam, thereby prolonging its inhibitory effect.<sup>26</sup>

- (3) Foam component: The foam component can imbue the newly developed material with its characteristics and improve its wetting, dispersion, fluidity, water saving, and spatial coverage. The foam component mostly belongs to the surfactant component, which could improve the wetting of the gas–liquid interface between water and coal. Additionally, the foam component could enhance the fluidity of the gel system, thus balancing the overall performance of the new material.<sup>27</sup>
- (4) Antioxidant inhibitor components: Antioxidants are known to scavenge free radicals, and the main function of the antioxidant component is to suppress the spontaneous combustion process of coal–oxygen, use chemical inhibitors to directly cut off the chain chemical reaction of coal–oxygen, reduce the chemical reaction heat, directly block the coal–oxygen reaction, and improve inhibition efficiency.<sup>28</sup>
- (5) Silica nanoparticles: Silica nanoparticles modify and enhance silica gel foam. They can aggregate on the foam liquid film, form a stable network structure, enhance the mechanical strength of the foam liquid film,<sup>24</sup> and slow down the foam liquid separation and coarsening effects, thereby improving the stability and durability of the silica gel foam.<sup>29</sup>

## 2.2. Preparation of Coal Samples and Inhibitors.

**2.2.1. Coal Sample.** Long-flame coal produced by the Daliuta Coal Mine of Shendong Company, Shenfu Coalfield, Yulin City, Shaanxi Province, China (Figure 1), was used as the research object in this study. Fresh coal blocks were collected, sealed, and stored at 5 °C for subsequent use following the method described in the “Coal and Rock Sample Collection Method” (No. GB/T 19222-2003). Subsequently, the coal blocks were processed, by first removing the surface of the coal block exposed to the air, thereafter removing the central portion of the coal block, and crushing it through a jaw crusher, following the method described in “Coal Sample Preparation Method” (GB/T 474-2008).

Thereafter, the crushed coal particles were sieved using a standard sieve, and 80–120 mesh (0.125–0.180 mm) coal samples were packed in sealed bags and stored at a low-temperature (5 °C). The obtained coal powder was dried in a constant temperature box at 20 °C for 48 h and denoted as the lignite raw coal sample of the blank group for subsequent inhibition coal sample experiments. The lignite coal sample was subjected to three analyses using a GF-A6 automatic industrial analyzer, and the average results are listed in Table 1.

**Table 1. Industry and Elemental Analysis of Coal Samples**

industrial analysis, %				
$M_{ad}$	$A_d$	$V_{daf}$	FC	
6.03	30.11	23.17	39.77	
elemental analysis, %				
C	H	O	N	S
79.22	5.71	13.49	1.14	0.60

Coal comprises aromatic rings, side chains, and functional groups.<sup>30</sup> The core is a polyaromatic ring system that is primarily composed of carbon, hydrogen, and oxygen.

**2.2.2. Preparation of Silica Gel Foam.** The foaming agent was sodium dodecyl sulfate (SDS, 99.9%, Tianjin Comio Chemical Reagent Co., Ltd.). The gel agent was sodium polyacrylate (99.9%, Sinopharm Group Chemical Reagent Co., Ltd.) The cross-linking agent was konjac gum (food grade, 85%, Zhongchen Biotechnology Co., Ltd., Henan, China). The foam stabilizer was xanthan gum (food grade, 99%, Pengda Chemical Co., Ltd., Shandong, China). The nanomaterial was nanosilica (20 nm, Yuante New Material Co., Ltd., Suzhou, China). The antioxidant was *tert*-butyl hydroquinone (TBHQ, 99%, Taikai Food Technology Co., Ltd., Jiangxi, China). The modification reagent was montmorillonite (99%, Shanlin-Shiyu Mineral Products Co., Ltd., Hunan, China).

Antioxidants can significantly delay or prevent the oxidation of substrates at low concentrations by scavenging free radicals. There are generally two types of antioxidants: natural and synthetic. Compared with natural inhibitors, synthetic inhibitors have notable advantages in terms of their efficacy, cost, and scale of production and are widely used. Studies have shown<sup>31,32</sup> that the antioxidant activity of TBHQ is more than 100 times that of vitamin C and that TBHQ has better antioxidant effects on various substances than those of other synthetic inhibitors. Moreover, researchers have attempted to introduce natural inhibitors such as tea polyphenols, anthocyanins, and L-ascorbic acid into composite inhibitors, which are effective in preventing spontaneous coal combustion.<sup>33–35</sup> However, there are relatively few studies on the use of synthetic inhibitors, such as *tert*-butyl hydroquinone (MMT), to prevent spontaneous coal combustion. Because of the marked advantages of TBHQ in terms of its antioxidant performance, we selected TBHQ as the basic antioxidant component of an antioxidant gel foam to prevent spontaneous coal combustion. However, owing to the facile volatilization of TBHQ at high temperatures, which reduces the antioxidant content, modified materials are needed to improve water retention and thermal stability. The modified material for this preparation experiment was montmorillonite (MMT). Chemical modification of the raw materials notably improved the antioxidant performance of TBHQ in high-temperature environments.<sup>36</sup>

The modification principle is based on the monomer solution intercalation polymerization method in which TBHQ can undergo a free-radical reaction under the action of an appropriate amount of potassium persulfate initiator, resulting in the breaking of hydrogen bonds in the hydroxyl group, thereby forming an active center. The generated active centers react chemically with the methyl and methylene groups in aliphatic acrylic acid/sodium acrylate, forming ether bonds and grafting TBHQ onto the acrylic acid/sodium acrylate molecules. Under high temperatures and in the presence of a potassium persulfate initiator (KPS), the acrylic acid monomer polymerizes to form longer carbon chains. Subsequently, the large molecular carbon chains of the poly(acrylic acid) salt are cross-linked using a cross-linking agent, forming a high-molecular-weight hydrogel with the poly(acrylic acid) salt as the skeleton, which has a three-dimensional network structure with a main chain, side chain, and cross-linking degree.<sup>37</sup>

The preparation process involved mixing the foaming agent (sodium dodecyl sulfate), foam stabilizer (xanthan gum), cross-linking agent (konjac gum), gelling agent (sodium

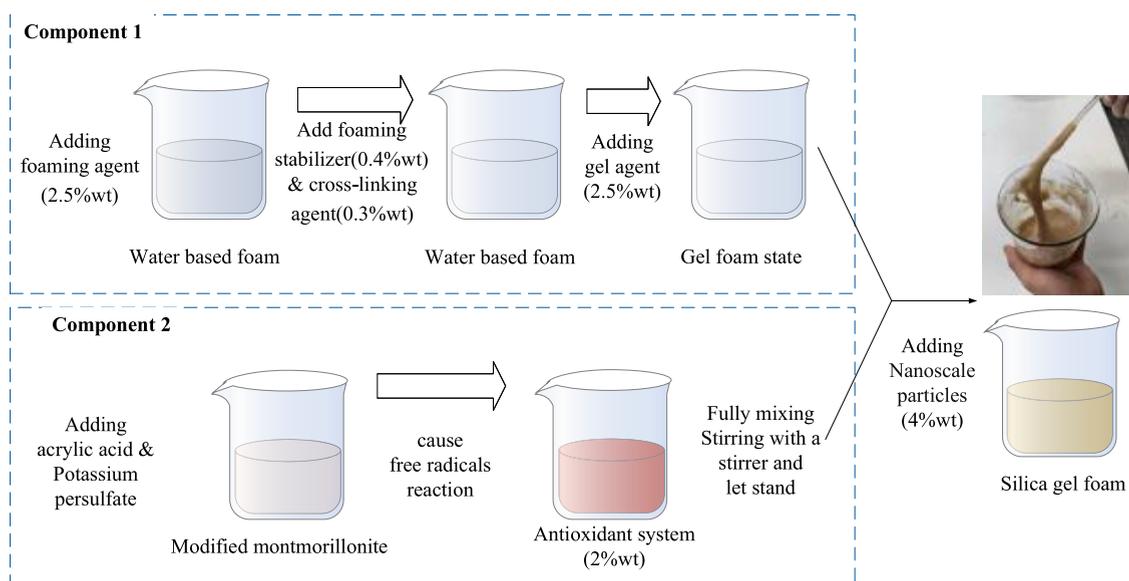


Figure 2. Schematic illustrating the preparation of silica gel foam.

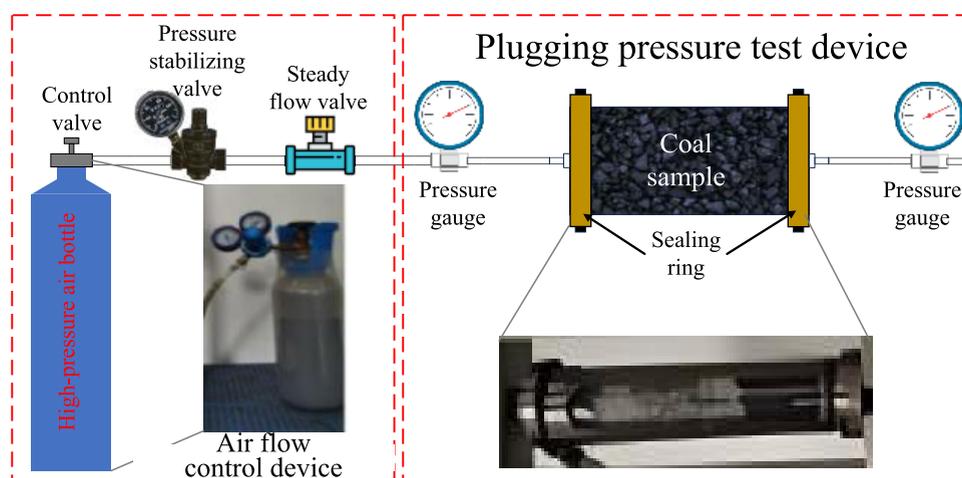


Figure 3. Blocking pressure testing system.

polyacrylate), nanoscale particles ( $\text{SiO}_2$ ), and modified “montmorillonite-acrylic acid-*tert*-butyl hydroquinone” antioxidant system in a certain proportion to obtain silica gel foam. First, modified montmorillonite (O-MMT) was obtained, and potassium persulfate, acrylic acid, and TBHQ were added to the O-MMT. The mixture was heated in a water bath to initiate the free-radical reaction and was cooled after approximately 120 min to obtain 50 mL of a 2 wt % modified antioxidant solution system. During the cooling process of the antioxidant system, 50 mL of water was weighed and mixed with a 0.3 wt % foaming agent, and a water-based foam was prepared. Thereafter, 0.4 wt % foam stabilizer was added to the water-based foam system and stirred for 5 min, and the foam height was stabilized, following which, 0.3 wt % cross-linking agent was added. After the solution in the beaker became pale yellow, 2.5 wt % gelling agent and 4 wt % nanoscale particles were added. The mixture was stirred while adding the previously prepared antioxidant system, finally completing the preparation of silica gel foam, as shown in Figure 2.

**2.3. Test Experiments.** 2.3.1. *Performance Testing Platform.* To test the efficacy of silica gel foam in preventing the spontaneous combustion of residual coal, we constructed a

sealing performance testing apparatus based on a permeation fluid loss instrument (Haitongda Special Instrument Co., Ltd., Qingdao, Shandong, China) while considering the specific needs of our research. The designed experimental platform consisted of an airflow control device (air pump, pressure gauge, and flow stabilizer) and a sealing pressure testing device (pressure gauge, 500 mL of acrylic transparent column). Figure 3 illustrates the experimental setup of the performance testing platform. The key feature of this experimental platform is that it allows the testing of silica gel foam representing different proportions of the original coal mass while maintaining a constant gaseous environment. During the experiment, we recorded the changes in the inlet and outlet pressures to evaluate the sealing capability of the silica gel foam in the air leakage channels.

For the experiment, the exposed surface of long-flame coal blocks was removed, and the remaining coal was crushed into coal particles of 40–70 mesh (0.42–0.21 mm) using a jaw crusher; the crushed coal particles served as the blank group raw coal samples. Subsequently, silica gel foam was added in proportions of 0 wt % (control group), 2, 4, and 6 wt % to the original coal mass to create differently inhibited coal samples.



**Figure 4.** Illustration of the blocking pressure testing system.

The experimentally inhibited coal samples were then loaded into a sealing pressure testing apparatus and left to settle for 20 h.

Finally, the experimental setup was connected, gas supply was enabled, and the gas inlet rate was increased linearly using a flow stabilizer. To ensure a steady flow of the airstream and avoid affecting the measurement of the accuracy of the gas pressure, a pressure regulator was installed to control the linear increase in the gas pressure. The data were then collected.

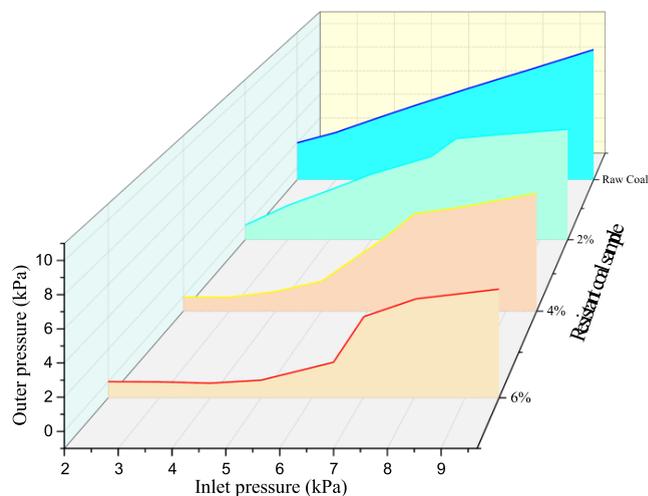
**2.3.2. Free-Radical Testing System.** All of the free-radical measurement experiments in this study were performed at a constant temperature of 25 °C. First, a specially made free-radical determination tube was inserted into the sample to obtain an appropriate amount (2 mg) of the sample, and the determination tube and sample were slowly transferred into the test cavity of the electron spin resonance spectrometer; the instrument heating, test, and gas supply modules were enabled and adjusted; the coal sample was scanned using an X-band magnetic field continuous wave. The experimental parameters were as follows: an aliquot of 1 mg of coal powder was used. The center magnetic field was 323.286 mT, scan width was  $4 \times 2$  mT, modulation frequency was 100.00 kHz, scan time was 30 s, modulation width was  $1.0 \times 0.1$  mT, microwave frequency was 9052.673 MHz, time constant was 0.03 s, temperature was increased from room temperature (20 °C) to 200 °C, and the heating rate was 2 °C/min. The free-radical measurement system is illustrated in Figure 4.

### 3. RESULTS AND DISCUSSION

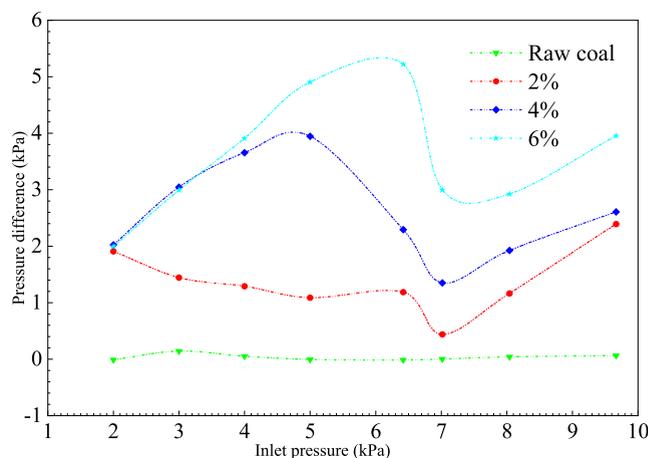
**3.1. Analysis of Macroscopic Characteristics of Blocking Oxygen and Preventing Coal Spontaneous Combustion.** By implementing the sealing system performance test experiment, we successfully obtained the relationship curve between the inlet and outlet pressures under different pressure conditions; the specific data are shown in Figure 5. During the experiment, we carefully monitored and measured the performance indicators of the sealing system to ensure the accuracy and reliability of the results.

As shown in Figure 6, when no silica gel foam is added, the pressure difference of the original coal sample increases linearly with the inlet pressure of the sealing pressure test tube. The outlet end pressure also increases linearly, and the pressure values of both spectral and electrical forces are close to zero. This occurrence indicates that the original coal sample developed cracks and strong permeability and that the original coal had almost zero sealing ability for air penetration.

After adding 2 wt % silica gel foam to the coal sample, when the inlet pressure was increased to 2.0 kPa, the outlet pressure

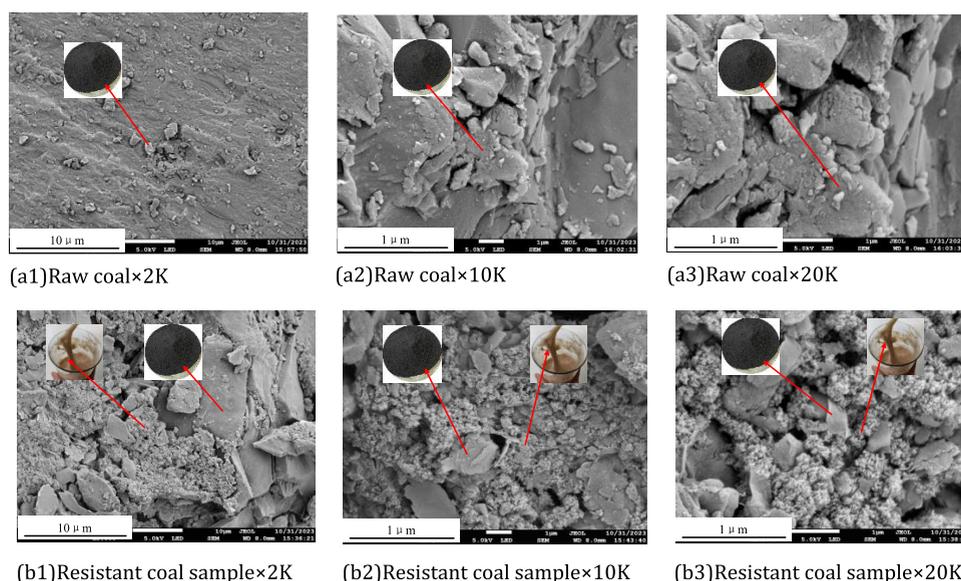


**Figure 5.** Relationship between the inlet pressure and outlet pressure.



**Figure 6.** Relationship curve between inlet pressure and pressure difference.

remained close to 0 Pa, and the corresponding pressure difference was 2.0 kPa. Similarly, when 4 and 6 wt % silica gel foam were added to the coal sample, the inlet pressure corresponding to the outlet pressure breaking to zero increased to 3.1 and 5.0 kPa, respectively. Thus, compared with that of the original coal sample, the leakage resistance of the inhibitory coal sample increased by 2.0, 3.1, and 5.0 kPa, respectively. As the inlet pressure increased, the pressure difference between the inlet and outlet decreased marginally and fluctuated; the decrease in pressure and the fluctuation occur because an



**Figure 7.** Scanning electron microscopy images of raw coal and resistance coal samples.

increase in pressure causes an increase in the flow velocity; however, the air leakage channel expands under pressure, an effect similar to that of hydraulic fracturing. These factors interact with each other to cause fluctuations.

The addition of silica gel foam markedly improved the air leakage resistance of the broken original coal. As the proportion of silica gel foam gradually increased, the air leakage resistance of the inhibited coal sample also increased. This trend indicates that in the process of mixing silica gel foam with broken coal particles, the foam is wetted and adsorbed onto the coal surface via its surface-active components, which include the foaming agent. The foam adsorbed on the coal surface effectively filled the air leakage channels between the coal particles, thereby markedly increasing the air leakage resistance. Simultaneously, the gel component in the silica gel foam formed a gel structure via gelation, which enhanced the strength and viscosity of the foam liquid film and increased the flow resistance of the foam while improving the stability of the filling foam liquid film. These changes comprehensively improved the air leakage resistance of the inhibition coal sample and the air leakage sealing ability of the silica gel foam.

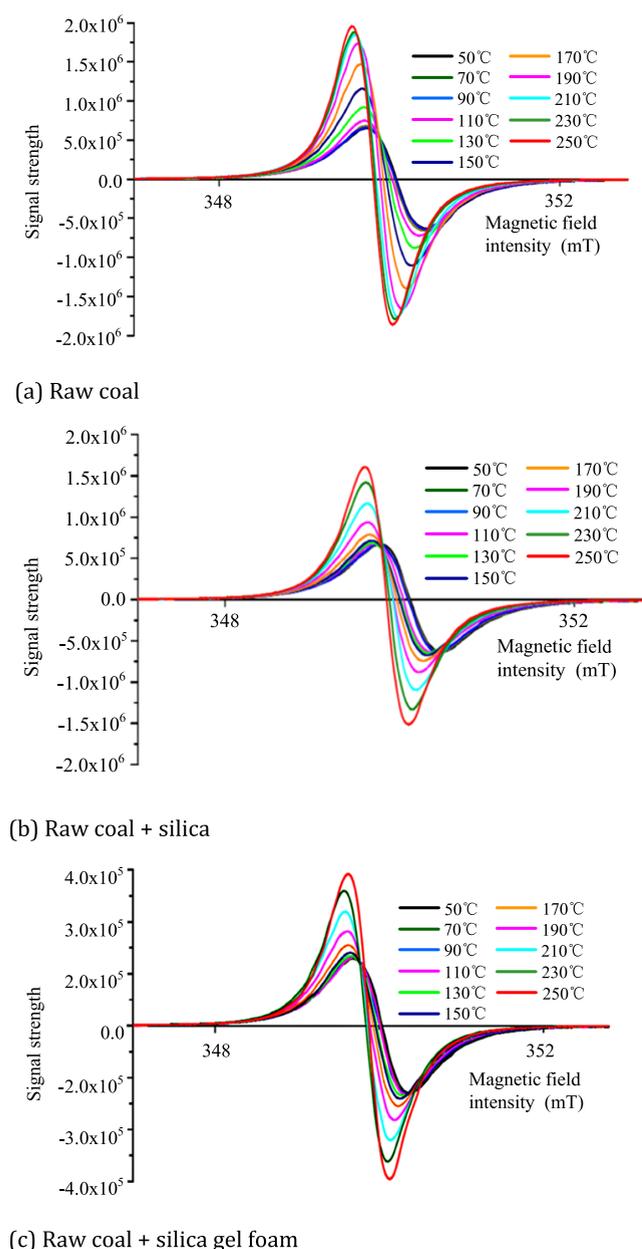
**3.2. Microscopic Characteristics of Silica Gel Foam Materials.** According to Figure 7a1–a3, the original coal particles were small and dispersed, and their surfaces were irregularly shaped with a large specific surface area, thus increasing the surface area exposed to oxygen. By contrast, a layer of adherent material was observed to cover the coal surface in the inhibition coal sample, which clearly indicated the adsorption of silica gel foam on the coal particle surface. For direct contact between the original coal surface and oxygen, after the inhibition treatment, the coal surface was coated with an antioxidation gel foam. The silica gel foam and coal surface exhibited an adsorption effect, forming a surface adsorption layer composed of silica gel foam, which effectively blocked the direct contact of oxygen with the coal surface. Therefore, this phenomenon accomplished the macroscopic prevention of silica gel foam on the coal surface and prevented spontaneous coal combustion. This finding explains the antioxidation mechanism of silica gel foam at the macroscopic

level and provides a substantial basis for the engineering application of preventing spontaneous coal combustion.

Silica gel foam is an inhibitor with high fluidity that can effectively prevent spontaneous combustion of coal. Its inhibition mechanism is mainly reflected in space and on the surface. In space, silica gel foam could fully penetrate the cracks of the goaf, especially the high-permeability areas, forming a three-dimensional covering layer, thus minimizing the contact between air and coal and reducing the oxygen supply. On the surface, silica gel foam has a surface-wetting property, which allows it to adhere closely to the surface of coal; simultaneously, due to the shape of foam, it could undergo the Jamin effect; the Jamin effect refers to the additional resistance that occurs when dispersed nonwetting phase fluids (such as bubbles or droplets) pass through pore throats or narrow openings during liquid–liquid or gas–liquid two-phase percolation, thus blocking the pores, thereby notably reducing the surface area of coal exposed to air and reducing the reaction channel of free radicals and oxygen.

**3.3. Analysis of Free Radicals during Spontaneous Combustion of Raw Coal and Blocked Coal Samples.** To compare and analyze the inhibitory effect of silica gel foam on the spontaneous combustion of residual coal in the goaf, antioxygen spontaneous combustion materials were added to the original coal at a standard of 15% of the original coal. In addition, based on the single-variable principle, a 15% addition ratio was used to add a SiO<sub>2</sub> suspension with a mass fraction of 20% to the original coal to serve as the original coal control group. Thereafter, the mixed coal samples were placed in a vacuum drying oven, and the vacuum drying mode was enabled, maintaining a low-temperature environment of 40 °C and drying for more than 30 h to reduce moisture, thereby eliminating the influence of external moisture on the experiment. The samples were sealed and stored after cooling.

**3.3.1. Electron Spin Resonance Spectra during the Spontaneous Combustion Process of Raw Coal and Resisted Coal Samples.** Electron spin resonance spectra were obtained during the spontaneous combustion process. The spectra of the original coal control group and the coal samples containing silica gel foam are shown in Figure 8. As



**Figure 8.** Electron spin resonance spectra obtained during the spontaneous combustion of raw coal and resisting coal samples.

the temperature increases, the electron resonance signal intensity of the coal body increases, and the peak area increases. Thus, temperature is still one of the important factors affecting free radicals, and the importance of the in situ measurement of free radicals is highlighted.

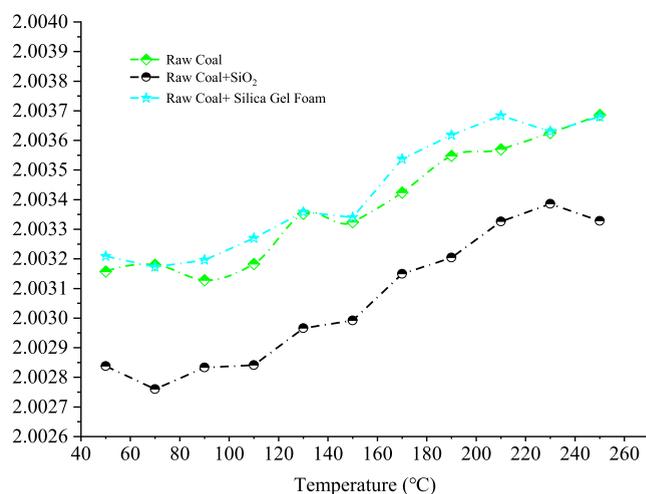
**3.3.2. Evolution Rules of  $g$ -Factor.** During spontaneous coal combustion, the condensation of benzene rings and the fracture of side chains and chemical bonds of macromolecular structures trigger the formation of a large number of new free radicals. These free radicals have more active chemical properties and combine easily with oxygen; additionally, the free radicals undergo oxidation reactions upon exposure to air, releasing large amounts of heat. This induces further decomposition and fission of the coal molecules, and more free radicals are formed, eventually leading to continuous oxidation of coal until spontaneous combustion occurs. The concentration of free radicals in coal is closely related to the

risk of spontaneous combustion, and the variables are positively correlated.<sup>38</sup> Therefore, by studying the evolution laws of the  $g$ -factor, line width, and concentration  $N_g$  of the electron paramagnetic resonance (EPR) spectra of spontaneous coal combustion processes with different inhibitors, we can scientifically and accurately evaluate the performance advantages and disadvantages of different inhibitors in preventing spontaneous coal combustion. EPR, also known as electron spin resonance (ESR), is the only direct method for detecting unpaired electrons and enables both qualitative and quantitative analyses of free radicals in samples. Thus, it is the primary method used for studying free radicals in coal samples. In this study, we performed an in-depth analysis of the  $g$ -factor and free-radical concentrations using EPR spectra. Thus, we investigated the dynamic changes in the concentrations of the free radicals in raw and inhibited coal samples during spontaneous combustion. This approach was used to further explore the effectiveness of silica gel foam in suppressing the spontaneous combustion of residual coal.

Table 2 and Figure 9 show the  $g$ -factor values and changing trends of the different coal spontaneous combustion inhibition

**Table 2.**  $g$ -Factors during Spontaneous Combustion of Raw Coal and Resistant Coal Samples

temperature (°C)	different styles of free-radical $g$ -factor values		
	raw coal	raw coal + silica	raw coal + silica gel foam
50	2.003157	2.002838	2.003090
70	2.003180	2.002760	2.003043
90	2.003128	2.002833	2.003137
110	2.003183	2.002841	2.003070
130	2.003352	2.002966	2.003256
150	2.003324	2.002992	2.003280
170	2.003424	2.003149	2.003397
190	2.003548	2.003204	2.003518
210	2.003470	2.003326	2.003584
230	2.003625	2.003386	2.003630
250	2.003686	2.003328	2.003572



**Figure 9.** Changes in  $g$ -factors during the spontaneous combustion process of different resistant coal samples.

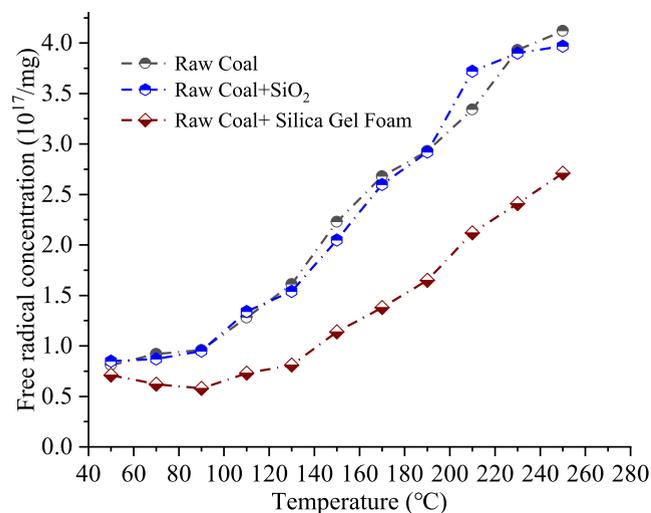
processes, respectively. The  $g$ -factor values of the EPR spectra of coal containing different inhibitors display a trend of first decreasing and then gradually increasing with increasing

temperature. The decrease followed by the increase in the  $g$ -factor proves that new free radicals are generated during the spontaneous combustion of coal.<sup>39</sup> Because of the single-variable principle, high-purity  $\text{SiO}_2$ , which does not participate in chemical reactions, was added to the control group coal and made the  $g$ -factor value of the original coal + silica dioxide lower than those of the original coal and other coal samples (containing mixed inhibition materials). Owing to the different substances added to the coal, the  $g$ -factors of the samples containing different additives changed, albeit the trend in the change was dissimilar. The  $g$ -factor of free radicals is generally related to the surrounding environment of paired electrons, which can be used to determine the type and complexity of free radicals. With an increase in temperature, the orbital–spin coupling effect in coal is enhanced, and the addition of inhibitors causes an increase in the number of hybrid atom free radicals, which promotes an increase in the  $g$ -factor of coal.

**3.3.3. Change Rules of Free-Radical Concentration.** The free-radical concentrations of the original coal control group and samples containing different inhibitors (added during the spontaneous combustion process) oxidized to different temperatures were calculated, as shown in Table 3, and the data are plotted in Figure 10.

**Table 3. Trend in the Evolution of Free-Radical Concentration**

temperature ( $^{\circ}\text{C}$ )	free-radical concentration of different samples ( $10^{17}/\text{mg}$ )		
	raw coal	raw coal + silica	raw coal + silica gel foam
50	0.81	0.85	0.71
70	0.92	0.87	0.62
90	0.96	0.95	0.58
110	1.28	1.34	0.73
130	1.61	1.54	0.81
150	2.23	2.05	1.14
170	2.68	2.60	1.38
190	2.93	2.92	1.65
210	3.34	3.72	2.12
230	3.83	3.90	2.41
250	4.12	3.97	2.71



**Figure 10.** Chart illustrating the trend in the change of the evolution of free radicals.

As shown in the figure, the free-radical concentrations of raw coal and coal samples containing inhibitors first increased slowly and then rapidly during the spontaneous combustion process, regardless of the type of inhibitor added. The initial free-radical concentrations of the coal samples containing silica gel foam were lower than those of the raw coal and coal samples containing inhibitory materials. The free-radical concentrations of the four different coal samples changed relatively steadily before the temperature reached  $100\text{ }^{\circ}\text{C}$ , and that of the coal samples containing silica gel foam exhibited a downward trend.

After the temperature reached  $140\text{ }^{\circ}\text{C}$ , the coal–oxygen complex reaction reached a turning point, and the reaction process rapidly increased, causing the free-radical concentration in the coal to increase continuously. The free-radical concentration of the coal samples containing silica displayed almost the same increasing trend as that of the raw coal samples. Even at higher temperatures, the free-radical concentration was higher, and the inhibitory effect was lost. The free-radical concentration of the coal samples containing silica inhibitors changed relatively slowly before the temperature rose to  $160\text{ }^{\circ}\text{C}$ , and the rate of the subsequent rising trend was lower than those of the raw coal and the samples containing silica but higher than that of the samples containing inhibiting materials. Compared with that of the sample containing silica inhibitor, the free-radical concentration of the coal samples containing silica gel foam displayed a notably decreasing trend in the oxidation temperature range between  $\sim 60$  and  $100\text{ }^{\circ}\text{C}$  before the slow increase, thereby indicating that the silica gel foam had a stronger inhibitory effect on the generation of free radicals at this stage, and the inhibitory effect was markedly better than those of other inhibitors.

#### 4. CONCLUSIONS

This paper introduces the composition optimization and preparation process of silica gel foam and analyzes the selection and function of its components; moreover, in the study, the principles and methods of the modified antioxidant system were investigated. Macroscopic performance tests, microscopic structural characterization, and studies on the chemical inhibitory mechanism of this composite material have allowed us to conclude the following.

Silica gel foam provided specific air leakage resistance under different leakage pressures. The air leakage resistance of the raw coal sample containing 2 wt % silica gel foam increased from 0 to 2.0 kPa. Spatially, three-dimensional coverage was accomplished in the high-permeability areas of the fissures in the goaf, owing to the high fluidity and low-concentration characteristics of the silica gel foam. The surface-wetting properties and foam form of silica gel foam enable it to adhere to coal surfaces. The jamming effect, which blocks the pores on the coal surface, significantly reduces the surface area of coal exposed to air, thereby eliminating the contact channels for reactions with oxygen and reducing the possibility of spontaneous combustion in mines.

The  $g$ -factor results from the EPR spectra of coal containing different inhibitors first decrease and then increase with rising temperature, indicating that new free radicals are generated during coal self-ignition. Despite the addition of various inhibitory materials, the  $g$ -factor in the coal samples still exhibited a decreasing and then increasing trend. Observing the trends of the  $g$ -factor helps determine the dynamic nature of free-radical formation during coal self-ignition, providing

information for devising more effective mine fire prevention strategies.

Regardless of whether it was the original coal or the coal samples containing inhibitors, the concentration of free radicals (Ng) during self-ignition first slowly increased and then rapidly increased. When the temperature increases to 160 °C, the concentration of free radicals in the inhibited coal samples containing added silica dioxide is similar to that of the original coal samples, indicating that the inhibited coal samples lost their inhibitory effect. However, in the inhibited coal samples containing silica dioxide, the concentration of free radicals during self-ignition shows a clear decreasing trend in the oxidation temperature range between 60 and 100 °C. This result indicates that during this phase, the inhibitory effect of silica gel foam on free-radical generation was stronger, and its inhibitory effect was significantly superior to those of other inhibitors. This approach offers an efficient and cost-effective solution for managing spontaneous coal combustion disasters in mines.

## 5. FUTURE OUTLOOK

Based on the fundamental theory of spontaneous coal combustion, this study used a combination of fluid dynamics and materials science to study the rheological characteristics, microstructure, and chemical inhibitory mechanisms of silica gel foam. However, owing to the complex natural oxidation process of coal, the variety of coal types, and the limitations posed by the current experimental conditions, time, and theoretical levels, further refinement is needed in future research, which includes the following.

An important future direction is the evaluation of silica gel foam in actual coal mine environments instead of in a laboratory environment, which should include field trials under various geological and climatic conditions to validate the practical applicability of the laboratory results. Furthermore, studies should ensure that the efficacy of silica gel foam in inhibiting coal self-ignition is not affected by environmental factors.

We conducted trials on the inhibitory effect of silica gel foam on the spontaneous combustion of different types of coal, further refining the material composition optimization and extending its application in different mines. We aimed to test the performance of the material under actual mine conditions, thereby optimizing its formula to enhance its fire prevention and extinguishing capabilities and ensure mine production safety.

Future research will also focus on the environmental impacts of using silica gel foam, particularly its long-term stability and ecofriendliness. We are committed to ensuring that the proposed solutions effectively prevent spontaneous coal combustion without negatively affecting the mining environment, thereby ensuring sustainability.

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All authors have given approval to the final version of the manuscript.

## Notes

The authors declare no competing financial interest.

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## ABBREVIATIONS

ERP, electron paramagnetic resonance; KGM, konjac glucomannan; FA, fly ash

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