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Effect of Coal Dust Content on the Low-temperature Oxidation of Silo Coal

Jianguo Liu,* Zihao Zhou, Longzhe Jin, Tianyang Wang, Shengnan Ou, Shu Wang, Yixuan Wei, and Mulati Jueraiti

Cite This: *ACS Omega* 2022, 7, 37442−37451

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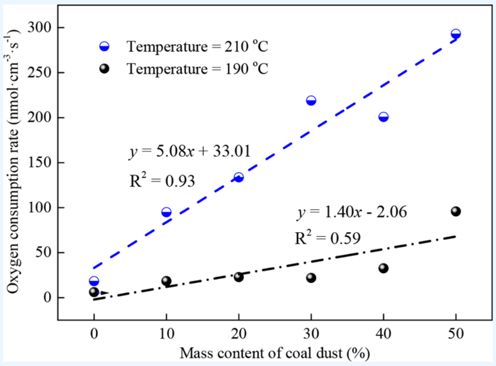
ABSTRACT: Coal's low-temperature oxidation (LTO) poses a significant threat to the safety of storing coal in silos. This study investigates the impact of coal dust content on the LTO characteristics of silo coal samples. The results indicate that the larger the coal dust content the higher the oxygen (O_2) consumption rate and carbon monoxide (CO) generation rate and the stronger the LTO capacity. To clear the mechanism of the impact, the thermal physical characteristics were studied and thermogravimetric and differential thermal analysis (TG-DTA) experiments were performed on various coal samples. The results show that, first, with the increase of coal dust content, the thermal conductivity of the silo coal samples initially increased and then decreased, whereas the thermal diffusion and heat capacity decreased and increased linearly, respectively. This indicates that the heat storage capacity of the silo coal sample is enhanced with the increase of the coal dust content. Second, the maximum oxygen absorption rate and differential thermal reduction value of the coal samples increased linearly with the decrease in their particle size; this result verifies that decreasing the particle size of silo coal can advance its LTO process. The study findings indicate that the risk of LTO and spontaneous combustion of silo coal can be effectively reduced by controlling the coal dust content (fine coal particles).

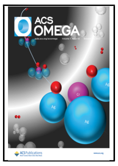


1. INTRODUCTION

Uncertainties in terms of epidemics, wars, and antiglobalization trades have increased in recent decades, leading to extensive investigations on energy security worldwide.[1−3] As a conventional fossil fuel, coal is expected to remain the primary source of energy in China in the coming decades.[4−6] To ensure energy security, the Chinese government has proposed increasing the deployable coal storage from 100 to 200 million tons.[7] For coal storage, silos are widely used in power plants, ports, and other places owing to the advantages of environmental protection, high space utilization rate, and convenient transportation.[8−10] However, the closed environment of silos results in a heat storage condition that could lead to the low-temperature oxidation (LTO) of coal, which increases the risk of spontaneous combustion of silo coal.[11−13] Therefore, the characteristics of the LTO and spontaneous combustion of silo coal should be investigated for ensuring the safety of silos.

Researchers have extensively studied the LTO and spontaneous combustion of coal dust.[14−16] Zou et al.[17] investigated the impact of oxygen (O_2) concentration on the LTO of coal dust at low temperatures; they reported that the minimum ignition temperature of coal dust can increase by 50 °C when the oxygen concentration is reduced from 21 to 5%. Wu et al.[18] analyzed the effect of O_2 and carbon dioxide (CO_2) on the spontaneous combustion of coal dust; their results indicated that the inhibitory effect of reducing the O_2 concentration on the spontaneous combustion of coal dust is significantly stronger than that of increasing the CO_2 concentration. A similar conclusion was reported in another study carried out based on numerical simulations.[19] Li et al.[20] investigated the impact of the particle size of coal dust on its spontaneous combustion, wherein they determined that the decrease in the particle size of coal dust could facilitate the decrease in its minimum ignition temperature. Similarly, the studies conducted by Benedetto et al.[21,22] also show that the particle size of dust can significantly affect its flammability and explosion behaviors. Ren et al.[23] considered the exothermal property of the LTO of coal dust and researched the variations in industrial components and thermal physical characteristics of coal dust under 300 °C using a laser-flash apparatus. They found that the specific heat capacity of coal dust is essential in determining its thermal conductivity. Another study[24] explored the exothermal characteristics of the LTO of coal dust using

Received: July 5, 2022
Accepted: October 6, 2022
Published: October 13, 2022



the thermogravimetric and differential scanning calorimetry (TG-DSC) apparatus; the results indicate that the heat flow of coal dust initially decreases, then increases, and finally decreases again with the increase in temperature. Additionally, Ramírez et al.[25] reported that the thermal susceptibility analysis performed using TG-DSC is better than thermal stability analysis when analyzing the LTO process of agriculture dust in terms of their spontaneous combustion in silos. Zhang et al.[9] investigated the distribution of high-temperature regions in large coal silos using experimental tests and numerical simulations; the study revealed target areas for preventing the spontaneous combustion of coal in silos.

The aforementioned studies almost all focused on LTO and spontaneous combustion of the coal dust with fine particle size; however, both large and fine particles of coal exist in silos. In other words, coal blocks with large particle sizes are mixed with a certain proportion of coal dust with fine particle sizes in silos. That is, the published research results of the LTO and spontaneous combustion of pure coal dust cannot really reflect the low-temperature oxidation of silo coal. Therefore, there is a research gap to conduct a comprehensive study for understanding the influence of coal dust content on the LTO and spontaneous combustion of silo coal. The study for the research gap can distinctly guide the particle size distribution of silo coal for preventing its combustion.

To this end, in the present study, first, we investigated the effect of coal dust content on the LTO and spontaneous combustion of silo coal using programmed heating experiments; then, to reveal the effect mechanism of coal dust content on the LTO process of silo coal, the thermal physical parameters, including thermal conductivity, thermal diffusivity, and heat capacity, of the silo coal samples with various coal dust content were measured; after that, the decomposition process of silo coal with different particle sizes was measured at low temperature using thermogravimetric and differential thermal analysis (TG-DTA) experiments. Based on these tests, it was discussed that the impact mechanism of the coal dust content on the LTO and spontaneous combustion of silo coal. The findings of this study are significant for preventing the spontaneous combustion of silo coal and ensuring the safety of coal silos.

2. MATERIAL AND METHODS

2.1. Preparation of Coal Samples. The experimental coal was sampled from a silo in Qinglong Temple Coal Mine, Yulin City, Shaanxi Province, China. Table 1 summarizes the industrial analysis of the coal sample conducted according to GB/T212-2008. The coal sample was crushed and screened using a ball mill device, and the LTO and decomposition samples were prepared subsequently.

Table 1. Industrial Analysis of the Silo Coal Sample (Mass Percentage (%))

M_{ad}[a]	A_{ad}[b]	V_{daf}[c]	FC_{ad}[d]
6.0	15.46	39.00	45.7

[a] M_{ad} = moisture under air-dried basis. [b] A_d = ash under air-dried basis. [c] V_{daf} = volatile matter under dried ash-free basis. [d] FC_{ad} = fixed carbon under air-dried basis.

2.1.1. Preparation of LTO Samples. The LTO samples were composed of lump coal and coal dust mixed with a certain mass fraction. The lump coal comprised four types of coal particles with particle sizes of 0.074−0.9 mm (20−200 mesh), 0.9−2 mm (9−20 mesh), 2−5 mm, and 5−10 mm at 25% mass fraction each. The particle size of the coal dust sample was less than 74 μm (>200 mesh). As shown in Table 2, six LTO samples were prepared by adding the coal dust sample to the lump coal sample with various mass fractions to investigate the effect of coal dust content on the LTO of silo coal.

Table 2. Mixing Principle of the Low-Temperature Oxidation (LTO) Samples for LTO Experiments

	mass fraction (%)					
sample	LTO-1	LTO-2	LTO-3	LTO-4	LTO-5	LTO-6
lump coal	100	90	80	70	60	50
coal dust	0	10	20	30	40	50

2.1.2. Preparation of Decomposition Samples. Four decomposition samples with particle sizes of 0.150−0.250 mm (60−100 mesh), 0.075−0.150 mm (100−200 mesh), 0.075−0.150 mm (200−300 mesh), and less than 0.050 mm (>300 mesh) were prepared by crushing and screening to investigate the impact of particle size on the LTO of silo coal; the samples were subjected to TG-DTA experiments.

2.2. Temperature-Programmed Experiments and Data Analysis Methods. *2.2.1. Experimental Apparatus and Method.* Figure 1 depicts the temperature-programmed apparatus used for the LTO samples. The apparatus primarily comprises a gas source, flow valve, heating furnace, cooling pool, gas collection bag, and gas chromatography. As reported in previous studies,[26−28] 100 g of LTO sample was placed in a cylindrical closed heating tank with a diameter of 8 cm. Compressed air was ventilated into the heating tank at a gas flow rate of 100 mL/min. The heating rate was 1 °C/min, and the initial and target temperatures were 30 and 210 °C, respectively. The gas generated during the heating process was collected at intervals of 20 °C, and the concentrations of O_2 and CO in the generated gas were determined using a gas chromatograph (Agilent 7890B).

2.2.2. Data Analysis Methods. According to ref 29, we considered O_2 and CO as the index gases of the LTO process. The O_2 consumption rate (OCR) and CO generation rate (CGR) were calculated at different temperatures using eqs 1 and 2, respectively.[26,27]

$$V_{O_2}(T) = \frac{Q\varphi_{O_2}^{1}}{SL}\ln\frac{\varphi_{O_2}^{1}}{\varphi_{O_2}^{2}} \tag{1}$$

$$V_{CO}(T) = \frac{(\varphi_{CO}^{2} - \varphi_{CO}^{2})V_{O_2}(T)}{\varphi_{O_2}^{1}\left[1 - \exp\frac{-SLV_{O_2}(T)}{Q\varphi_{O_2}^{1}}\right]} \tag{2}$$

where $V_{O_2}(T)$ denotes the OCR at temperature T (mol/(cm^3·s)); Q indicates the air supply rate (mL/min); S represents the sectional area of the heating tank (cm^2); L denotes the height of the coal sample (cm); $\varphi_{O_2}^{1}$ and $\varphi_{O_2}^{1}$ indicate the O_2 concentrations at the inlet and outlet of the heating tank, respectively (%); $V_{CO}(T)$ denotes the CGR at temperature T (mol/(cm^3·s)); and φ_{CO}^{1} and φ_{CO}^{2} indicate the CO concentrations at the inlet and outlet of the heating tank, respectively, (%).

https://doi.org/10.1021/acsomega.2c04219
ACS Omega 2022, 7, 37442−37451

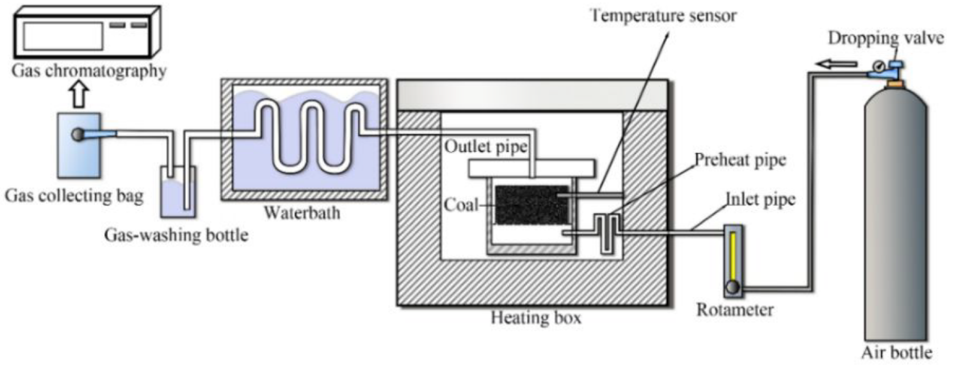


Figure 1. Temperature-programmed apparatus of coal samples.

2.3. Test of the Thermal Physical Parameters.

The varying regulations of the bulk density and thermal physical parameters of the LTO samples were measured to explore its effect mechanism on the LTO of silo coal.

2.3.1. Bulk Density Test. The bulk density of coal particles significantly impacts the thermal conductivity of coal.[30] Therefore, the bulk density of the LTO samples was measured using a volumetric cylinder and an electronic analytic balance according to eq 3

$$\rho = m/V \tag{3}$$

where ρ denotes the bulk density (g/cm^3) and m and V indicate the mass (g) and volume (mL) of the sample, respectively. The masses of all test samples in this study were 10 g.

2.3.2. Thermal Physical Parameter Test. The thermal conductivity of silo coal determines its heat storage capacity, which largely affects its spontaneous combustion characteristics.[14] Figure 2 illustrates the measurement instrument for testing the thermal physical parameters of the LTO samples. The thermal physical parameters include thermal conductivity (λ), thermal diffusion (α), and thermal capacity (C), measured via the hot-wire method using the instrument of XIATECH-TC3000 according to ref 31. Specifically, during the measurement, 50 cm^3 of the LTO sample was placed in the sample box (Figure 2b), and the temperature sensor (Figure 2a) was buried in the sample box. The thermal physical parameters were automatically calculated using the software attached to the instrument.

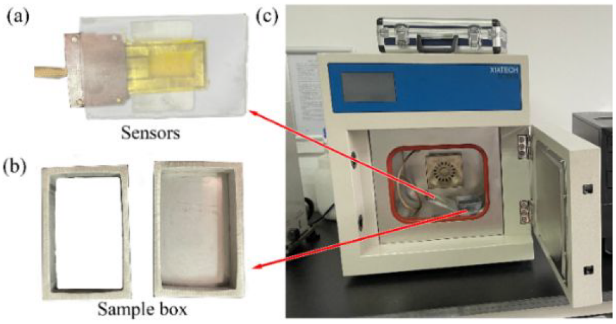


Figure 2. Measurement instrument of thermal physical parameters: (a) temperature sensor, (b) sample box, and (c) measurement apparatus.

2.4. TG-DTA Test and Data Analysis Method.

2.4.1. TG-DTA test. TG-DTA is a common method used to study the LTO characteristics of coal dust.[31] To explore the effect of particle size on the LTO of silo coal, TG-DTA tests were performed on four decomposition samples under an air atmosphere at a flow rate of 100 mL/min. The mass of the measurement sample was 2 mg, and the temperature was increased from 25 to 300 °C at a heating rate of 1 °C/min.

2.4.2. Data Analysis Method. Figure 3a depicts the typical TG and derivative thermogravimetric (DTG) curves of the coal dust samples. As indicated in the figure, the coal sample experienced three stages under low temperatures (25−300 °C); stage 1 involved the weight loss of the sample caused by water evaporation, and stage 2 exhibited the weight gain of the sample caused by oxygen absorption, and stage 3 involved the weight loss again caused by the decomposition of samples.[32] To quantitatively evaluate the influence of particle size on the LTO characteristics of the coal samples, the percentage of oxygen absorption (Δ_m (%)) and maximum oxygen absorption rate (V_{max} (%/min)) were extracted as the characteristic parameters of the LTO process (Figure 3a).

Figure 3b depicts the typical DTA curve of the coal sample. It was observed that when the temperature was higher than 100 °C, the rate of heat absorption gradually reduced under the oxidative exothermic action of the coal sample;[24] the DTA curve reached a minimum value at the temperature of approximately 200 °C. Subsequently, the DTA curve began to increase and a maximum value was attained at the temperature of approximately 260 °C. After that, the value of the DTA curve rapidly decreased in the final stage owing to the heat adsorption process caused by the decomposition of the coal sample. In this study, the reduction value (Δ_h, μV) of the DTA between temperatures 200 and 260 °C was extracted as an index parameter to characterize the oxidation strength of the coal samples. Figure 3b depicts the extraction method.

3. RESULTS AND DISCUSSION

In this study, to investigate the impact of coal dust content on the LTO of silo coal, first, the LTO parameters of the LTO samples with different coal dust contents were measured using a temperature-programmed apparatus, and the OCR and CGR at various temperatures were computed based on the measurement results. The experimental results indicate that the LTO parameters of silo coal samples regularity changed with an increase of coal dust content. Then, to ascertain the effect mechanism of coal dust content on the LTO characteristics of silo coal, the impacts of the coal dust content on the bulk density and thermal conductivity parameters of the LTO samples were tested. At the same time, the effect of

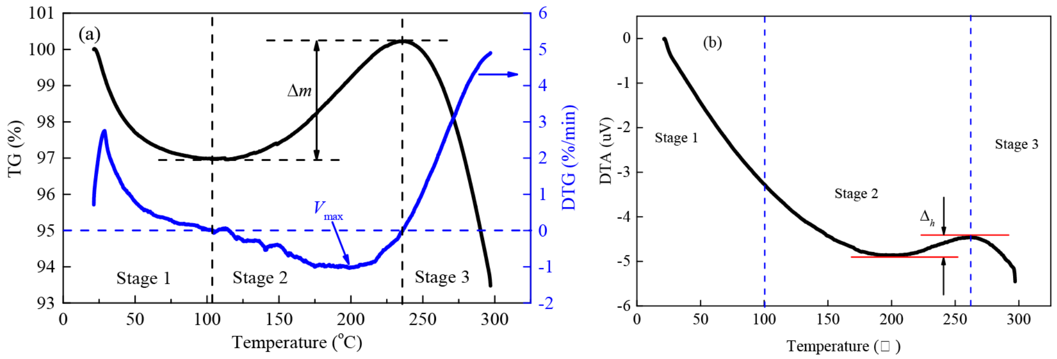


Figure 3. Typical curves of the thermogravimetric and derivative thermogravimetric (TG-DTG) (a) and differential thermal analysis (DTA) (b) of coal at low temperatures.

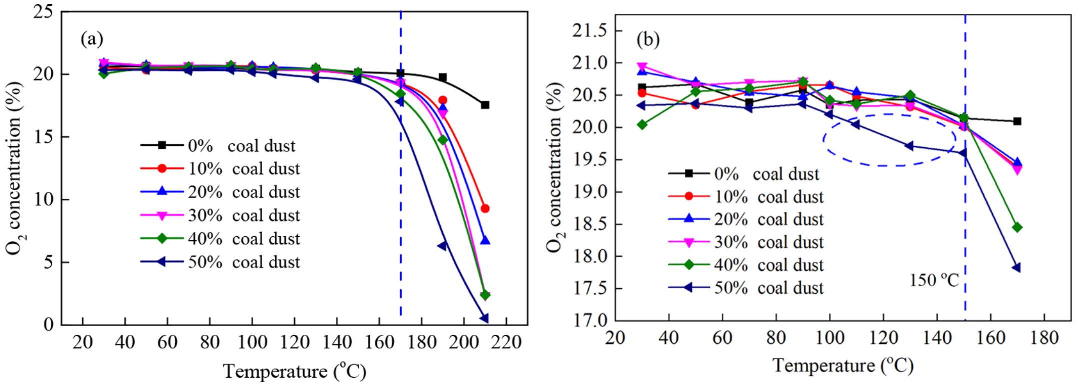


Figure 4. O_2 concentrations of the low-temperature oxidation (LTO) samples when temperatures less than 210 °C (a) and 170 °C (b).

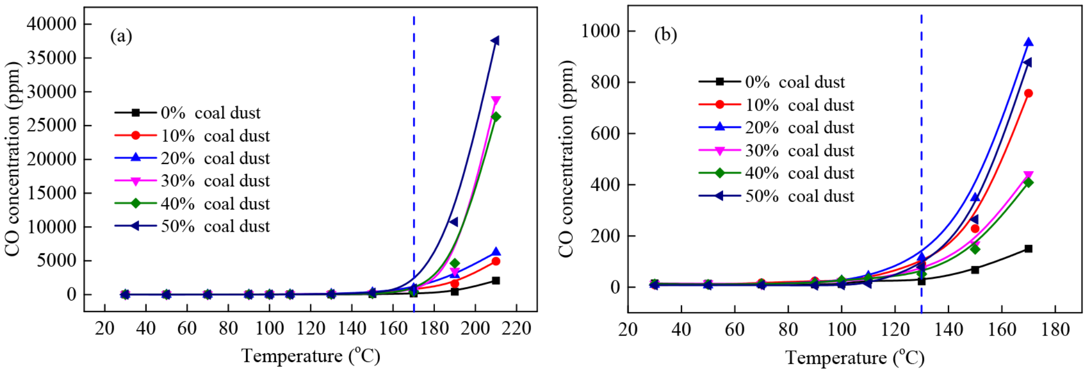


Figure 5. CO concentrations of the low-temperature oxidation (LTO) samples when temperatures are less than 210 °C (a) and 170 °C (b).

particle size of coal dust on its LTO process was explored using TG-DTA experiments. The results and corresponding analysis are explained in subsequent sections.

3.1. Effect of Coal Dust Content on the LTO Process of Silo Coal. As the coal sample can adsorb O_2 and generate CO during the oxidation process, they are commonly used as index gases for analyzing the LTO process of coal.[29] The adsorption of O_2 includes physical and chemical adsorptions, and CO is generated by the oxidation reaction of coal.[29] Figure 4 depicts the O_2 concentration variation curve of the LTO samples with different coal dust contents. As indicated in Figure 4a, the O_2 concentration varied significantly above 170 °C in six LTO samples, and the larger the coal dust content, the lower the O_2 concentration. At 210 °C, the O_2 concentration of the LTO sample containing 50% coal dust was only 0.53%, which was 3% of the O_2 concentration (17.54%) of the LTO sample without the coal dust. This implies that the coal dust content significantly promotes the LTO process of silo coal. Furthermore, as indicated in Figure 4b, the O_2 concentration of the LTO sample containing 50% coal dust reduced gradually after 100 °C and rapidly after 150 °C. This result implies that the addition of coal dust can significantly enhance the physicochemical O_2 adsorption of coal samples.

Figure 5 shows the CO concentrations of the samples in the LTO process. As depicted in Figure 5a, the CO concentrations of the LTO samples exponentially increased after 170 °C; namely, the higher the coal dust content, the higher the CO concentration. At 210 °C, the CO concentration of the LTO sample containing 50% coal dust was 37 565 ppm, which was 18.2 times that of the LTO sample without the coal dust. As indicated in Figure 5b, the CO concentrations of the LTO samples changed at 130 °C, wherein the samples began to oxidate. To quantitatively analyze the influence law of coal dust content on the LTO characteristics of silo coal, the OCR and CGR of each LTO sample were calculated and analyzed.

3.1.1. Effect of Coal Dust Content on the OCRs of the LTO Samples. Figure 6 depicts the OCRs of the LTO samples.

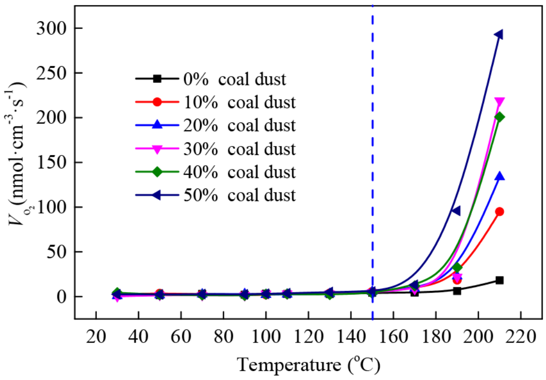


Figure 6. O_2 consumption rates of the low-temperature oxidation (LTO) samples concerning temperature.

Similar to the variation of the O_2 concentration, the OCRs exponentially increased with temperature after 150 °C, and the OCRs increased with the increase in the coal dust content at 210 °C. It was observed that the OCR of the LTO sample containing 50% coal dust was 16.1 times that of the sample containing no coal dust. The results indicate that the coal dust content could significantly promote the LTO process of silo coal.

Figure 7 shows the variation of OCRs concerning the coal dust content. As indicated in Figure 7a, no significant difference was observed in the OCRs of the six samples at 150 °C. But when the temperature was increased to 170 °C, the OCRs of the samples linearly increased with the increase in the coal dust content and exhibited a goodness-of-fit (R^2) of 0.73. After that, with the further increase in the temperature, the OCRs of the samples also linearly increased with a larger increase rate (the slope of the fitting linear); specifically, the values of the slope increased from 0.14 °C/min at 170 to 5.08 °C/min at 210 °C. These results indicate that the coal dust content can significantly improve the LTO process of the silo coal samples. In other words, the higher the coal dust content, the stronger the oxygen adsorption capacity, which results in a higher OCR at the same temperature.

3.1.2. Effect of Coal Dust Content on the CGRs of the LTO Samples. Figure 8 depicts the variation curves of the CGRs of the LTO samples. It was observed that the CGRs of the six samples were initially similar to each other with low values when the temperature was below 170 °C, implying that the oxidation reaction of the samples is weak in this temperature range. Conversely, as depicted in Figure 7, the OCRs of the LTO samples linearly increased with the coal dust content at 170 °C. That is, in comparison with lump coal, coal dust exhibits a smaller particle size, larger specific surface area, and a more developed pore structure. These characteristics increase the number of places for oxygen adsorption on the surface of coal dust more than that on the lump coal surface. This enables the samples containing more coal dust content to exhibit stronger oxygen adsorption effects. In addition, it is known that the oxygen adsorbed on the sample surface is not completely involved in the oxidation reaction below 170 °C. Consequently, at 170 °C, the OCRs of the LTO samples differ significantly with coal dust content (Figure 8), whereas the CGRs remain similar (Figure 7a). After that, when the temperature is above 170 °C, the LTO samples begin to distinctly generate CO (Figure 8), and the CGRs rapidly increase with the increase of temperature; this implies that the oxidation reaction of the samples is enhanced. Simultaneously,

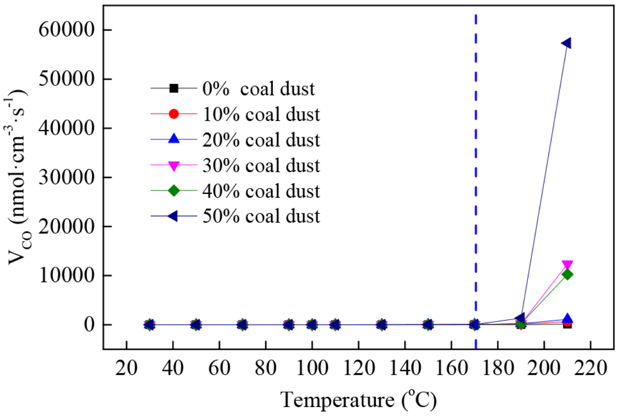


Figure 8. CO generation rates of the low-temperature oxidation (LTO) samples concerning temperature.

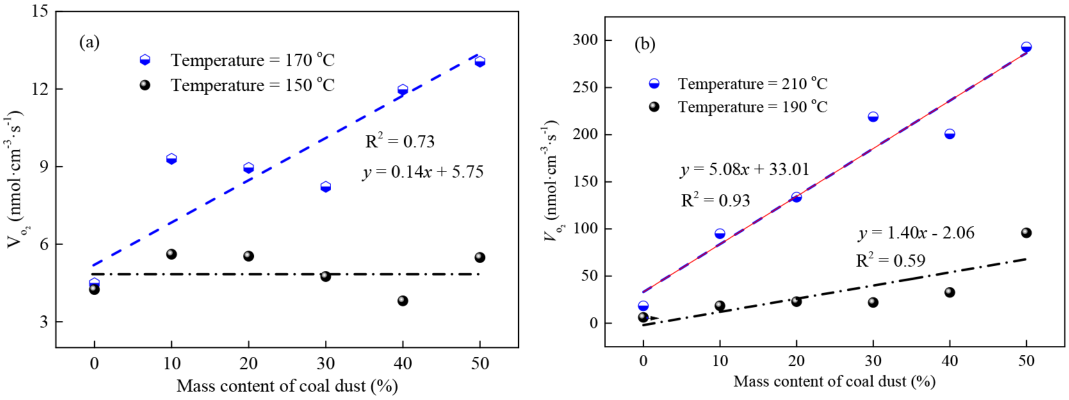


Figure 7. O_2 consumption rates of the low-temperature oxidation (LTO) samples under temperatures of 150 and 170 °C (a), 190 °C, and 210 °C (b).

the CGRs of the LTO samples increase substantially with the increase of the coal dust content. In particular, the CGR of the LTO sample containing 50% coal dust content is 479.4 times that of the LTO sample without the coal dust at 210 °C. This further validates that the coal dust content can obviously enhance the oxidation reaction of silo coal.

Figure 9 depicts the variation curves of the CGRs concerning the coal dust content. As indicated in the figure, no significant difference was observed in the CGRs of the six LTO samples at 170 °C with an average value of 32.6 nmol/(cm^3·s). As the temperature increased, the CGRs of the samples exponentially increased with the coal dust content. At 190 and 210 °C, the CGRs of the LTO sample containing 50% coal dust content were 64.0 and 479.4 times that of the LTO sample containing no coal dust, respectively. Combining both Figures 7 and 9, we can find that the LTO samples containing coal dust adsorbed a substantial amount of oxygen at low temperatures (less than 170 °C), whereas the adsorbed oxygen only can react with the coal sample when the temperature was increased to the oxidation temperature of coal; thus, a large amount of CO was generated.

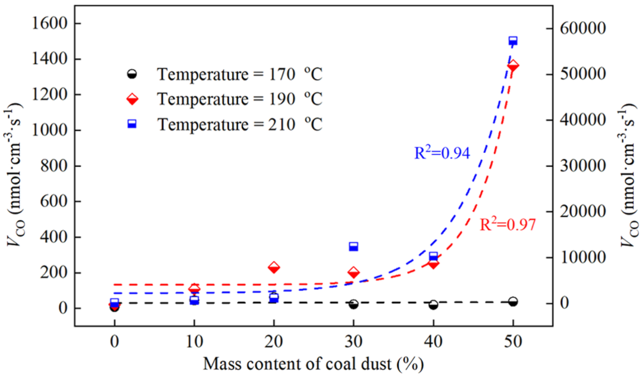


Figure 9. Variation curves of the CO generation rates concerning the coal dust content under different temperatures.

The LTO process of the coal heap is codetermined by the heat storage capacity of the environment (external factor)[23] and the self-oxidation characteristics of coal (internal factor).[32] Based on this conclusion, the effect of coal dust content on the thermal conductivity parameters of the LTO samples was discussed in section 3.2, and section 3.3 explains the impact of the particle size of coal on its self-oxidation parameters.

3.2. Effect of Coal Dust Content on the Thermal Physical Characteristics and the Bulk Density of the LTO Samples. To analyze the effect mechanism of the coal dust content on the LTO process of silo coal, we investigated the relationships between the coal dust content and thermal physical characteristics of the LTO samples. These characteristics include thermal conductivity, thermal diffusion, and thermal capacity. Additionally, the variation in the bulk density of the LTO samples was investigated concerning the coal dust content.

3.2.1. Effect of Coal Dust Content on the Thermal Physical Parameters of the LTO Samples. Figure 10 depicts the thermal physical parameters of the LTO samples. As indicated in Figure 10a, the thermal conductivity of the samples initially increased and then decreased with the increase of the coal dust content; in particular, when the content of coal dust increased from 0 to 20%, the thermal conductivity of the LTO samples increased by 19.1%. With further increase in the coal dust content, the thermal conductivity of the LTO samples gradually decreased, and the thermal conductivity of pure coal dust (100% coal dust) reduced to 0.0714 W/(m·K), which is lower than that of the LTO sample without the coal dust (0.0758 W/(m·K)).

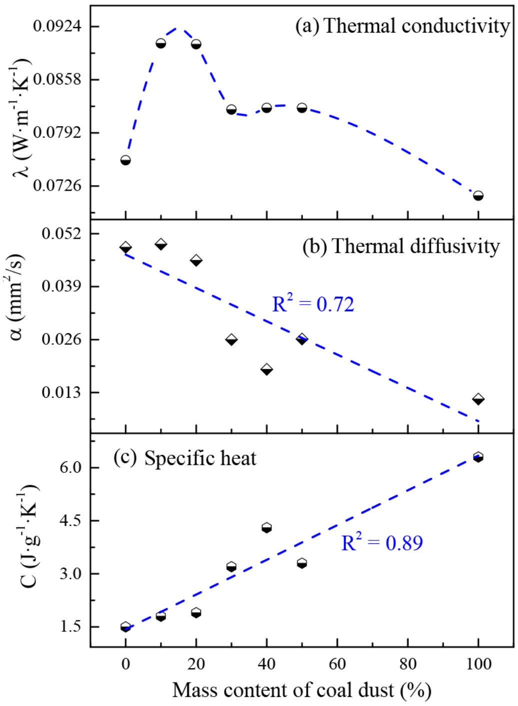


Figure 10. Thermal conductivity (a), thermal diffusion (b), and thermal capacity (c) of the low-temperature oxidation (LTO) samples.

When the content of coal dust is less than 20% (Figure 11), the addition of coal dust can effectively fill the gaps in the lump coal. As the thermal conductivity of the coal dust is higher than that of air, the thermal conductivity of the LTO samples increases in this stage with the increase in the coal dust content. When the coal dust content increases by more than 20%, the gaps in the lump coal are filled, and the relative content of the lump coal decreases. As the thermal conductivity of the coal dust is lower than that of the lump coal, the thermal conductivity of the LTO samples begins to decrease when the coal dust content increases beyond 20%.

Thermal diffusion is a common parameter used for evaluating the heat storage capacity of objects. The larger the thermal diffusion, the poorer the heat storage capacity.[23,33] Figure 10b depicts the variation curve of the thermal diffusion of the LTO samples with respect to the coal dust content. As indicated in the figure, the thermal diffusion linearly decreased with the increase in the coal dust content with an R^2 of 0.72. This result indicates that the addition of coal dust can effectively reduce the thermal diffusion of the samples and improve their heat storage capacity, facilitating the LTO process of the coal samples. By contrast, Figure 10c indicates that the thermal capacity of the LTO samples linearly increases with the increase in coal dust content with an R^2 = 0.89. The thermal capacity of pure coal dust was 4.2 times that of the LTO samples containing no coal dust; this result concurs with the finding reported in a previous study.[23] Therefore, the LTO samples containing coal dust exhibit a

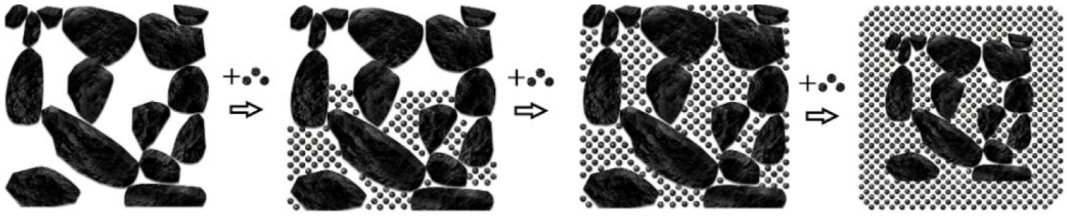


Figure 11. Filling diagram of the coal dust in lump coal.

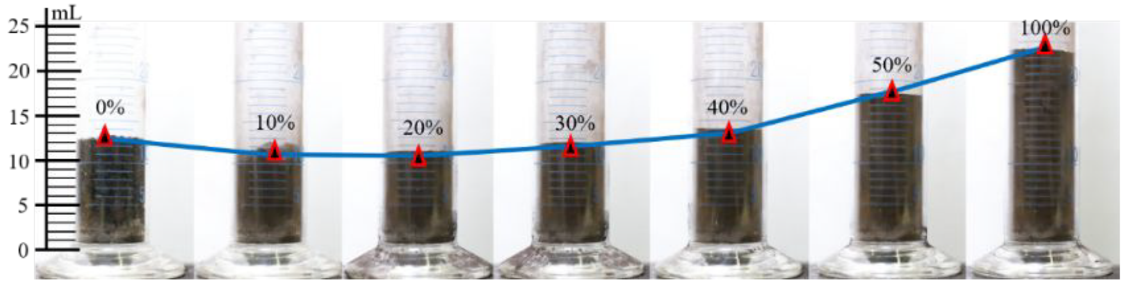


Figure 12. Variation in the stacking volumes of the low-temperature oxidation (LTO) samples concerning the coal dust content.

stronger heat absorption capacity. In other words, more energy can be absorbed in silo coal with more coal dust content by increasing the unit temperature, which is the primary reason for the reduction of thermal diffusion in silo coal when adding coal dust to it. In conclusion, the LTO process of silo coal could be improved by increasing the thermal capacity when adding coal dust content.

3.2.2. Effect of Coal Dust Content on the Bulk Density of the LTO Samples. To further understand the effect mechanism of the coal dust content on the thermal physical parameters of the LTO samples, we investigated the variation in the bulk density of the LTO samples with different coal dust content. Figure 12 illustrates the stacking volumes of the six LTO samples with identical mass (10 g). It was observed that when less than 20% (mass percentage) of coal dust was added, the stacking volumes of the LTO samples gradually decreased with the increase in the coal dust content. Subsequently, the stacking volume began to increase with a further increase in the coal dust content. The stacking volume of pure coal dust (100% coal dust) was 2.1 times that of pure lump coal.

Figure 13 shows the bulk density of the LTO samples. Herein, we observed that the bulk density initially increased and then decreased with the increase in the coal dust content; it reached a maximum value of 0.85 g/cm^3 at 20% content of the coal dust. This variation is similar to that of the thermal conductivity depicted in Figure 10a, indicating that the coal dust content mainly affects the thermal conductivity of silo coal samples by changing their bulk density.

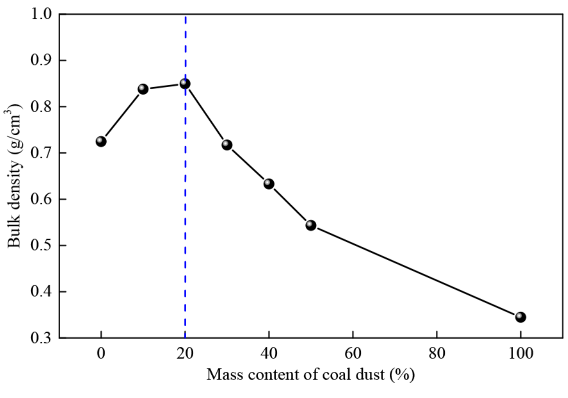


Figure 13. Variation in the bulk density concerning the coal dust content.

In summary, the thermal diffusion of the LTO sample linearly decreases with the increase in the coal dust content. This in turn enhances the heat absorption capacity and reduces the thermal conductivity capacity, which can advance the LTO of silo coal.

3.3. Effect of Particle Size on the Decomposition Process of Coal. To understand the effect mechanism of the coal dust content on the LTO process of silo coal, the impact of the particle size on the LTO process of silo coal was investigated. The TG-DTA experiments were performed using four decomposition samples. Figure 14a depicts the TG curves of the decomposition samples, wherein the mass of the samples experiences three variation stages. The mass initially decreases, then increases, and decreases again with the increase in the temperature; the inflection temperature was approximately between 100 and 236 °C. Wang et al.[32] reported that the three stages of the mass variation correspond to water evaporation, oxygen absorption, and thermal decomposition of coal under low temperatures.

Figure 14b depicts the DTA curves of the decomposition samples, indicating that the differential thermal values of the samples are constantly decreasing when the temperature is less than 200 °C. This result shows that the samples exhibit endothermal reactions in this temperature range, which primarily includes water evaporation and volatilization of small-molecule organic matters. When the temperature reaches close to 200 °C, the differential thermal values begin to increase and attain a maximum value at approximately 260 °C. This indicates that the samples exhibit oxidative exothermic reactions, and the heat released by the oxidation reaction reduces the overall heat absorption value of the samples. After 260 °C, the differential thermal values of the samples decrease again, indicating the samples enter the decomposition stage.

Additionally, the temperature entering the oxidation reaction tends to decrease with the decrease in the particle size of coal samples (Figure 14b). In other words, the coal samples with

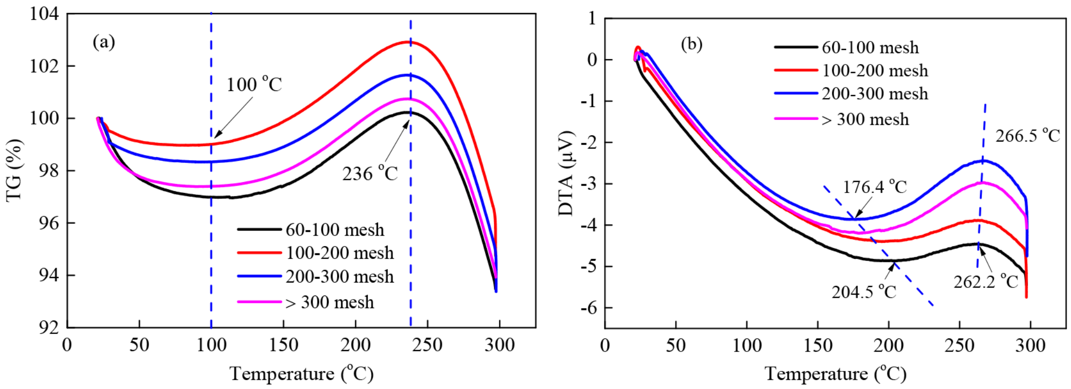


Figure 14. Thermogravimetric (TG) (a) and differential thermal analysis (DTA) (b) curves of the decomposition samples.

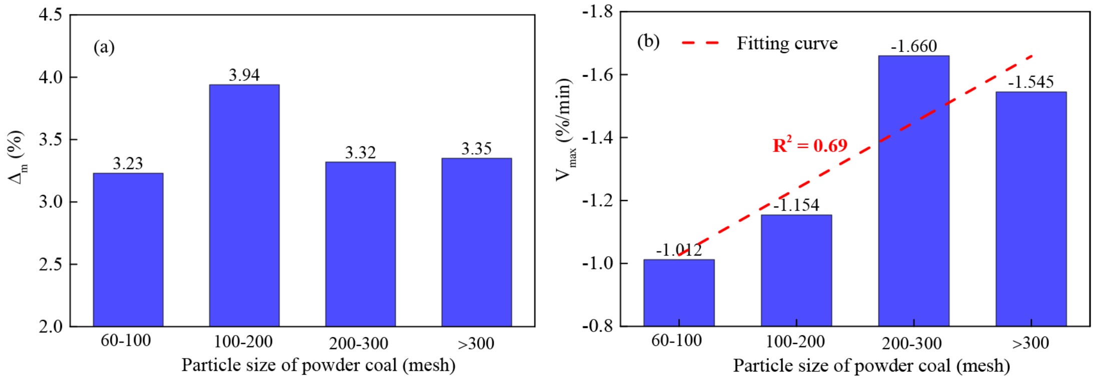


Figure 15. Percentage of oxygen absorption (Δ_m) (a) and the maximum oxygen absorption rate (V_{max}) (b) of the decomposition samples.

smaller particle sizes undergo oxidation reactions at a lower temperature. Similar conclusions were also obtained in previous studies,[34,35] wherein the decrease in the particle size of coal could increase its specific surface area; this causes oxygen to be more easily absorbed when passing through the coal surface, which in turn improves the LTO capacity of coal. To further understand the effect mechanism of the particle size on the LTO process of coal, the characteristic parameters of the TG-DTA experiment were quantitatively analyzed, including the percentage of oxygen absorption (Δ_m (%)), the maximum oxygen absorption rate (V_{max} (%/min)), and the reduction value (Δ_h (μV)) of the DTA.

3.3.1. Effect of the Particle Size on TG Parameters. Figure 15a,b depicts the values of Δ_m and V_{max}, respectively. As indicated in the figures, the value of Δ_m varies slightly with an average of 3.46% following the decrease of the particle size, whereas the value of V_{max} linearly increases with R^2 = 0.69. This implies that at the low-temperature decomposition stage of coal, although the amount of oxygen adsorbed remains unchanged, the oxygen absorption rate increases linearly with the decrease in the particle size of coal. During the LTO process of coal, the temperature is the most critical factor affecting its oxidation; in other words, although large amounts of oxygen are absorbed on the coal surface, only a portion of the oxygen is oxidated with the coal sample.

In summary, the smaller the particle size, the larger the specific surface area and the higher the number of reactive oxidation groups exposed to the surface, which increases the maximum oxygen absorption rate of coal samples.

3.3.2. Effect of Particle Size on the Differential Thermal Parameters. Figure 16 depicts the values of Δ_h of the decomposition samples during the LTO process, wherein the Δ_h values increase linearly (R^2 = 0.59) with the decrease in the particle size. That is, the heat released by the oxidation of the sample increased linearly. This result concurs with the finding shown in Figure 15b. Therefore, the oxidation capacity of the coal sample was significantly enhanced with the decrease in its particle size. The result implies that the particle size of silo coal, particularly the content of the coal dust with smaller particle sizes, should be closely regulated to prevent the spontaneous combustion of silo coal.

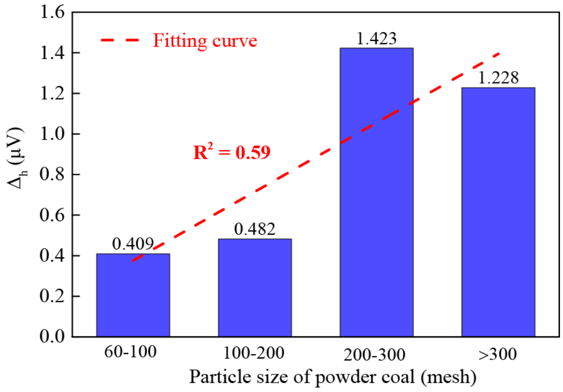


Figure 16. Differential thermal analysis (DTA) reduction value (Δ_h) of the decomposition samples.

3.4. Effect Mechanism of Coal Dust Content on the LTO Process of Silo Coal. The aforementioned experimental results validate that the coal dust content primarily affects the LTO process of silo coal in two aspects, namely the external and internal factors. The thermal diffusion of the samples decreased linearly with the increase in the coal dust content in lump coal samples. Consequently, the heat generated by the LTO of the samples cannot be dissipated in time. This provides an advanced heat storage condition for further oxidation of silo coal,[9,25] thus enhancing its spontaneous combustion process. This is the external factor that facilitates the spontaneous combustion of silo coal at low temperatures.

Additionally, the overall particle size distribution of silo coal decreased after the addition of a certain amount of coal dust. In this scenario, the specific surface area significantly increased with the decrease in the particle size, exposing more reactive oxidation groups to the surface. This advances the LTO process of coal, and the heat released by the LTO of coal provides a heat source for its spontaneous combustion.[16,20] This is the internal factor that advances the spontaneous combustion of silo coal at low temperatures.

In summary, improving the heat storage capacity of silo coal and enhancing the LTO characteristics of silo coal enable the coal dust content to facilitate the spontaneous combustion of silo coal. Therefore, the coal dust content (fine coal particles content) in silo coal should be closely regulated to reduce the possibility of its spontaneous combustion.

4. CONCLUSIONS

In this study, the effect of coal dust content on the LTO of silo coal samples was comprehensively investigated using temperature-programmed apparatus, TG-DTA experiments, and thermal physical parameter tests. The primary conclusions of the study can be summarized as follows.

(1) The coal dust content significantly impacts the LTO of silo coal. With the increase in the coal dust content, the OCR and CGR of the samples increased linearly and exponentially, respectively; at 210 °C, the OCR and the CGR of the coal sample containing 50% coal dust were 16.1 and 479.4 times that of the coal sample without coal dust, respectively. Therefore, controlling the mass fraction of the coal particulars with fine particle size is important to prevent the spontaneous combustion of silo coal.

(2) The coal dust content significantly affects the thermal physical parameters of the silo coal sample. The increase in coal dust content initially increased the thermal conductivity and then decreased the same. Furthermore, the thermal diffusion decreased linearly, whereas the thermal capacity exhibited a linear increase. Enhancing the thermal storage capacity is the external factor that enables the coal dust content to facilitate the spontaneous combustion of silo coal at low temperatures.

(3) The particle size significantly affects the LTO of silo coal. When the temperature was less than 300 °C, the maximum oxygen absorption rate ($V_{\max}$) and the differential thermal reduction value (Δ_h) of the samples increased linearly with the decrease in the particle size. Therefore, the oxidation capacity of silo coal can be enhanced at low temperatures by decreasing the particle size. This is the internal factor that enables the coal dust content to advance the spontaneous combustion of silo coal at low temperatures.

To more truly reflect the influence of coal dust content on the LTO of silo coal, in the future, it is of great practical significance to establish a mesoscale closed silo experimental platform and use temperature sensors to real-time monitor the temperature change of the coal samples with different coal dust content in the silo.

AUTHOR INFORMATION

Corresponding Author

Jianguo Liu − *Research Institute of Macro-safety Science, University of Science and Technology Beijing, Beijing 100083, China; Key Laboratory for Engineering Control of Dust Hazard, National Health Commission of People's Republic of China, Beijing 100083, China;* orcid.org/0000-0003-0736-6550; Email: liujg@ustb.edu.cn

Authors

Zihao Zhou − *School of Civil and Resource Engineering, University of Science and Technology Beijing, Beijing 100083, China*

Longzhe Jin − *Research Institute of Macro-safety Science, University of Science and Technology Beijing, Beijing 100083, China; Key Laboratory for Engineering Control of Dust Hazard, National Health Commission of People's Republic of China, Beijing 100083, China*

Tianyang Wang − *School of Civil and Resource Engineering, University of Science and Technology Beijing, Beijing 100083, China*

Shengnan Ou − *School of Civil and Resource Engineering, University of Science and Technology Beijing, Beijing 100083, China*

Shu Wang − *Key Laboratory for Engineering Control of Dust Hazard, National Health Commission of People's Republic of China, Beijing 100083, China; School of Civil and Resource Engineering, University of Science and Technology Beijing, Beijing 100083, China*

Yixuan Wei − *Key Laboratory for Engineering Control of Dust Hazard, National Health Commission of People's Republic of China, Beijing 100083, China; School of Civil and Resource Engineering, University of Science and Technology Beijing, Beijing 100083, China*

Mulati Jueraiti − *School of Civil and Resource Engineering, University of Science and Technology Beijing, Beijing 100083, China*

Complete contact information is available at:
https://pubs.acs.org/10.1021/acsomega.2c04219

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was financially supported by the National Nature Science Foundation of China (No. 52204198), the fellowship of the China Postdoctoral Science Foundation (2022M710355), and the National Key Research and Development Program of China (Nos. 2022YFC2903901, 2021YFB2301704).

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