Experimental Study on the Ignition Temperature of Combustible Dust Clouds with the Effect of $CH_4/CO/H_2$

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1 Introduction

Combustible dusts and flammable gases are widely existing in coal, petroleum, chemical, metallurgical, textile and pharmaceutical industries. However, the coupling effects between flammable gas and dust cloud introduce complexity and uncertainty on the ignition and explosion of dust clouds. Consequently, it is of significance to systematically study the effect of flammable gas on the explosion characteristics of dust clouds. By far, many studies have focused on explosion characteristics of dusts or gases, including both sensitivity (e.g., minimum ignition temperature/energy, MIT/MIE) and severity (e.g., maximum pressure rise $P_{\max}$ and maximum pressure rise rate (dP/dt) max). Some studies only investigated the explosion characteristics of dust clouds [1-4]. Other studies focused on inert mediums to prevent explosion [5-6]. Regarding the ignition mechanism, most of the literatures are related to single particle ignition [7-10]. The previous studies [11-16] found that the explosion characteristics including flame propagation behaviors of hybrid mixtures were more severe than that of the corresponding pure dust or gas. Nevertheless, few studies focused on the effect of flammable gas concentration on combustible dust explosion. Therefore, this paper aims to study the ignition temperature of dust/flammable gas with an emphasis on the role of mole fraction of flammable gas.

2 Experimental

2.1. Experimental apparatus and procedure

The basic experimental apparatus included a high-temperature furnace following the international standard ISI/IEC 80079-20-2 [17], i.e., Godbert-Greenwald (G-G) furnace was used to determine the MIT of dust clouds, and the procedure was similar with the description in [4, 18]. The volume of the cylindrical chamber was around 220 mL. The temperature of tube wall was adjusted to a desired value (max. to 1000 °C) by the

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temperature controller. For the test, the furnace tube was heated to a fixed temperature at first, then the oven was flushed more than five times with the gas mixture before each experiment. The premixed test gas was injected into the gas reservoir under a desired dispersion pressure (0.01~0.05 MPa), and then the weighted dust (0.1~0.2 g) was placed in the dust chamber, the dust was dispersed through the heated furnace tube with the flush of gas. The ignition criterion was an observation of the flame in the bottom of tube. Once an ignition occurred, the temperature was lowered with the interval of 10 ℃ until no flame observed in ten attempts. The lowest temperature with a successful ignition was the minimum ignition temperature (MITC) of dust cloud. To determine the role of small amount of flammable gas, the dust mass obtained the MITC for a given dust sample in air was taken as the basis for the ignition temperature of dust cloud (ITC) in other gas atmospheres.

2.2. Dust samples and flammable gases

Three dust samples with different volatile matters (anthracite coal, bituminous coal and sweet potato starch) as well as three flammable gases (methane, hydrogen and carbon monoxide) were used in the study. The two coal samples were from Yunnan province, China and the starch was a common grain dust in China. All the flammable gases premixed with air were no more than 60% of their lower explosion limit (LEL) in the standard condition. The components of gases were as follows: 1% CH_4/H_2/CO in 99% air, 2% CH_4/H_2/CO in air, 2.5% H_2 in air and 3% CH_4/CO in air for investigating the effect of flammable gas. The industrial analysis of dust samples and the properties of the flammable gases including autoignition temperature (AIT) were given in Table 1 and Table 2, respectively.

Table 1. Properties of dust samples

Properties	Median diameter (μm)	Volatile content (%) in mass basis	Moisture content (%) in mass basis	Ash (%) in mass basis	Fixed carbon (by diff.)	Heat of combustion (kJ/kg)
Anthracite	9.95	8.16	1.3	27.42	63.12	34000
Bituminous	18.33	23.55	1.17	27.93	47.35	24280
Starch	22	88.18	2.9	2.07	6.85	17200

Table 2. Properties of flammable gas [19]

Properties	Density (g/m^3)	Molecular weight	Explosible range (vol./%)	AIT (℃)	Specific heat capacity (kJ/kg·K)	Heat of combustion (kJ/mol)
CH_4	660	16	4.4~17	595	2.238	890.3
H_2	89.9	2	4~77	560	14.44	285.5
CO	1250	28	10.9~75.6	605	1.039	283.0

3 Ignition kinetics of dust cloud

Ignition of solid-fuel particles is a complex interaction phenomenon among different mechanisms including heat and mass transfer, evaporation, devolatilization, heterogeneous oxidation on the particle surface, and volatiles oxidation in the gas-phase [20]. The possible reaction pathways for the ignition of dust clouds are proposed in Fig. 1. With high heating rates, both the rates of heterogeneous reactions on the particle surface and the homogenous reactions in the gas phase are increased. The addition of extra flammable gases may have a strong impact on the ignition kinetics. The concentration and type of the flammable gas may also affect the ignition temperature of dust.

MIT is one of the most important factors for industrial safety [21]. However, the ignition mechanism of dust clouds under MIT conditions has not been well studied. Our previous work concluded that heterogenous ignition dominates at MIT conditions for a dense dust cloud of three bituminous coal with volatile matter from 26.9% to 38.2% [22]. However, the ignition mechanism of starch dust clouds with extremely high volatile content may not follow this conclusion. Dust samples presented in the G-G furnace in this work (0.1 ~ 0.2 g) can be regarded as a dense cloud, i.e., the number of particles N numerically far outweighs the ratio of cloud diameter d_{cl} to particle diameter d_{p}, $N \gg d_{\mathrm{cl}} / d_{\mathrm{p}}$ [23]. Moreover, pure homogeneous ignition only occurs under high temperature and low oxygen for high volatile coal [24].

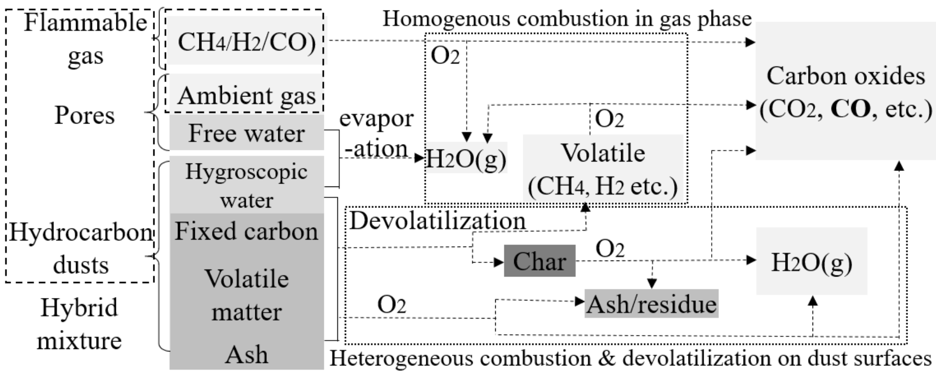

Fig. 1. Proposed reaction pathways proceeding in the ignition of dust cloud [25].

4 Results and discussion

Figure 2 shows ignition temperatures (ITCs) of anthracite coal dust (low volatile) with presence of one-component flammable gas. The addition of all three flammable gases reduces the ITC of anthracite, and the ITC decreases significantly with increasing flammable gas mole fraction. The ITC of a fix mass/concentration (0.2 g) of anthracite coal decreases from 610 ℃ to 560 ℃, 580 ℃ and 570 ℃ as 3% CH_4, 3% CO and 2.5% H_2 are added in air, respectively. This finding agrees with Addai's work [14] that the presence of 2% CH_4 or 3% H_2 can respectively decrease the ITC of CN4 dust from 640 ℃ to 630 ℃ and 620 ℃. However, the current promoted effect of the addition of small amount of flammable gases is much stronger than the observations of Addai's work [14]. Considering the similarity of chemical composition between the anthracite coal and CN4, i.e., both dust samples have high fixed carbon content and low volatile matter content, the probable reason is that the addition of flammable gases has a greater impact on the ignition of dense dust cloud. Because Addai's study focuses on dilute dust clouds, while the current work studies dense dust clouds. Specifically, the heterogeneous combustion reaction occurring on dust surface dominates the ignition of the anthracite coal dust cloud because of its low volatile matter. Exothermic oxidation reaction of additional flammable gases increases the gas phase temperature in the furnace, promoting the ignition on the particle surface of the anthracite dust cloud. Therefore, the actual temperature of furnace with the presence of the flammable gases is higher than the set temperature, suggesting a reduction of IT as a macroscopic performance. The promotion effect on the ignition of the anthracite coal dust cloud follows CH_4 > H_2 > CO as shown in Fig. 2. Firstly, CH_4 with the highest combustion heat compared with the other two gases could release more heat during oxidation reactions (see Table 2), i.e., heating promotion effect. Regarding to H_2 and CO having the comparable combustion heat, the reason why the promotion impact of CO is inferior to H_2 might be that the addition of hydrocarbon fuel C_xH_y (e.g. CH_4 and H_2) compensates the volatile matter content for anthracite coal dusts since C_xH_y is the main component of the pyrolyzed or devolatilized products of coal. In other words, the hybrid mixture of anthracite coal dust and C_xH_y might has an equally global ignition characteristic with a medium-volatile bituminous coal dust.

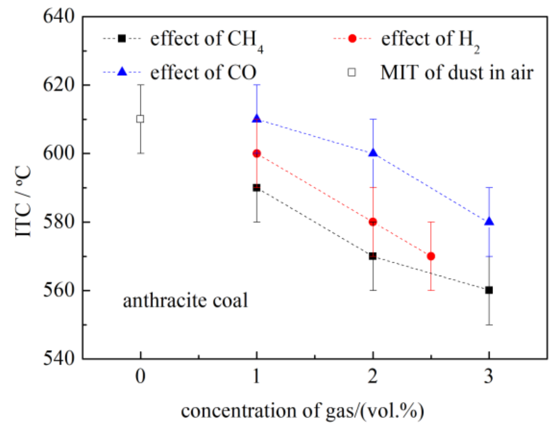

Fig. 2. Effect of one-component flammable gas on IT of anthracite dust (0.2 g).

In order to find out the effect of flammable gases on IT of dust cloud with high volatile, sweet potato starch dust is used in the current work. Fig. 3a illustrates that all three flammable gases have an ignorable effect on ITC of starch dust (0.15 g), considering the experimental error ± 10 °C. This result presents a large difference compared with the observations of anthracite coal dust shown in Fig. 2, which is caused by the physicochemical properties of these two dusts. Unlike the predominated heterogenous ignition of anthracite coal dust, ignition mechanism of starch dust might be different due to its high volatile matter content. Homogeneous ignition occurs when the volatile matter content or the cloud density is high [7, 23]. In this scenario [7], MIT of dust cloud (T_{mci}) only depends on the diameter (d_{cl}) and density (ρ_{cl}) of dust cloud, showing an independent correlation with the particle size of dust according Eq. (1).

$$T_{\mathrm{mci}}^{\alpha-2} = \frac{\mathrm{constant}}{d_{\mathrm{cl}}^{2}\rho_{\mathrm{cl}}} \tag{1}$$

To estimate the ignition mechanism of starch dust cloud, additional experiments were conducted to determine the dependence of MITC on the particle size (d_{p}). The result reveals that MITC of starch dust does not vary with particle size (see Fig. 3b), i.e., homogeneous ignition dominates for starch dust cloud. In this scenario, the presence of extra flammable gases has a limited effect. As a result, ITCs of the hybrid mixtures are roughly the same with pure starch dust as shown in Fig. 3a.

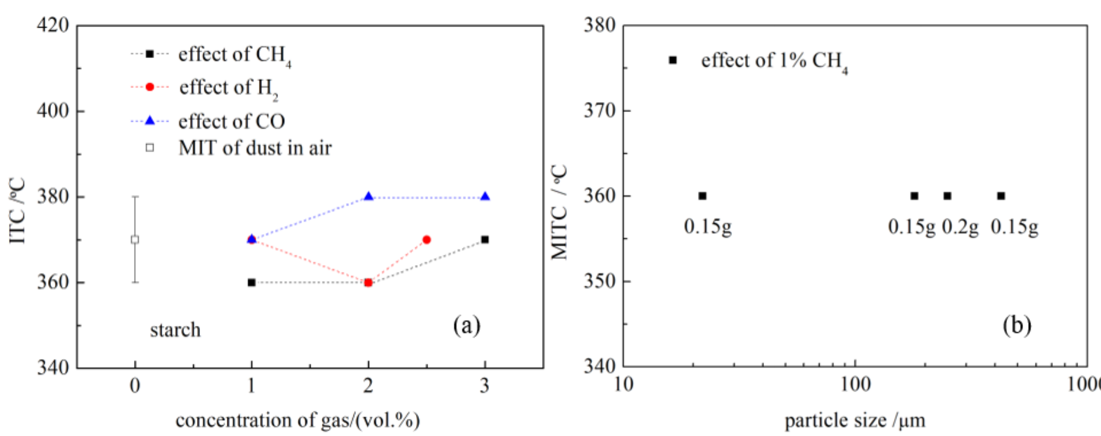

Fig. 3. (a) effect of flammable gas on ITC of starch (0.15 g), (b) effect of particle size on MITC of starch.

Figure 4 shows the effect of the presence of flammable gases on the ITCs of bituminous coal (0.1 g). The ITCs decrease by 10 ℃ with the addition of 1% gas for all three flammable gases, however, clearly vary with the type and the mole fraction of flammable gases. The ITC of bituminous coal dust decreased first and then increased with further increasing of CH_4 mole fraction in explosion atmosphere. Strangely enough, the ITC of bituminous coal is even higher than pure bituminous coal when CH_4 mole fraction exceeds 2%. The reason behind this unnormal phenomenon needs further investigations. The addition of H_2 or CO was not found to have this negative effect on the ITC of bituminous coal. The ITC of bituminous coal decreases with the addition of H_2 or CO, but this trend tends to be stable after the mole fraction of these two gases reaching a certain value. Unlike the ignition of anthracite and starch dust clouds, the combustion of bituminous coal is much more complicated, existing both two ignition mechanisms, i.e., homogenous ignition and heterogeneous ignition [9]. Therefore, the ignition process of hybrid bituminous coal/flammable gas mixtures exists much more prominent competition ignition mechanisms and parallel reactions in both gas phase and dust particle surface.

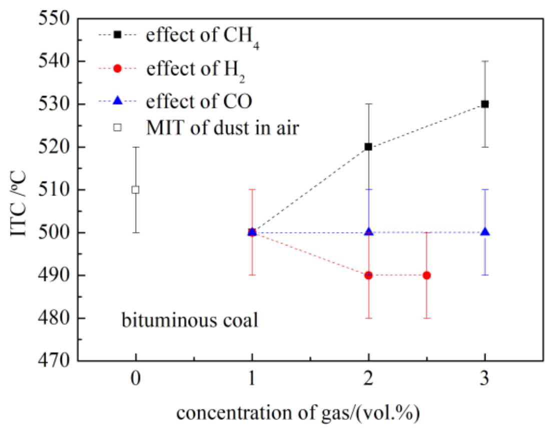

Fig. 4. Effect of one-component flammable gas on ITC of bituminous coal (0.1 g).

5 Conclusions

The effects of three flammable gases on the ITC of dusts with different volatiles have been investigated with the help of Godbert-Greenwald furnace. Our present work has affirmed that the addition of flammable gas had distinct influence on the ITC of dust containing different volatiles. The ignition of anthracite coal dust was heterogeneous, the addition of flammable gases significantly decreased the ITC of dust, and the IT of hybrid mixture was inversely proportional to the increase of gas mole fraction. Conversely, homogeneous ignition dominated for starch dust cloud because of its high volatile matter, thus the presence of extra flammable gases had a limited effect on the ITC of starch dust. The ignition mechanism of bituminous coal dust was complex, heterogeneous ignition and homogeneous ignition existed simultaneously, the results of hybrid mixtures of bituminous coal dust and three flammable gases were very different from the other two dusts: the addition of CO or H_2 reduced the ITC of bituminous coal but the trend tended to be stable after the mole fraction of these two gases reaching a certain value, whilst the ITC of bituminous coal decreased at first when CH_4 added, but then increased even higher than pure bituminous coal as CH_4 mole fraction exceeds 2%. These results improve our understanding of the ignition behavior of dust clouds with the presence of small amount of flammable gas.

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