Effect of Inert Gas CO₂ on Deflagration Pressure of CH₄/CO
Zhenmin Luo,* Yang Su, Ruikang Li, Xiaokun Chen, and Tao Wang

ABSTRACT: To study the effect of CO₂ on the explosion characteristics of CH₄/CO, the explosion experiments of the effect of different volume fractions of CO₂ on CH₄/CO deflagration were carried out by using the self-developed pipeline gas explosion experimental platform. The explosion characteristics of premixed gas are studied from the aspects of explosion peak pressure and time of reaching the peak pressure. The results show that the effect of CO on the deflagration of methane with a different volume fraction is the result of the interaction of the elementary reaction and the oxygen content in the reaction system. Two percent of the CO promoted the methane explosion in the oxygen-rich state, while it showed a damping effect in the oxygen-poor state. CO₂ has different inhibitory effects on different volume fractions of methane. Experiments show that the addition of 20% CO₂ can effectively inhibit the deflagration of methane. The addition of CO₂ has a stronger inhibitory effect on the mixed gas of CH₄/CO under the condition of poor oxygen but less on the mixed gas under the condition of rich oxygen.

1. INTRODUCTION
CO and CH₄ are common gases encountered in chemical production and coal mines. If they are mixed with air, then they can form explosive mixed gases under certain conditions. Because of the complexity of underground gas in coal mine and the wide use of multicomponent mixed gas in industrial production, explosion accidents of mixed combustible gas occur from time to time.1-3 Furthermore, it has the characteristics of multiplication and recurrence, which are serious threats to the safety of people and property. The deflagration of the CH₄/CO gas mixture has been widely studied by its predecessors.3-8 In terms of the promotion and damping effect of CO on gas explosion, Deng et al.5,10 numerically simulated the explosion process of CH₄/CO two-component combustible gas and explored the damping effect of CO on multicomponent gas. Luo et al.11 found that the existence of CO has a certain damping effect on the explosive chain reaction of CH₄/CO mixed gas. Jia et al.12,13 used the method of numerical simulation to analyze the kinetic process of the elementary reaction of CH₄ explosion affected by H₂ and CO and revealed their promotion and damping mechanism. In the aspect of the influence of CO on the limit of gas explosion, Wang et al.14 found that the addition of CO can restrain the explosion of CH₄ to a certain extent after CO reaches the optimal explosion concentration. Hu et al.15 studied the explosion limit of the mixture of H₂, CH₄, and CO under different influencing factors. The explosion limit characteristics of many kinds of mixed combustible gases are analyzed theoretically by Shrestha et al.16 Kondo et al.17 have tested the explosion limit of a large number of binary and ternary mixed combustible gases. Chen et al.18 studied the effect of CO addition on the laminar combustion rate by numerically simulating the CH₄/CO/air flame structure under lean burn conditions. Cheng and Deng19 studied the influence of CO on the position and size of the CH₄ explosion triangle and concluded that CO gas will cause the CH₄ explosion triangle to move and extend to the left and downward. Luo et al.20 studied the effects of CO on the explosion of CH₄ and the emission spectra of free radicals.

The above studies focus on the explosion limit of mixed gases and the effect of CO on methane, and some scholars
have found that CO₂ can affect the formation of nitrogen oxides in syngas.²¹⁻²³ At the same time, at present, the research on the explosion of inert gas to mixed combustible gas is mostly focused on the laminar combustion velocity of inert gas to mixed gas.²⁴⁻³² For example, Lamoureux et al.,³³ Dahoe,³⁴ and Qiao et al.³⁵ have measured the laminar combustion velocity of mixed gases with different equivalence ratios under the action of diluted gases. In addition, Lu et al.³⁶ used inert gas to study the effect of inert gas on methane deflagration from three aspects: the limit oxygen volume fraction, the explosion limit, and the explosion suppression effect. Yossefi et al.³⁷ used the method of numerical simulation to get the very important chemical effect of CO₂ on mixed combustible gas. Mitu et al.³⁸ studied the effect of inert gas on a methane–air deflagration propagation index and the effect of initial pressure on the deflagration of methane–air in a small airtight container.³⁹ In addition, in the aspect of LPG, Razus et al.⁴⁰,⁴¹ studied the deflagration characteristics of LPG. However, most studies focus on the effect of inert gas on the explosion characteristics of a single combustible gas, such as methane, but there are few studies on the deflagration of multicomponent combustible gas. In this paper, the combustion characteristics of methane gas containing CO under CO₂ dilution are studied to better understand the combustion and inhibition characteristics of multicomponent mixed gas and provide a theoretical basis for the safe use of multicomponent combustible gas.

2. EXPERIMENTAL SETUP AND METHODS

2.1. Experimental Apparatus. The comprehensive experimental system of pipeline gas (dust) explosion suppression developed by our university is used in this paper. The experimental system consists of an explosion reaction pipeline, an automatic control unit, a vacuum unit, a gas distribution unit, an ignition device, a dynamic pressure data acquisition and storage unit, a notebook computer, and so on. The structure of the reaction device is shown in Figure 1. During the experiment, the external temperature for all experiments was 22–25 °C, the humidity was 52–64% RH, and the initial pressure was 0.1 MPa.

2.2. Materials. In the experiment, the volume fractions of CO added were 0, 0.4, 1.2, and 2.0%. The volume fractions of CH₄ were 7, 8.5, 9.5, 10.5, 11, and 12%, corresponding to oxygen-rich state, equivalent state, and oxygen-poor state, respectively. The volume fractions of CO₂ added were 0, 10, 15, and 20%. The purities of CO, CH₄, and CO₂ gases used are all greater than 99.9%.

ϕ₂ represents the percentage of CO₂ mentioned in the gas mixture.

\[
\phi_{CO_2} = \frac{V_{CO_2}}{V_{CO} + V_{CO_2} + V_{CH_4} + V_{AIR}} \times 100\%
\]

In eq 1, \(V_{CO_2}\), \(V_{CO}\), \(V_{CH_4}\), and \(V_{AIR}\) stand for the volume fractions of CO, CO₂, CH₄, and air, respectively.

The partial pressure gas distribution method was used in the experiment. First of all, a vacuum pump was used to remove part of the air from the explosion container to produce negative pressure in the container, and then, the experimental gas was put into the pipeline to reach the preset working condition concentration. The experimental process was controlled by one button to ensure the reliability of the experiment. No less than three repeated tests were carried out for each experimental condition to ensure the repeatability of the experiment and the accuracy of the data.

3. RESULTS AND DISCUSSION

3.1. Effect of CO on CH₄ Deflagration Pressure. The effect of different volume fractions of CO on methane deflagration is different. As can be seen from Figure 2, when the volume fraction of CO is low, CO has a certain damping effect on each volume fraction of methane. As shown in Figure 2, with the addition of 0.4% CO, the peak explosion pressure of each volume fraction of methane is less than that of pure methane. When the addition of CO is 1.2%, the addition of CO can promote methane explosion, that is, the peak...
explosion pressure is higher than that without CO. When 2% CO was added, CO promoted the explosion of methane in the oxygen-rich state, while it showed a damping effect when the methane was oxygen-poor.

Figure 3 shows the time when the CH₄/CO gas mixture reaches the peak explosion pressure under the action of CO. As can be seen from the figure, the effect of the CO addition ratio on methane explosion can be divided into three categories: ① under the oxygen-rich condition, that is, when the volume fractions of methane are 7.0 and 8.5%, the 0.4% volume fraction of CO has a certain damping effect on the explosion of methane, that is, when the volume fraction of added CO is low, it increases the time for the mixed gas to reach the peak pressure. When the volume fractions of CO are 1.2 and 2%, the addition of CO shortens the time for the mixture to reach its peak, that is, it promotes the explosion of methane. ② Under oxygen-poor conditions, that is, when the volume fractions of methane are 10.5, 11, and 12%, the addition of CO increases the time for the CO/CH₄ mixture to reach the peak explosion pressure, and it can be considered that the addition of CO weakens the reaction rate of methane. ③ When the volume fraction of methane is 9.5%, CO shortens the time for the explosion pressure of the mixture to reach its peak.

In addition, when the addition ratio of CO is low, except for the addition of 9.5% methane, the time for the other volume fraction of methane to reach the peak increases slightly, that is, the addition of CO slows down the reaction rate of methane and increases the time for reaching the peak value. When the volume fraction of CO is high, it can promote the explosion of the mixed gas under non-oxygen-poor conditions. To sum up, the addition of CO in the experimental range did not change the peak pressure trend of each volume fraction of methane, that is, the peak pressure increased at first and then decreased with the increase of the methane volume fraction, which reached the maximum when the methane volume fraction was 9.5%. From the peak pressure and the time of reaching the peak pressure, it can be seen that the addition of CO can promote and inhibit methane, when the inhibition effect is greater than the promotion effect, the damping effect appears, and when the promotion effect is greater than the inhibition effect, it can enhance the methane explosion. The specific manifestations are as follows: when 0.4% volume fraction of CO is added, it has a certain inhibitory effect on the deflagration of methane gas under oxygen-poor and oxygen-rich conditions, and it can be seen from the figure that the peak pressure decreases and the time to reach the peak pressure increases, indicating that the addition of CO slows down the reaction rate of methane and increases the time for it to reach the peak value. This is because the presence of CO reduces the key free radicals HCO, HO₂, etc., needed in the process of CH₄ explosion reaction. It also reduces the formation of important intermediates such as H₂O₂ and CH₃, which leads to the damping effect. When 1.2% CO is added, the peak deflagration pressure of all volume fractions of methane is higher than that of pure methane, but the time to reach the peak pressure under oxygen-rich and equivalent conditions decreases and increases under oxygen-poor conditions, indicating that the addition of 1.2% CO can promote methane reaction under oxygen-rich and equivalent conditions. When 2.0% volume fraction of CO is added, the pressure peak value of the mixed gas increases and the time to reach the peak value decreases under oxygen-rich conditions compared with pure methane, while under oxygen-poor conditions, the pressure peak decreases and the peak time increases, indicating that, when 2.0% CO is added, oxygen in the mixed gas plays a key role in the explosion, and the addition of 2.0% CO under oxygen sufficient conditions promotes methane. On the contrary, it has an inhibitory effect. The above phenomena show that the effect of CO on methane deflagration with a different volume fraction is the result of the interaction of the elementary reaction and the oxygen content in the reaction system.

3.2. Effect of CO₂ on CH₄ Deflagration. It can be seen in Figure 4 that the addition of CO₂ has a great influence on the deflagration of methane. When the volume fraction of CO₂ is less than 5%, all volume fractions of methane can be ignited, and the peak explosion pressure first increases and then decreases with the increase of methane volume fraction and reaches the maximum value when the volume fraction is 9.5%. When the volume fraction of CO₂ is more than 5%, the deflagration pressure decreases gradually with the increase of methane volume fraction. In addition, when the addition of CO₂ can cause the methane to detonate, the mixed gas containing CO₂/methane is not detonated (at the dotted coil in Figure 4). This is because the inhibitory effect of CO₂ on methane is mainly reflected in three aspects: on the one hand, the inert gas is diluted and cooled; on the other hand, it is the direct combustion product of methane and CO, which aggravates the difficulty of forward reaction; the third is to accelerate the annihilation of key free radicals in the process of reaction kinetics in the form of a third body.

Figure 4 shows that, when the volume fraction of methane is large, there are more cases of flameout. For example, when the volume fraction of methane is 12%, 10% CO₂ can completely inhibit the explosion of methane. The reason for this...
phenomenon may be that the volume fraction of oxygen in the mixed gas is insufficient to meet the needs of methane deflagration. The experimental data show that 20% volume fraction of CO₂ can effectively inhibit the deflagration of methane, which is also similar to the results in the literature. The peak deflagration pressure of methane shows different phenomena under different CO₂ addition ratios. Figure 5 shows that, under the action of CO₂, the peak explosion pressure of methane decreases with the increase of the addition ratio, which can be divided into two categories: ① when the addition ratio of CO₂ is less than 10%, the addition ratio does not affect the peak explosion pressure of methane, that is, with the increase of the volume fraction of methane, the explosion pressure increases at first and then decreases and reaches the maximum pressure peak when the volume fraction of methane is 9.5%. ② When the addition ratio of CO₂ is more than 10%, the peak explosion pressure of methane decreases with the increase of methane volume fraction. This is similar to the results in the literature.

In addition, the effect of CO₂ on different volume fractions of methane is different under a different addition ratio, and the addition of CO₂ has a lower effect on the smaller volume fraction of methane. For example, under the 5% CO₂ addition ratio, the pressure reduction rates of methane in each volume fraction from 7 to 12% are 2.9, 1.3, 5.7, 7.5, 16.5, and 33.8%. The increase of the CO₂ addition ratio decreases the volume fraction of oxygen in the mixed gas, slows down the reaction rate, and increases the time for the explosion to reach the peak pressure. In summary, the addition of CO₂ has two inhibitory effects on the deflagration of methane: physical effect and chemical effect. In the physical aspect, by diluting the combustible gas mixture and reducing its concentration, the chemical reaction is carried out very slowly, which slows down or even suppresses the deflagration of methane. The chemical effect refers to the secondary reaction of the inert gas CO₂ with the intermediate products of methane deflagration reaction under the condition of high temperature and high pressure produced by methane combustion and explosion, which can restrain the development of chain explosion reaction by absorbing and capturing active groups so that methane deflagration can be suppressed.

3.3 Effect of CO₂ Addition on CH₄/CO. Under a different methane volume fraction, the effect of CO₂ on the CH₄/CO mixture is different. Figure 6 shows the peak pressure change of methane with different volume fractions of CO under the action of CO₂. It can be seen from the figure that the addition of CO₂ decreases the deflagration pressure of the CH₄/CO mixture, but the effect on each volume fraction of methane mixture containing CO is different. Figure 6a shows the 7% volume fraction of methane. It can be seen from the figure that, when the volume fraction of CO₂ is less than 15%, the peak pressure of the CH₄/CO gas mixture increases first and then decreases and then increases with the increase of the CO volume fraction. However, when CO₂ is added to 20%, the peak pressure of the gas mixture decreases with the addition of CO. Figure 6b and Figure 6c shows the peak pressure when the volume fractions of methane are 8.5 and 9.5%, respectively. When the volume fraction of CO₂ is 5%, the change of the peak pressure is close to that without adding CO₂. With the increase of methane volume fraction, the effect of CO₂ on the CH₄/CO gas mixture increases gradually. In addition, the effect of CO₂ on the CH₄/CO gas mixture under oxygen-rich and oxygen-poor conditions is different. Take 5% CO₂ as an example. When the volume fraction of methane is 11%, the pressure drop rates of mixed gas (oxygen-poor condition) are 17.86, 18.90, 24.78, and 19.40%, while when the volume fraction of methane is 8.5%, the pressure drop rates of mixed gas (in oxygen-rich condition) are 6.77, 8.51, 5.39, and 1.68% compared with those without CO₂. It can be seen that the addition of CO₂ has a stronger effect on the mixed gas under the condition of poor oxygen but less on the mixed gas under the condition of rich oxygen. The reason for this phenomenon may be that the mixed gas can be in better contact with oxygen under oxygen-rich conditions. Taking the addition of 10% CO₂ to the CH₄ of 8.5 and 10.5% volume fraction as an example, the equivalent ratio of 8.5% CH₄ is 0.844, the equivalent ratio of 8.5% CH₄ + 10% CO₂ is 0.993%, the equivalent ratio of CH₄ is 1.258, and the equivalent ratio of 10.5% CH₄ + 10% CO₂ is 1.117. It can be seen that the addition of CO₂ makes the equivalent ratio of the whole mixed gas close to 1 under the oxygen-rich condition, which makes the reaction easier to a certain extent, while under the oxygen-poor condition, the equivalent ratio of the mixed gas added by CO₂ is still greater than 1, which has little effect on the reaction.

4. CONCLUSIONS
In this paper, the effect of the inert gas CO₂ on the explosion pressure of mixed gas in the pipeline is studied experimentally. The main conclusions are as follows:

1) During the explosion of CH₄/CO mixed gas, CO has a damping effect on the deflagration of methane under oxygen-poor conditions, while it can promote the explosion of methane under oxygen-rich conditions. In terms of the time to reach the peak pressure, the addition of CO has different effects on different volume fractions of methane, that is, oxygen-rich conditions, oxygen-poor conditions, and methane explosion with a volume fraction of 9.5%. The effect of CO on the deflagration of methane with a different volume fraction is the result of the interaction of the elementary reaction and the oxygen content in the reaction system.

2) Compared with methane under oxygen-rich conditions, CO₂ has a better inhibitory effect on methane under oxygen-poor conditions. With the increase of CO₂ addition, methane is completely inhibited in different degrees. For example, methane with a volume fraction of 12% begins to be completely inhibited when the volume fraction of CO₂ is
10%, while 9.5% methane is not completely inhibited by the addition of CO2 in the experimental conditions.

3) The deflagration pressure of the CH4/CO mixture decreases with the addition of CO2, and the effect of CO2 on the deflagration of the CH4/CO mixture with a different volume fraction is different. The addition of CO2 has a stronger inhibitory effect on the mixed gas under the condition of poor oxygen but less on the mixed gas under the condition of rich oxygen.

**Figure 6.** Effect of CO2 on the CO/CH4 mixture.

![Graphs](https://pubs.acs.org/10.1021/acsomega.0c02686)

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**Notes**

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