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Experimental Investigation of the Impact of CO, C_2H_6, and H_2 on the Explosion Characteristics of CH_4

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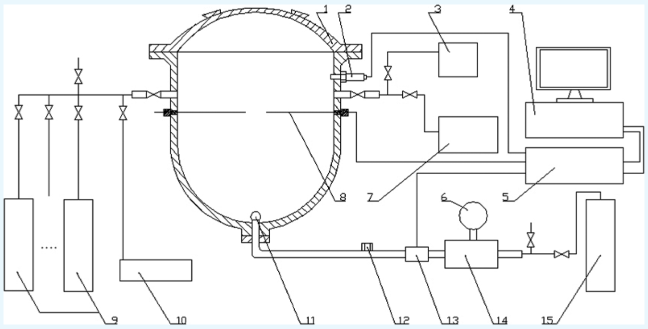
ABSTRACT: Gas explosions are destructive disasters in coal mines. Coal mine gas is a multi-component gas mixture, with methane (CH_4) being the dominant constituent. Understanding the process and mechanism of mine gas explosions is of critical importance to the safety of mining operations. In this work, three flammable gases (CO, C_2H_6, and H_2) which are commonly present in coal mines were selected to explore how they affect a methane explosion. The explosion characteristics of the flammable gases were investigated in a 20 L spherical closed vessel. Experiments on binary- (CH_4/CO, CH_4/C_2H_6, and CH_4/H_2) and multicomponent ($CH_4/CO/C_2H_6/H_2$) mixtures indicated that the explosion of such mixtures is more dangerous and destructive than that of methane alone in air, as measured by the explosion pressure. Furthermore, a self-promoting microcirculation reaction network is proposed to help analyze the chemical reactions involved in the multicomponent ($CH_4/CO/C_2H_6/H_2$) gas explosion. This work will contribute to a better understanding of the explosion mechanism of gas mixtures in coal mines and provide a useful reference for determining the safety limits in practice.



1. INTRODUCTION

Gas explosions are ruinous disasters in coal mines.[1−4] Explosions of fuel−air mixtures are characterized by specific parameters, such as explosive limits, maximum explosion pressure, maximum rate of pressure rise, and time to reach the maximum explosion pressure. These parameters reflect explosion intensity and destructiveness. A number of experimental studies on methane (CH_4) explosion characteristics can be found in the literature in the last decades.[5−9] Coward and Jones[10] and Zabetakis[11] investigated the flammability characteristics of combustible gases and vapors under a variety of environmental conditions. It is worth noting that the experimental results depend on certain factors of the investigated process, such as the size and type of explosion chambers, energy and type of ignition source, initial pressure and temperature, and flammable mixture flow.[12,13] The explosion characteristic tests of the flammable gases were conducted from the mid-1980s through the 1990s at the Pittsburgh Research Laboratory (PRL) in different volume chambers (8, 20, 120, 25, and 500 L).[14−17] The experimental data reported include lower explosive limit (LEL), upper explosive limit (UEL), peak explosion pressures, and the maximum rate of pressure rise. The tests were performed at ambient temperature and pressure under both quiescent and turbulent conditions. Initial temperature and pressure have tremendous effect on explosion parameters.[18−22] The experimental results show that the explosive limits of methane/natural gas can be significantly extended at high temperatures and high pressures, and the UEL is more sensitive than the LEL as pressure and temperature increase.[23] The peak explosion pressure is slightly reduced at high-temperature conditions and gradually increases with the initial pressure.[23,24] Moreover, many scholars have investigated the influence of initial ignition energy and initial turbulence on the explosion behavior of methane/air mixtures.[16,17,25,26] The scholars proved that the level of initial ignition energy significantly impacts the flame and explosion characteristics and also extends the explosive limits of methane.[27] The gas flow turbulence increases the maximum explosion pressure and burning velocity.[28] Because of the great catastrophe caused by coal dust explosions, many researchers have made great efforts to explore the mechanism of CH_4/coal dust mixtures in recent years. The researchers found that the presence of coal dust with methane not only increases the explosion pressure but also accelerates the time of the explosion.[29] The explosion risk of hybrid CH_4/coal dust is much higher than that of pure coal

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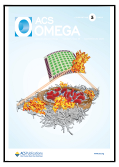

dust explosion.[30] Furthermore, coal mine gas is a multi-component gas mixture, with methane being the dominant constituent. However, most reported studies treated the mine gas as pure methane without considering carbon monoxide, hydrocarbons, hydrogen, and other flammable gases; as a consequence, the results may significantly deviate from the reality in coal mines.

Hydrogen (H_2), although appearing in small amounts in coal mine gas, has a wide explosion range with a low minimum ignition energy, thus posing a high explosion risk.[31−34] Several studies have reported the effects of H_2 addition on the explosion characteristics of hydrocarbon fuel streams or natural gas, in particular an increase in the laminar burning velocity[35−38] and also a decrease in the laminar flame thickness.[36] Jackson et al.[39] carried out a combined experimental and numerical investigation on the effects of H_2 addition to lean premixed CH_4 flames. The results indicate a significant enhancement of lean flammability limits for relatively small amounts of H_2. The effects of hydrogen concentration on spherically propagating laminar hydrogen/methane/air flames were studied at different equivalence ratios at atmospheric pressure by Okafor et al.[40] The results showed that an increase in hydrogen concentration in the binary fuel led to an increase in laminar burning velocity. Yu et al.[41] investigated the effects of hydrogen addition on the propagation characteristics of methane/air premixed flames at different equivalence ratios in a venting duct. The results indicated that the tendency toward flame instability increased with the fraction of hydrogen, and the premixed hydrogen/methane flame underwent a complex shape change with the increasing hydrogen fraction. Using a standard 20 L spherical explosion vessel, the explosion characteristics of H_2/CH_4/air and CH_4/coal dust/air mixtures were investigated by Li et al.[42] The results showed that the presence of molecular hydrogen would significantly increase the maximum explosion pressure and the rate of pressure rise of H_2/CH_4/air mixtures.

Some research was performed on the explosion characteristics of binary mixtures such as CO/CH_4, C_2H_4/CH_4, and C_2H_6/CH_4[43−45] or ternary systems such as $H_2/CH_4/CO$ in the context of the nitrogenous fertilizer industry.[46] Experimental results revealed that the flammability limits of the mixture gas are related to many factors, such as the inherent properties of the flammable gases, the state of the mixture gas (temperature, pressure, and composition), ignition energy, size and geometry of the container, and the flame spread direction. Zheng et al.[47] proposed a BP neural network model to predict the minimum and maximum explosive limits of a flammable gas mixture containing H_2, CH_4, and CO based on experimental data. It is worth noting that the flammable gas mixtures investigated belong to rich H_2/lean CH_4 fuel, which is quite different from the coal mine gas.

With respect to the explosion characteristics of coal mine gas, existing results are still largely based on single- or binary-component flammable gases. Beyond binary mixtures, few studies were conducted with gas compositions that are of interest to the chemical industry but not necessarily relevant to coal mines. In coal mines, the composition of the flammable gases varies according to the conditions in the coal mine. Besides CH_4, CO is the main product of low-temperature oxidation of coal, just like C_2H_6, C_3H_8, C_2H_4, and other hydrocarbon gases or the product of degradation and cracking of coal after the temperature reaches a certain threshold.[48,49] C_2H_6 is a gaseous hydrocarbon produced during the low-temperature oxidation of coal and accounts for a large proportion, especially in the coalbed gas of kerosene symbiotic mines.[49] H_2 is mainly the product of degradation and cracking of coal after the temperature reaches the threshold value.[49] The H_2 content is generally small, but H_2 has the highest explosion risk in coal mine gas. Therefore, the content and composition of the flammable gases are not fixed but vary with time and location.

As mentioned above, the explosion characteristics of multicomponent flammable gases including CH_4, CO, C_2H_6, and H_2 have been reported in the literature. However, existing studies are either focused on binary mixtures, for example, CH_4/C_2H_4,[43] CH_4/CO,[43,44] and CH_4/C_2H_6,[45] or the mixture composition is very different from that of typical gases in coal mines, for example, Zheng et al.[47] studied a $CH_4/H_2/CO$ mixture relating to the rich H_2/lean CH_4 fuel. Against this background, the purpose of the present study is to help bridge this knowledge gap by experimentally investigating the effect of multicomponent flammable gases on the explosion characteristics of methane in the context of coal mines. Therefore, three flammable gases (CO, C_2H_6, and H_2) were selected and their individual explosion properties were studied, prior to the exploration of the binary- (CH_4/CO, CH_4/C_2H_6, and CH_4/H_2) and multicomponent ($CH_4/CO/C_2H_6/H_2$) mixtures. The novelty of our work is thus about the explosion characteristics of $CH_4/CO/C_2H_6/H_2$ mixtures, with the compositions relevant to the situations in coal mines. The quantitative results of the explosive limits and pressures of such mixtures have not been reported before. Furthermore, a self-promoting microcirculation reaction network was proposed to help analyze the chemical reactions involved in the multicomponent ($CH_4/CO/C_2H_6/H_2$) gas explosion. The reaction network suggests that chain initiation and chain-branching reactions in $CH_4/CO/C_2H_6/H_2$/air mixtures could happen more easily and faster than in CH_4/air mixtures, which could help further study the explosion reaction mechanism of multicomponent flammable gases.

2. RESULTS AND DISCUSSION

2.1. Parameters for Assessing Explosion Intensity. The explosion experiments at particular gas concentrations will generate pressure−time curves, which normally show the pressure increases to a peak value before it falls. This peak pressure is designated the maximum explosion pressure (P_{max}). P_{max} is specific to the concentration of the flammable gas, and thus we can obtain the extreme explosion pressure ($\mathrm{Max}_{P_{max}}$), that is, $\mathrm{Max}_{P_{max}} = \mathrm{Max}\{P_{max1}, P_{max2}, P_{max3}, ..., P_{maxn}\}$, where 1, 2, 3, ..., n are the different gas concentrations investigated in the experiments. The corresponding gas concentration at $\mathrm{Max}_{P_{max}}$ is the most dangerous concentration (C_m), which is normally slightly greater than the stoichiometric concentration for this gas−air reaction.

Another measure used is the explosion risk degree (F), proposed by Kondou and Rock:[50] $F = 1 - (L/U)^{0.5}$, where U and L are the UEL and LEL, respectively, of a particular flammable gas.

2.2. Explosive Limits of CH_4, CO, C_2H_6, and H_2. The explosive limits of CH_4, CO, C_2H_6, and H_2 were measured and given in Table 1, which also show that the explosion risk degree of CO, C_2H_6, and H_2 is higher than that of methane. Therefore, it is necessary and significant to study how the presence of a small amount of CO, C_2H_6, or H_2 affects the

Table 1. Explosive Limits of Monocomponent Flammable Gas

flammable gas	experimental data LEL/%	experimental data UEL/%	explosion risk degree F
CH_4	5.05	15.35	0.426
CO	12.95	70.15	0.570
C_2H_6	3.15	12.55	0.499
H_2	4.55	71.85	0.748

Table 2. Impact of CO, C_2H_6, or H_2 Addition on the Explosive Limits of CH_4

flammable gas	added concentration/%	explosive limits of CH_4 LEL/%	explosive limits of CH_4 UEL/%
CO	0.5	5.05	15.25
	1.0	4.95	14.95
	2.0	4.75	14.45
	3.0	4.25	13.95
C_2H_6	0.5	4.55	14.35
	1.0	3.95	13.45
	1.5	3.25	12.75
	2.0	2.25	12.15
H_2	0.5	4.75	15.85
	1.0	4.15	15.55
	1.5	3.55	15.25
	2.0	2.85	15.05

explosion characteristics of methane in the context of coal mines.

2.3. Impact of CO, C_2H_6, and H_2 on Methane Explosion Characteristics. In typical coal mine gases, the CO concentration is no more than 3.0% and the C_2H_6 and H_2 concentrations less than 2.0%. Therefore, CO of four different concentrations (0.5, 1.0, 2.0, and 3.0%) and C_2H_6 and H_2 of four different concentrations (0.5, 1.0, 1.5, and 2.0%) were added to CH_4/air mixtures for experimentation, respectively.

The explosive limits of flammable gas have been well investigated by Coward, Hughes and Raybould, and Zabetakis.[5,10,11] Le Chatelier's formula is widely used to determine the explosive limits of the flammable gas mixtures. For hydrocarbon−air mixture, the prediction of Le Chatelier's formula is relatively accurate, but for the gas mixture containing H_2 or CO, this does not fit.[10,43,47,51,52] The explosive limits of the binary mixed gases CH_4 + CO and CH_4 + C_2H_4 were determined by Deng et al.[43] The results show that there is a certain gap between the value calculated by Le Chatelier's formula and the experimental data. Furthermore, the UELs of binary fuel mixtures of hydrogen with methane, ethylene, and propane in air were determined experimentally at elevated temperatures by Wierzba and Ale.[52] It was shown then that the experimental limits of hydrogen−methane mixtures deviate slightly from those calculated using Le Chatelier's rule and the UELs of hydrogen−ethylene mixtures deviate significantly from those calculated using Le Chatelier's rule over the range of temperatures tested and at a residence time of 10 min. It was suggested that the narrowing of the UEL is due to surface reactions on the stainless steel wall during the waiting time that tends to change the mixture composition just prior to spark ignition. The explosive limits of these mixtures exposed to longer residence times do not obey Le Chatelier's rule. Hence, the explosive limits of CH_4/CO, CH_4/C_2H_6, or CH_4/H_2 are measured. The impacts of CO, C_2H_6, and H_2 on the explosive limits of methane are presented in Table 2. The experimental data show that the addition of CO, C_2H_6, and H_2 tends to reduce the LEL of CH_4 in varying degrees. This is especially the case for C_2H_6: when the added C_2H_6 reaches 2.0%, the LEL of CH_4 significantly reduces from 5.05 to 2.25%. Adding CO and C_2H_6 decreases the UEL of CH_4, whereas adding H_2 increases the UEL of CH_4 first before it reduces it.

Tables 3−5 and Figures 1−3 illustrate the impact of CO, C_2H_6, and H_2 on P_{max} for different CH_4 concentrations, whereas the results on $\mathrm{Max}_{P_{max}}$ and C_m are shown in Table 6. It should be noted that C_m of pure CH_4 is ∼10%. After adding CO, C_2H_6, or H_2, the C_m value of CH_4 moves toward LEL at different levels with an increased amount of the other gas added; this effect is particularly pronounced for C_2H_6. With the same amount of the other gas added, the impact on C_m of binary-component mixtures is as follows: $C_m^{H_2} \approx C_m^{CO} > C_m^{C_2H_6}$. The P_{max} value of binary mixtures (CH_4/CO, CH_4/C_2H_6, CH_4/H_2) is higher than that of pure CH_4 when the concentration of CH_4 is between LEL and C_m; on the other hand, when the CH_4 concentration is between C_m and UEL, P_{max} of binary mixtures is lower than that of pure CH_4.

Furthermore, after adding CO, C_2H_6, or H_2, the $\mathrm{Max}_{P_{max}}$ value of the binary-component mixtures is higher than that of pure CH_4. Increasing the amount of the added CO, C_2H_6, or H_2 leads to a more significant rise of $\mathrm{Max}_{P_{max}}$. With the same amount added, the impact on $\mathrm{Max}_{P_{max}}$ of binary-component mixtures is as follows: $\mathrm{Max}_{P_{max}}^{H_2} > \mathrm{Max}_{P_{max}}^{CO} > \mathrm{Max}_{P_{max}}^{C_2H_6}$. In summary, the explosion intensity and destructive power of binary-component mixtures (CH_4/CO, CH_4/C_2H_6, and CH_4/H_2) are significantly higher than that of pure CH_4.

Normally, the explosions in low gaseous mines belong to oxygen-rich explosion, and 5.0% has been viewed as the critical value for CH_4 concentration as it is approximately the LEL. However, our experimental results show that the LEL of CH_4 will be significantly reduced in the presence of CO, C_2H_6, or H_2, exposing the increasing risk even at a low CH_4 concentration. Meanwhile, the most dangerous concentration (C_m) for CH_4, producing the highest explosion overpressure, also reduces in the presence of CO, C_2H_6, or H_2. Therefore, the effects of CO, C_2H_6, or H_2 on the explosive limits and C_m of CH_4 must be fully considered, and the alarm threshold of CH_4 needs to be lowered accordingly in coal mine gas monitoring and early alarm system.

2.4. Impact of CO/C_2H_6/H_2 Mixtures on Methane Explosion Characteristics. Usually CO, C_2H_6, H_2, and CH_4 coexist in coal mines. To gain further understanding of the explosion behavior of multicomponent flammable gases, CO, C_2H_6, and H_2 were added to CH_4/air mixtures with four different ratios, as given in Table 7. The results are shown in Tables 7 and 8 and Figure 4.

When CO, C_2H_6, and H_2 were added together to CH_4/air mixtures, both the LEL and UEL of CH_4 decreased. Note that the C_m value of pure CH_4 is approximately 10%; this C_m of CH_4 shifts toward the LEL with the increase of the CO/C_2H_6/H_2 amount. With regard to the maximum explosion pressure, when the CH_4 concentration is between LEL and 9.0%, P_{max} of the CH_4/CO/C_2H_6/H_2 mixtures is higher than that of pure CH_4, whereas for the CH_4 concentration between 11.0% and UEL, P_{max} of the CH_4/CO/C_2H_6/H_2 mixtures is lower than that of pure CH_4. With the increase of the added CO/C_2H_6/

Table 3. Impact of CO Addition on P_{max} for Different Concentrations of CH_4

	concentration of CH_4/%								
	P_{max} of different concentrations of CH_4/CO/MPa								
concentration of CO/%	5	6	7	8	9	10	11	12	13
0		0.510	0.605	0.680	0.720	0.778	0.738	0.710	0.632
0.5		0.525	0.627	0.684	0.750	0.795	0.735	0.660	0.604
1.0	0.485	0.542	0.701	0.761	0.808	0.822	0.715	0.633	0.542
2.0	0.574	0.612	0.731	0.785	0.845	0.800	0.689	0.563	0.500
3.0	0.593	0.620	0.780	0.810	0.856	0.797	0.666	0.531	0.427

Table 4. Impact of C_2H_6 Addition on P_{max} for Different Concentrations of CH_4

	concentration of CH_4/%								
	P_{max} of different concentrations of CH_4/C_2H_6/MPa								
concentration of C_2H_6/%	5	6	7	8	9	10	11	12	13
0		0.510	0.605	0.680	0.720	0.778	0.738	0.710	0.632
0.5	0.486	0.613	0.666	0.753	0.791	0.748	0.694	0.635	0.533
1.0	0.609	0.670	0.731	0.805	0.769	0.731	0.676	0.553	0.424
1.5	0.639	0.705	0.769	0.812	0.778	0.688	0.658	0.347	
2.0	0.758	0.791	0.830	0.784	0.753	0.669	0.624	0.151	

Table 5. Impact of H_2 Addition on P_{max} of Different Concentrations of CH_4

	concentration of CH_4/%								
	P_{max} of different concentrations of CH_4/H_2/MPa								
concentration of H_2/%	5	6	7	8	9	10	11	12	13
0		0.510	0.605	0.680	0.720	0.778	0.738	0.710	0.632
0.5	0.427	0.543	0.628	0.693	0.760	0.803	0.783	0.745	0.629
1.0	0.502	0.585	0.664	0.771	0.794	0.826	0.761	0.724	0.618
1.5	0.567	0.599	0.743	0.778	0.796	0.839	0.743	0.717	0.604
2.0	0.600	0.621	0.776	0.791	0.884	0.806	0.718	0.668	0.597

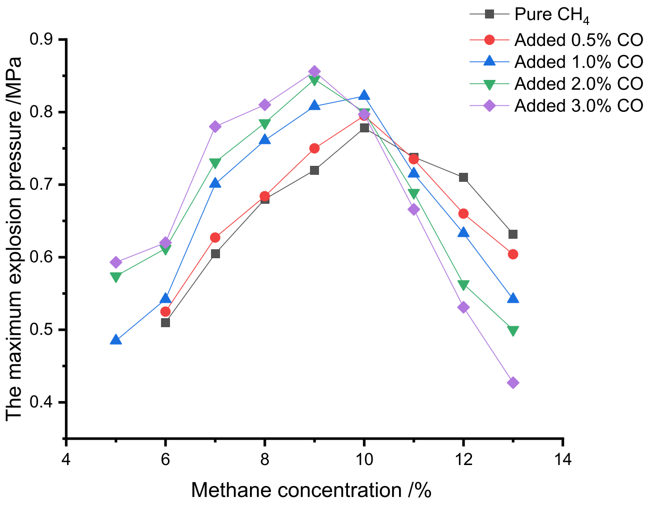

Figure 1. Impact of CO addition on P_{max} for different concentrations of CH_4.

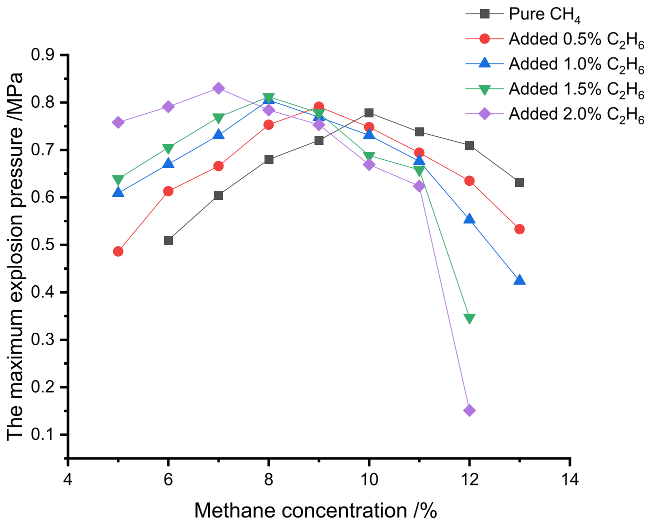

Figure 2. Impact of C_2H_6 addition on P_{max} for different concentrations of CH_4.

H_2, the experimental results also indicate a more significant rise in $\mathrm{Max}_{P_{max}}$ of the $CH_4/CO/C_2H_6/H_2$ mixtures.

The reaction mechanism of multicomponent flammable gases is complicated in coal mines. Other flammable gases (CO, C_2H_6, and H_2) with a slight change in concentration could make a significant impact on the explosion characteristics of CH_4. Moreover, if a gas explosion occurs, a certain amount of H_2, CO, and other flammable gases may be produced. This is likely to trigger the second explosion which would be more dangerous than the first one.[53] Therefore, the likelihood and risk of the second gas explosion should be fully assessed in the emergency rescue system, in particular, with regard to a small change of other flammable gases.

Furthermore, the explosion characteristic parameters of multicomponent flammable gases such as explosive limits, maximum explosion pressure, and C_m may not be obtained by

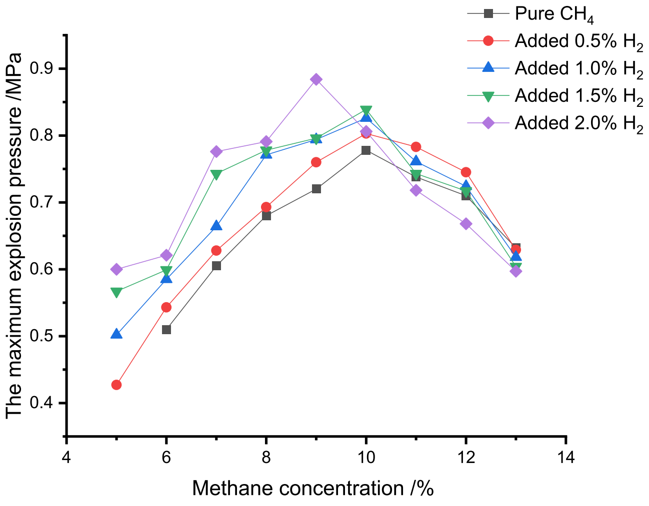

Figure 3. Impact of H_2 addition on P_{max} for different concentrations of CH_4.

simply superimposing the values from single- or binary-component flammable gases.

2.5. Theoretical Analysis of the Impact of CO, C_2H_6, and H_2 on the Explosion Characteristics of CH_4. From the perspective of the chemical reaction kinetics, we proposed a self-promoting microcirculation reaction network of the multicomponent flammable gases (Figure 5)

$$H_2 + O_2 \rightarrow HO_2 + H^{\bullet} \tag{1}$$

$$H_2 + M \rightarrow H^{\bullet} + H^{\bullet} + M \tag{2}$$

$$H^{\bullet} + O_2 \rightarrow O^{\bullet} + OH^{\bullet} \tag{3}$$

$$O^{\bullet} + H_2 \rightarrow H^{\bullet} + OH^{\bullet} \tag{4}$$

$$CH_4 + H^{\bullet} \rightarrow CH_3^{\bullet} + H_2 \tag{5}$$

$$CH_4 + OH^{\bullet} \rightarrow CH_3^{\bullet} + H_2O \tag{6}$$

$$CH_4 + O^{\bullet} \rightarrow CH_3^{\bullet} + OH^{\bullet} \tag{7}$$

$$H_2O + H^{\bullet} \rightarrow OH^{\bullet} + H_2 \tag{8}$$

$$CO + OH^{\bullet} \rightarrow CO_2 + H^{\bullet} \tag{9}$$

$$C_2H_6 + H^{\bullet} \rightarrow C_2H_5^{\bullet} + H_2 \tag{10}$$

$$C_2H_6 + OH^{\bullet} \rightarrow C_2H_5^{\bullet} + H_2O \tag{11}$$

Table 7. Impact of $CO/C_2H_6/H_2$ Added Simultaneously on the Explosive Limits of CH_4

				explosive limits of CH_4	
no.	CO/%	C_2H_6/%	H_2%	LEL/%	UEL/%
1	0	0	0	5.05	15.35
2	0.8	0.3	0.1	4.25	14.05
3	1.0	0.5	0.2	3.85	13.45
4	1.5	0.8	0.5	3.25	13.15
5	2.0	1.0	0.5	2.65	12.95

Reaction 1 would be the primary chain initiation reaction in the initial reaction period, as reaction 1 is easier to be triggered than reaction 2 according to the reaction activation energy.[54] With the reaction progressing, the temperature rises, which would make reaction 2 the main chain initiation reaction. Reactions 3 and 4 are the main branching chain reactions, and reactions 5−11 are the main elementary reactions in the multicomponent flammable gas reaction system. Either reaction 1 or 2 provides $H^{\bullet}$ radicals that develop a radical pool of $OH^{\bullet}$, $O^{\bullet}$, and $H^{\bullet}$ by the chain reactions 3 and 4. The reactions 1−4 are of great importance in the oxidation reaction mechanisms of hydrocarbon in that they provide the essential chain-branching and propagating steps as well as the radical pool for fast reactions. Moreover, the reactions 5−11 will produce new H_2, $H^{\bullet}$, and $OH^{\bullet}$ radicals which further accelerate the rates of reactions 1−11. Thus, as illustrated in Figure 5, the above reactions may lead to a self-promoting microcirculation system and a positive feedback mechanism. With the progress of the reaction, the reaction rate, the heat release, and the pressure will increase constantly until the explosion pressure reaches the maximum value. It is important to realize that any high-temperature hydrocarbon mechanism involves H_2 and CO oxidation kinetics and that most, if not all, of CO_2 that is formed results from reaction 9. However, experimental evidence indicates that the oxidation of CO to CO_2 comes late in the reaction scheme[7] because reaction 9 is slower than the reaction 6 or 11. Hence, the chain initiation of H_2 can produce highly active $H^{\bullet}$ and $OH^{\bullet}$ radicals, and CO may be mainly involved in the later reaction of CH_4, which will further accelerate the reaction speed of the main reactant CH_4. In reaction 2, M is the usual third body. CO may participate in molecular collisions as the usual third body to help produce $H^{\bullet}$ radicals. Therefore, CO may increase the collision frequency and make the chain initiation reaction of $CH_4/CO/C_2H_6/H_2/$air mixtures easier than CH_4/air mixtures. It is worth to note that the chain initiating reaction of methane/air mixtures is difficult and slow. However, in the presence of OH, O, and H

Table 6. Comparison of the Impact of CO, C_2H_6, and H_2 Added Respectively on $\text{Max}_{P_{\text{max}}}$ and C_m of the Gas Mixtures

added concentration/%	added flammable gas	$\text{Max}_{P_{\text{max}}}$ of gas mixtures/MPa	C_m of the gas mixtures/%	concentration of CH_4/%
0.0		0.778	10.0	10.0
0.5	C_2H_6	0.791	9.5	9.0
	CO	0.795	10.5	10.0
	H_2	0.803	10.5	10.0
1.0	C_2H_6	0.805	9.0	8.0
	CO	0.822	11.0	10.0
	H_2	0.826	11.0	10.0
2.0	C_2H_6	0.830	9.0	7.0
	CO	0.845	11.0	9.0
	H_2	0.884	11.0	9.0

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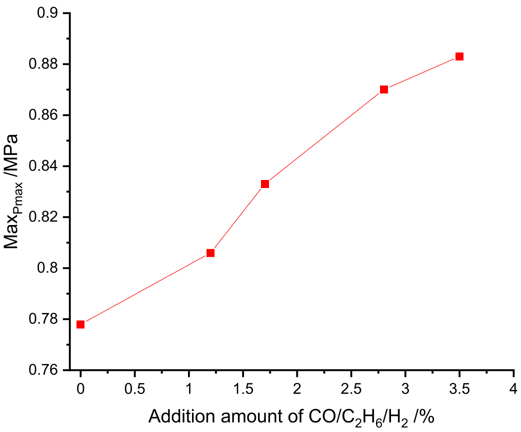
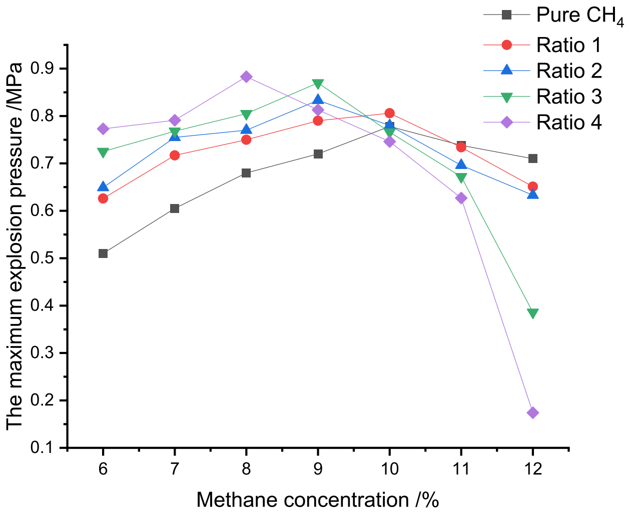
Table 8. Impact of $CO/C_2H_6/H_2$ Added Simultaneously on P_{max} for Different Concentrations of CH_4

				concentration of CH_4/%						
				P_{max} of different concentrations of $CH_4/C_2H_6/CO/H_2$ mixtures/MPa						
no.	CO/%	C_2H_6/%	H_2/%	6	7	8	9	10	11	12
1	0	0	0	0.510	0.605	0.680	0.720	0.778	0.738	0.710
2	0.8	0.3	0.1	0.626	0.717	0.750	0.790	0.806	0.734	0.651
3	1.0	0.5	0.2	0.649	0.755	0.770	0.833	0.780	0.696	0.633
4	1.5	0.8	0.5	0.725	0.768	0.805	0.870	0.766	0.672	0.386
5	2.0	1.0	0.5	0.773	0.791	0.883	0.813	0.746	0.627	0.174



(a) Impact on $Max_{P_{max}}$



(b) Impact on P_{max}

Figure 4. Impact of $CO/C_2H_6/H_2$ addition on P_{max} and $Max_{P_{max}}$ of $CH_4/CO/C_2H_6/H_2$ mixtures.

radicals, the reactions 5−7 that involved methane are all fast. Furthermore, the positive feedback mechanism of the self-promoting microcirculation may make the reaction rate of $CH_4/CO/C_2H_6/H_2$/air mixtures faster than CH_4/air mixtures, resulting in more heat release and higher explosion pressure. C_2H_6 oxidizes much more slower than hydrogen, and very small quantities of hydrogen will increase the rate of CO oxidation substantially.[7] Therefore, the influence of C_2H_6 on the explosion process is likely smaller than that of CO and H_2. Chain initiation and chain branching reactions initiated by H_2 in multicomponent flammable gas mixtures are easier and faster than that in the methane/air mixture. CO and C_2H_6 also may accelerate the chain initiation reaction as the third body. Thus, the rate of CH_4 oxidation is substantially faster than that of the pure methane reaction system. The above theoretical analysis gives further support to the observations in the experiments that binary- (CH_4/CO, CH_4/C_2H_6, and CH_4/H_2) and multicomponent ($CH_4/CO/C_2H_6/H_2$) mixtures are more dangerous, and the resulting explosion is more destructive than that by pure CH_4.

3. CONCLUSIONS

Three representative gases, CO, C_2H_6, and H_2, were selected to investigate the impact of their presence on CH_4 explosion characteristics. The explosion strength and explosion destructive power are higher for binary- (CH_4/CO, CH_4/C_2H_6, and CH_4/H_2) and multicomponent mixtures ($CH_4/CO/C_2H_6/H_2$) than for pure CH_4 by measuring the explosion pressure P_{max} and $Max_{P_{max}}$. Because of the decrease of the LEL and C_m of CH_4 in the presence of CO, C_2H_6, and H_2, the impact of other flammable gases on the explosion characteristics of CH_4 must be fully considered, and the alarm threshold of CH_4 needs to be lowered accordingly in coal mine gas monitoring and early alarm system. Meanwhile, other flammable gases (CO, C_2H_6, and H_2) with a slight change in concentration could make a significant impact on the explosion characteristics of CH_4. Experimental results indicate that the characteristic explosion parameters of multicomponent flammable gases such as explosive limits, maximum explosion pressure, and C_m may not be obtained by simply superimposing the values from single- or binary-component flammable gases. Experiment is still the primary way to obtain these parameters. The experimental data will also potentially provide guidance for the further study of the reaction mechanism of multicomponent gas explosion. Furthermore, a self-promoting microcirculation reaction network of the multicomponent flammable gases ($CH_4/CO/C_2H_6/H_2$) was proposed, combining the theory analysis with experimental data. This reaction network reflects the impact of CO, C_2H_6, and H_2 on the explosion characteristics of CH_4 and aids to reasonably infer the explosion reaction mechanism of multicomponent flammable gases.

For multicomponent flammable gases, the dynamics of the reaction and the interactions between components can become quite complex. The investigation on the explosion microscopic reaction mechanism of multicomponent flammable gases and the influence of temperature, pressure, ignition energy, and turbulence on multicomponent flammable gas characteristics will be conducted in future work. Furthermore, the scale of the experiments is comparatively small in relation to large industrial scales, and advanced computational tools combined with experiments should indeed be welcomed.

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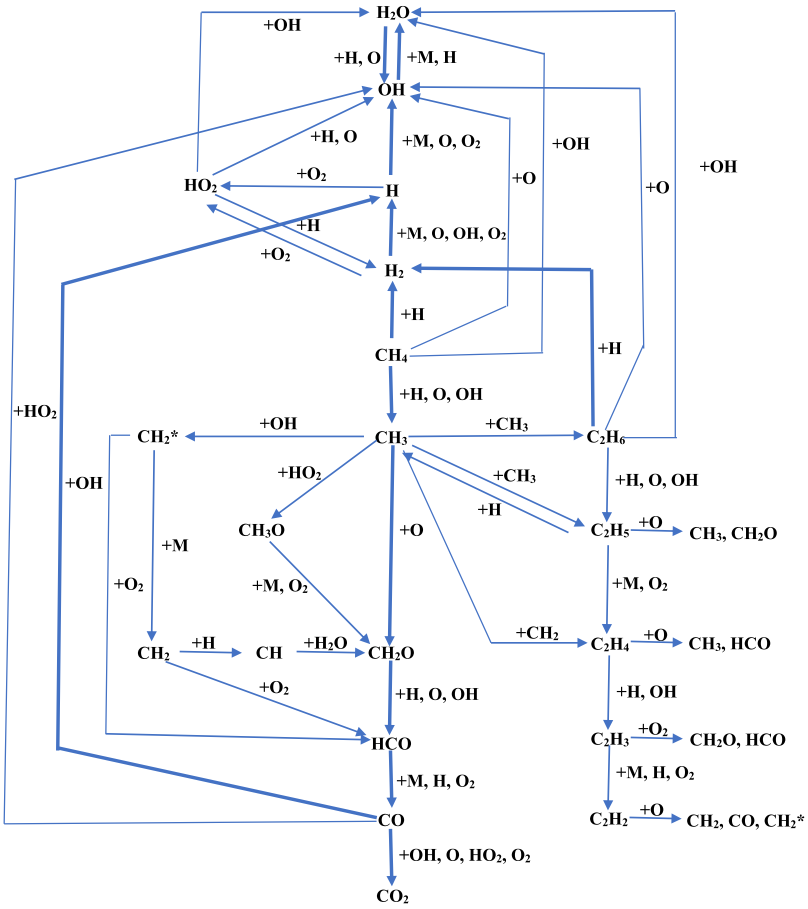

Figure 5. Self-promoting microcirculation reaction network of the multicomponent flammable gases.

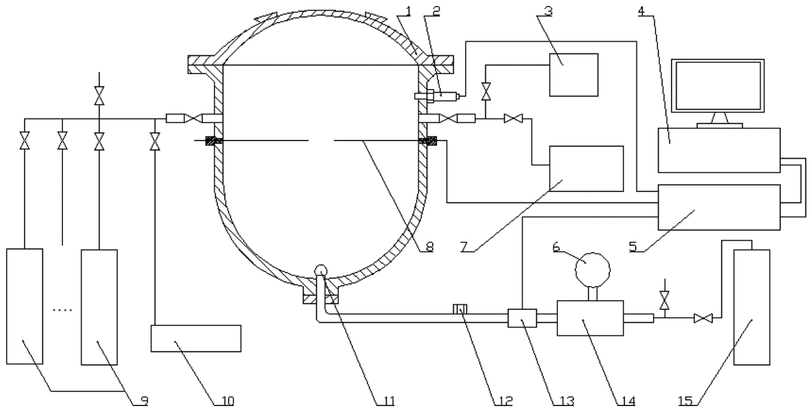

Figure 6. Schematic diagram of the experimental setup. 1 Explosion reactor; 2 pressure transducer; 3 air compressor; 4 computer; 5 controller; 6 manometer; 7 vacuum pump; 8 ignition electrode; 9 gas steel bottle group; 10 precision digital pressure gauge; 11 dispersion nozzle; 12 powder addition mouth; 13 electromagnetic valve; 14 compressed air storage room; 15 compressed air steel bottle.

4. EXPERIMENTAL METHODS

Experiments were performed in a 20 L spherical closed vessel which consists of an explosion vessel, a gas distribution system, an ignition system, and a measurement system, as shown in Figure 6. The explosion vessel (designed and produced by the Chongqing Branch of China Coal Research Institute, China), which can withstand a maximum pressure of 3.0 MPa, is made of stainless steel and is nearly spherical. The approximate dimensions are 34 cm in height, 30 cm in diameter, and 19,900 cm^3 in effective volume. The gas distribution system is composed of bottles of pure CH_4, CO, C_2H_6, and H_2, an air compressor, a vacuum pump, and a pressure gauge. The flammable gases used in this experiment were provided by the Shanghai Pujing Gas company. The purity of each flammable gas was higher than 99.99%. The partial pressure method was used for mixture preparation, with a high-accuracy sensitive pressure transducer. The ignition source for the experimental setup was a detonating pyrotechnic ignition device (supplied by Liuyang Wenchi Electric Ignition Co., China) with a calorimetric energy of 5 J. The ignition position is in the center of the vessel. For the measurement of the static pressure, an NTS-2A precise digital pressure gauge (produced by NTS Co., Japan) was used. The measurement of the dynamic explosion pressure was achieved using a CY-DB 1303-type pressure sensor (produced by Baoji Huarui Sensor Institute, China) and a multifunction explosive reaction controller (produced by the Chongqing Branch of China Coal Research Institute).

The explosion characteristics were determined at ambient temperature and pressure. The electric igniter was placed at the center of the reactor, and the explosion vessel was evacuated and purged with fresh air three times. Then, the required mixture of flammable gases and air was injected into the vessel using the partial pressure method, waiting for at least 5 min to allow the gas to fully mix in the reactor. Afterward, the igniter was ignited by the ignition controller, and the pressure data were recorded and saved into the computer. Both the data acquisition instrument and ignition controller were connected with a synchronizer trigger to ensure the synchronization of ignition and data acquisition. A minimum of three experiments were performed for each initial condition of the flammable mixtures. The maximum explosion pressure listed in the tables is the maximum value among the three experiments.

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Notes

The authors declare no competing financial interest.

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