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High methane natural gas/air explosion characteristics in confined vessel

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HIGHLIGHTS

- The effect of methane fraction on pressure evolution during natural gas combustion is examined.
- Increasing pressure will increase the explosion pressure and the maximum rate of pressure rise.
- Increasing pressure will decrease the combustion duration.
- An empirical correlation for the combustion phasing is proposed.

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ABSTRACT

The explosion characteristics of high methane fraction natural gas were investigated in a constant volume combustion vessel at different initial conditions. Results show that with the increase of initial pressure, the peak explosion pressure, the maximum rate of pressure rise increase due to a higher amount (mass) of flammable mixture, which delivers an increased amount of heat. The increased total flame duration and flame development time result as a consequence of the higher amount of flammable mixture. With the increase of the initial temperature, the peak explosion pressures decrease, but the pressure increase during combustion is accelerated, which indicates a faster flame speed and heat release rate. The maximum value of the explosion pressure, the maximum rate of pressure rise, the minimum total combustion duration and the minimum flame development time is observed when the equivalence ratio of the mixture is 1.1. Additionally, for higher methane fraction natural gas, the explosion pressure and the maximum rate of pressure rise are slightly decreased, while the combustion duration is postponed. The combustion phasing is empirically correlated with the experimental parameters with good fitting performance. Furthermore, the addition of dilute gas significantly reduces the explosion pressure, the maximum rate of pressure rise and postpones the flame development and this flame retarding effect of carbon dioxide is stronger than that of nitrogen.

1. Introduction

Depletion of fossil fuels and the urgent requirement of reducing environmental pollutions caused by combustion have recently driven the studies on the combustion characteristics of alternative fuels. Natural gas is thought to be one of the best alternative fuels and it has been widely used in city bus and taxi engines in China. Natural gas is a mixture of light hydrocarbons such as methane, ethane, propane, etc. Components of natural gas from different production sites may vary but the primary component is methane [1]. There are several advantages for the application of natural gas in the internal combustion engines; engines can operate under high compression ratio because of high fuel octane number [2]. Natural gas is the clean energy and has the lowest C/H ratio among all the hydrocarbons thus allowing the reduction emission of CO_2 [2–5]. Moreover, most of the technology developed for the internal combustion engines can be easily applied to burn natural gas as engine fuel.

Previously, extensive investigations have been carried out to evaluate the fundamental combustion characteristics of natural gas. Gu et al. [6] and Liao et al. [7] measured the laminar burning velocity of methane and natural gas mixed with air at different

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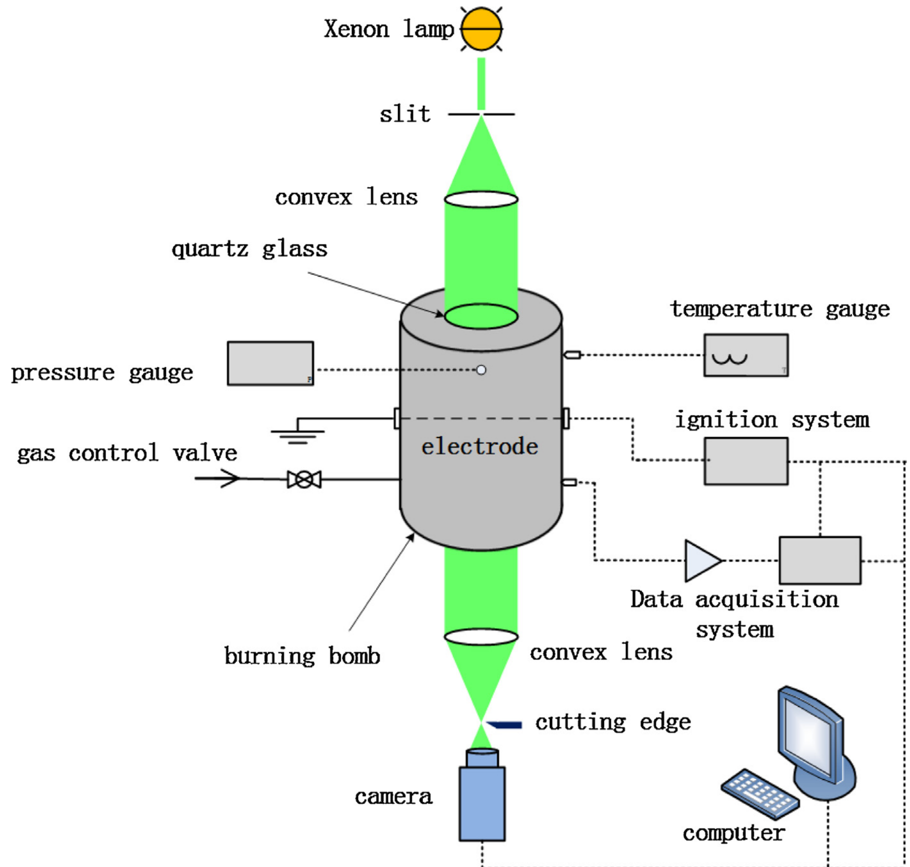

Fig. 1. Experimental setup.

equivalence ratios and investigated the flame instability at high pressures. Huang et al. [8] and Hu et al. [9] studied the laminar burning characteristics of hydrogen enriched natural gas. They obtained the stretched flame propagation speed and the burning velocity. Cho et al. [10] reviewed the spark ignition natural gas engines. They highlighted some indexes such as emissions, combustion efficiency, and strategies to get stable combustion of the natural gas engines. Slavinskaya et al. [11] developed a kinetic mechanism to predict the emission of methane–ethane flame, especially the poly-aromatic hydrocarbons (PAH). However, there are some problems of the application of natural gas. One of the problems for utilizing natural gas is the disasters about combustion such as ignition and explosion [12–14]. The dominant damages of the fire resulting by leakage are the heat release from the sustained fire and collapse of buildings because of the explosion [15]. But there has been little study focusing on the explosion characteristics of natural gas. Another problem is about purification. Purification is difficult and expensive. But the research about the influence of other non-methane gases such as ethane is very little.

The explosion pressure (P_{max}), the rate of pressure rise (dP/dt), and the time when P_{max} reaches (t_R) (also defined as the combustion duration) [16] are the most important parameters of explosion in appraising the risk of the application of natural gas [17,18]. They are also important for the transportation and storage of natural gas. The parameters such as P_{max}, dP/dt and t_R are very sensitive to initial pressure, initial temperature, equivalence ratio, and the fuel compositions. The dilute gas also has significant influence on those parameters because it reduces the flame temperature due to increased average mixture specific heat. Theoretically, constant volume adiabatic equilibrium pressure (P_e) for given mixture composition is the maximum pressure that the system can thermodynamically achieve [19]. However, the explosion pressure (P_{max}) measured in confined vessel is expected to be significantly lower than the adiabatic equilibrium pressure (P_e). The main reason is the heat loss resulted by the thermal conduction, convection and radiation [19]. Razus et al. [20] have observed the influence of initial temperature and pressure on the explosion characteristics in a spherical vessel. They found that the $(dP/dt)_{max}$ and the deflagration indices were linear functions of initial pressure, when the initial temperature and the fuel/oxygen were constant. And when the initial pressure and the composition were constant, the $(dP/dt)_{max}$ and the deflagration indices were influenced by the initial temperature.

The objectives of this work are the following. Firstly, the influence of the initial conditions such as pressures, temperatures, equivalence ratios, dilution ratios and different types of dilute gases on the explosion characteristics of high methane natural gas including the explosion pressure, the pressure rise rate, the combustion duration will be investigated. Additionally, some natural gas companies in China are trying to produce high methane concentration natural gas (up to 99.9% in volume) because of high octane number of methane, though this purification process is extremely expensive. Thus our second objective is to see to what extend the explosion pressures are affected by the variation of methane concentration. Finally, the peak pressure and the combustion phasing of these high methane concentration natural gas mixtures will be studied so as to provide some fundamental data for natural gas engine timing control.

2. Experimental setup and procedures

Fig. 1 is the sketch of the experimental system. It includes the constant volume combustion vessel, the ignition system, the heating system and the data acquisition system. The diameter of the stainless steel cylinder combustion vessel is 180 mm and the length is 210 mm. The combustion vessel is wrapped by heating tapes so as to control the temperature of the mixture in the vessel. Temperatures of the gas in the vessel can be monitored by a thermocouple fixed in the inner wall of the vessel. Two quartz windows are mounted at the both sides of the cylinder for optical access. The gas is filled into the vessel by accurately controlling the inlet valve, so that every gas component is sequentially added into the vessel to its required partial pressure at the specific equivalence ratio. The prepared combustible mixture is ignited by the centrally installed pair of electrodes whose spark energy (45 mJ) is higher than the minimum ignition energy of all the mixtures so as to ensure the flame propagation is successfully initiated. The flame propagation is also captured by the high speed camera. The dynamic pressure during combustion is then acquired by a pressure transducer (Kistler 7001) at a sample rate of 100 kHz, combined with a Charge Amplifier (Kistler 5011). Once the combustion was finished, the combustion vessel was vacuumed and flushed with dry air at least three times to avoid the influence of the residual gas on the next combustion. A time interval of 5 min was awaited to make sure the mixtures are motionless. An interval time of 30 min was tested for several initial conditions, and there is no noticeable difference. Additionally, the reproducibility of the experiment has also been tested and there is no significant difference in the pressure history.

The purities of each component in the study are all higher than 99.99%. The natural gas used in this experiment is provided by the Shanghai Weichuang Standard Gas Company who has the China Metrology Certification, and it only contains main components are methane and ethane, the fractions of other components is less than

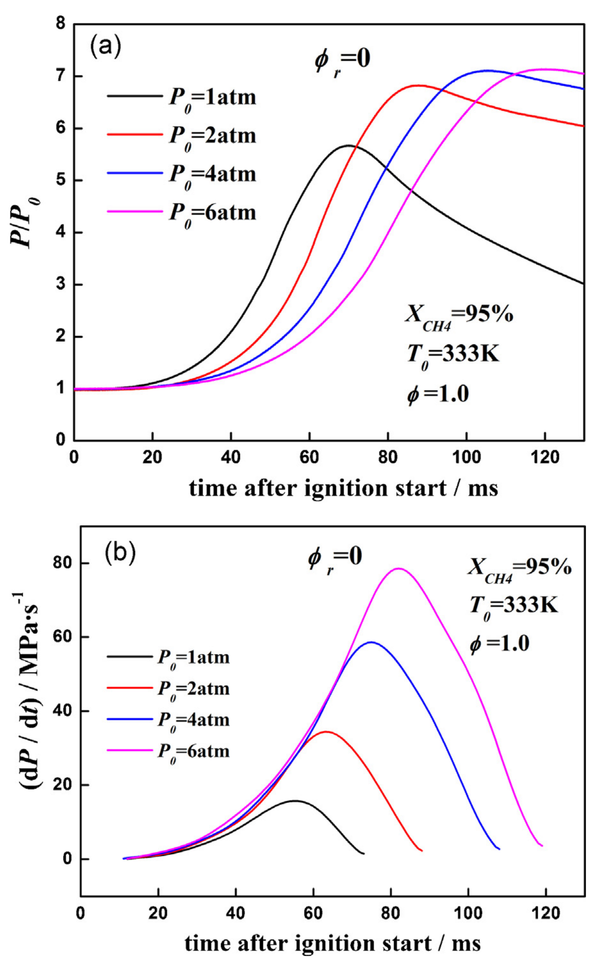

Fig. 2. Pressure history and the rate of pressure rise for different initial pressures.

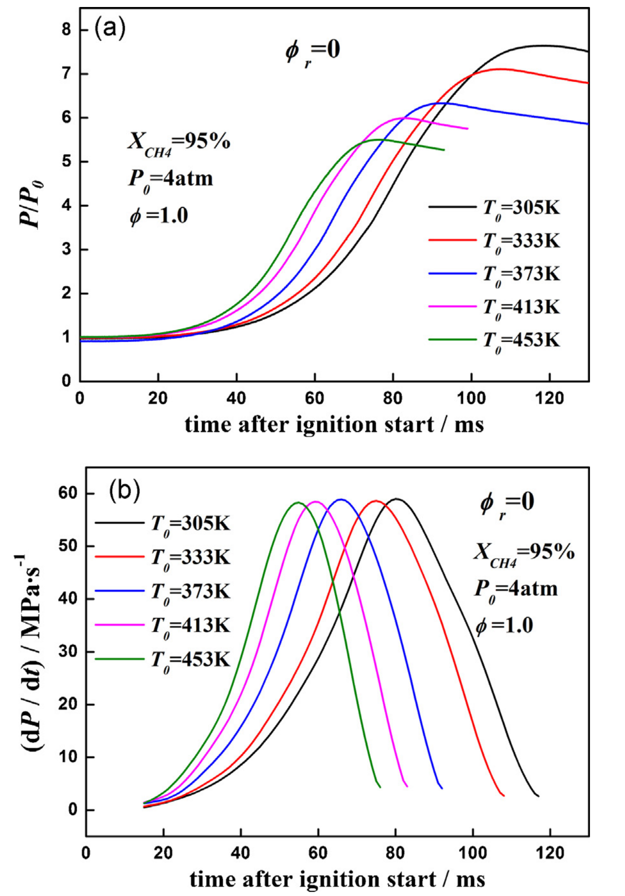

Fig. 3. Pressure history and the rate of pressure rise for different initial temperatures.

0.01%. The dilution ratio is defined as the volumetric fraction of diluents (N_2/CO_2) addition in the pre-mixture:

$$\phi_r = \frac{V_{diluent}}{V_{fuel} + V_{air} + V_{diluent}} \quad (1)$$

In this experiment, the initial temperatures are chosen as 305 K, 333 K, 373 K, 413 K and 453 K. The error of the temperature is ±3 K. The initial pressures are set as 1 atm, 2 atm, 4 atm and 6 atm. The equivalence ratio ranges from 0.7 to 1.4. The dilution ratios are 5%, 10% and 20%. The methane fraction is defined as the volumetric fraction of methane in the natural gas. In this work, the methane fractions are from 95% to 100%. The dilute gas is the mixture of N_2 and CO_2 and the proportion is 0/100, 85/100 and 100/0.

Combustion pressure history data was recorded by an oscilloscope. The combustion duration is defined as the time interval between the start of ignition and the instant when the maximum value of pressure is obtained. The flame development time is defined as the time starting from ignition to the moment that the pressure increases by 7%, same as the definition in Ref. [21].

3. Results and discussions

Experimental observations show that all the recorded pressure–time curves exhibit similar behavior: after the mixture is ignited, the pressure increases gradually to the moment that the pressure rise rate reaches to its maximum value (as shown in Fig. 2(b)), then the pressure continues to increase though the pressure rise rate is decreased, and after the maximum pressure is obtained, (as shown in Fig. 2(a)) the pressure begins to decrease because of the end of combustion and the heat loss.

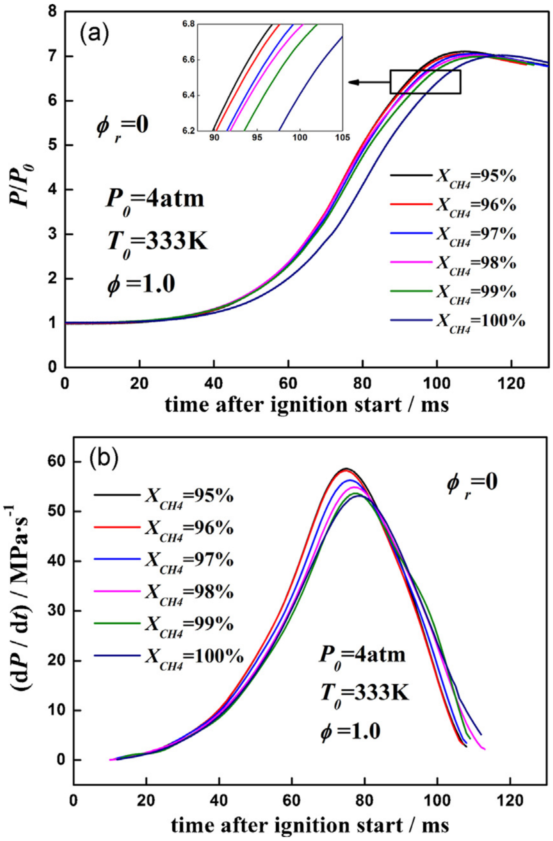

Fig. 4. Pressure history and the rate of pressure rise for different methane fractions.

3.1. Parameter dependence of the pressure evolution

Figs. 2–7 give the pressure evolution and the pressure rise rate during combustion process. The pressure is normalized with respect to the initial pressure (P_0). Fig. 2(a) gives the pressure–time curves at different initial pressure when the initial temperature is 333 K, the methane fraction is 95%, and the equivalence ratio is 1.0. Fig. 2(b) shows the pressure rise rate corresponding to the pressure history. The explosion pressure (P_{max}) is increased significantly with the increase of P_0. However, the maximum value of normalized pressure is not sensitive to the initial pressure. Because the flame propagation speed is decreased with the increase of P_0 [22], which is evidenced by the flame propagation video, the time that the pressure reaches to the maximum value (t_R) increases with the increase of P_0, so as to the time of the maximum value of pressure rise rate. The maximum value of pressure rise rate increases obviously with the increase of the initial pressure. This pressure evolution dependence on the initial pressure implies that for practical engines, introduction of higher compression ratio significantly increases the engine power output.

Fig. 3 illustrates the influence of initial temperature on the pressure history and the pressure rise rate when the initial pressure is 4 atm, the methane fraction is 95%, and the equivalence ratio is 1.0. Fig. 3(a) shows that the maximum value of pressure decreases significantly with the increase of the initial temperature. The similar behavior also happens to the time that the pressure reaches to its maximum value and the time of the maximum value of pressure rise rate. This is because the adiabatic temperature increases with the increase of the initial temperature, leading to the increase of the flame propagation velocity. Moreover, the P_{max} is decreased with the increase of T_0 due to the decreased mass of the charge and

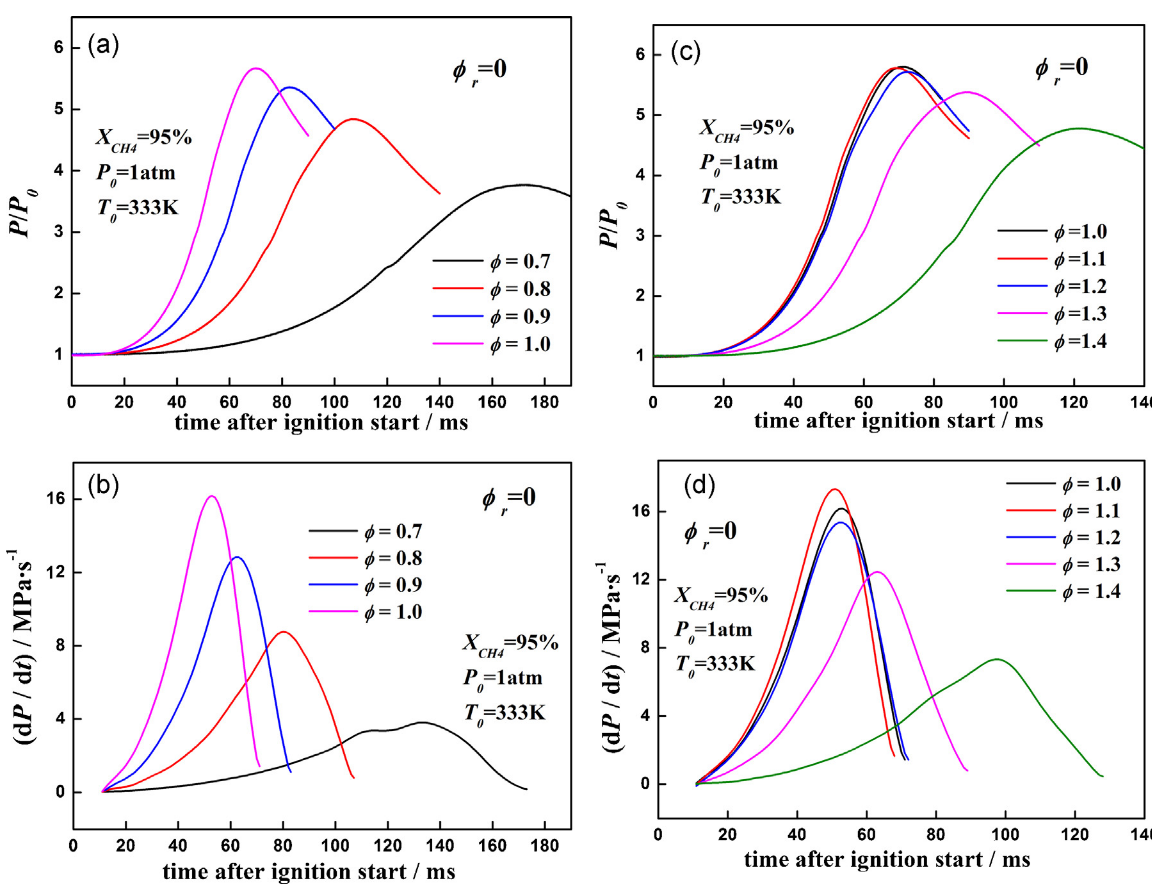

Fig. 5. Pressure history and the rate of pressure rise for different equivalence ratios.

hence the heat release. Fig. 3(b) shows the maximum values of the pressure rise rate are almost the same with the alternation of initial temperature. This illustrates T_0 has little influence on the pressure rise rate (dP/dt).

Fig. 4 shows the influence of methane fraction on the pressure evolution and the pressure rise rate for $P_0 = 4$ atm, $T_0 = 333$ K, and the equivalence ratio of 1.0. Fig. 4(a) shows that with the increase of methane fraction, both the P_{max} and the maximum pressure rise rate, $(dP/dt)_{max}$, are slightly decreased. Additionally, it is clearly seem that the increase of methane fraction slightly postpones the pressure evolution, as seen by the inset of Fig. 4(a), time that P_{max} and $(dP/dt)_{max}$ appears is slightly postponed. Because the molecular heating value of ethane is slightly higher than methane, with the increase of methane fraction in the mixture, the total heating value of the fuel is decreased, resulting in a slightly decrease of P_{max} and $(dP/dt)_{max}$. Additionally, because the flame speed of ethane is slightly higher than that of methane [23]. It is expected that the fuel mixture with higher methane fraction burns less slower, resulting in the observed postponed pressure evolution.

Fig. 5 gives the pressure–time curves and the pressure rise rate–time curves at different equivalence ratios when the initial pressure is 1 atm, the initial temperature is 333 K, the methane fraction is 95%. It is seen that for very lean mixtures ($\phi = 0.7$), there is significantly excess oxidizer provided in the mixture, pressure increase during combustion is very slow and the peak pressure is around 3.5 times of the initial pressure. As the equivalence ratio increases, the heating value of the fuel mixture is increased due to the increase of fuel concentration, and the combustion becomes more robust, the combustion pressure increases faster and P_{max} is also increased. At the equivalence ratio of 1–1.1, the combustion is the most complete and P_{max} has the highest value. As the equivalence ratio is further increased, there are excess fuel provided and the combustion is incomplete, the pressure increase is slowed down.

Fig. 6 shows the effect of the ratio of dilution ratio ϕ_r on the explosion pressure and the pressure rise rate when the initial pressure is 1 atm, the initial temperature is 423 K, the methane fraction is 95%, the equivalence ratio is 1.0 and the dilution gas is N_2(85%)/CO_2(15%). With the increase of the dilution ratio, the maximum value of pressure decreases significantly and the pressure rise ratio becomes lower with the increase of the dilution ratio. Additionally, maximum pressure rise rate also decreases significantly. The decrease of the pressure is caused by the reduced total heat value of the mixture due to the decreased concentration of the reactants. Additionally, the adiabatic flame temperature decreases with the increase of the dilution ratio, which also contributes to the decreased maximum pressure. The decrease of the maximum pressure rise can still be explained by the fact that the flame speed of the mixture decreases with the increase of the dilution ratio. The pressure evolution dependence on the dilution ratio implies that for practical engines, the introduction of the exhausting gas recirculation is a very effective way to reduce the combustion temperature and pressure so that the NO_x can be reduced. However, the EGR ratio cannot be too high because extra techniques are required to compensate the pressure loss and thus the power output decrease due to the usage of EGR.

Fig. 7 compares the effect of different dilution gases on the pressure evolution. The initial pressure is 1 atm, the initial temperature is 423 K, the methane fraction is 95%, the equivalence ratio is 1.0 and the dilution ratio is 10%. With the increase of N_2/CO_2 ratio, the P_{max} appears earlier and its value is higher. Additionally, the maximum pressure rise rate increases with the increase of N_2/CO_2 ratio, pressure rises also faster. It is expected that for different dilution gas compositions, the total heating value of the reactants is constant. Because the diatomic molecule N_2 has lower specific heat than that of the tri-atomic molecule CO_2, the final combustion temperature

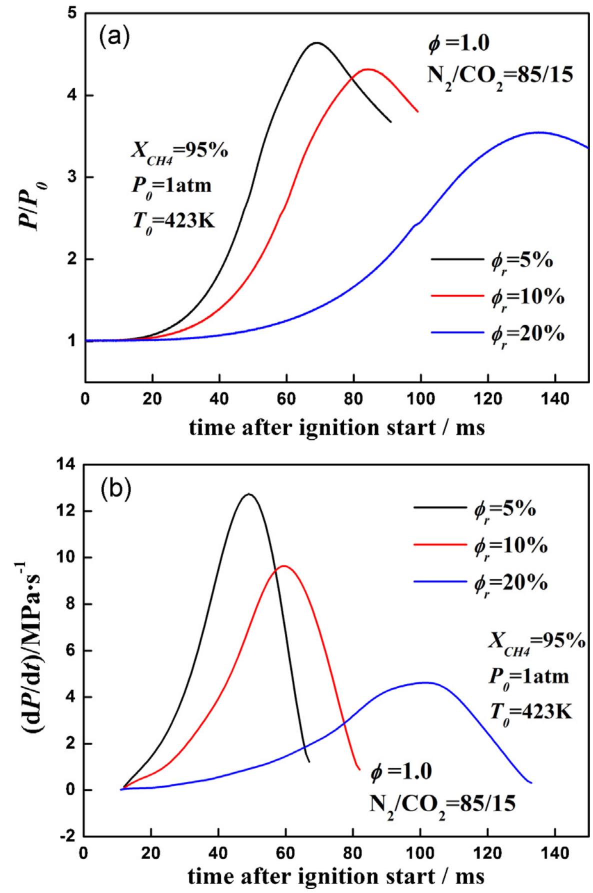

Fig. 6. Pressure history and the rate of pressure rise for different dilution ratios.

of the diluted mixture increases with the increase of N_2/CO_2 ratio. Furthermore, since the effectiveness of different diluents such as CO_2 and N_2 on the flame speed decrease has been discussed by Chen et al. [24]. They found that CO_2 has a higher flame speed suppression effect because CO_2 has larger specific heat and it also participate in chemical reactions, thus with the increase of CO_2 concentration in the diluent mixture, the pressure rise rate is significantly reduced.

3.2. The peak pressure

Ideally, the combustion pressure in a closed vessel can be calculated through thermodynamic equilibrium by using the adiabatic assumption. Thus the constant volume adiabatic equilibrium pressure (P_e) is calculated in this work. However, practically, because combustion takes place in a finite time scale and there is no real adiabatic combustion chamber, thus the peak pressure (P_{max}) of combustion is significantly lower than P_e.

Fig. 8 shows the normalized peak pressure (P_{max}/P_0) and the normalized peak equilibrium pressure (P_e/P_0) as a function of equivalence ratio at different initial conditions. The adiabatic equilibrium pressures are calculated by CHEMKIN, equilibrium model.

Fig. 8(a) shows that pressure gives its maximum value at the condition that the equivalence ratio is 1.1. Both rich and lean mixtures lead to the reduction of P_{max}/P_0. This illustrates the dominant effect of flame propagation speed on the pressure evolution process. When the methane fraction is 95%, the value of P_{max}/P_0 is higher than that of pure methane. This reveals that the presence of ethane in natural gas actually increases the final combustion pressure thus favors the enhancement of the power output. Fig. 8(b)

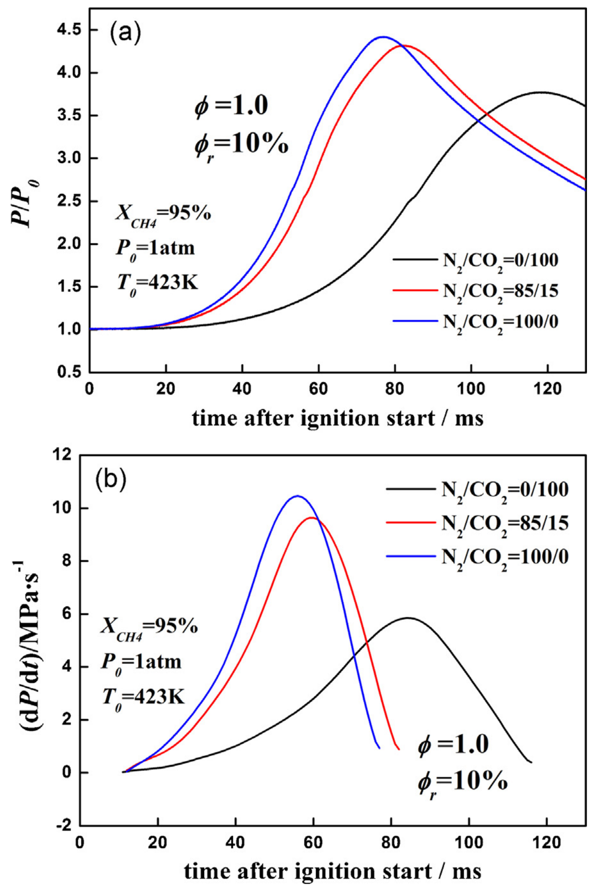

Fig. 7. Pressure history and the rate of pressure rise for different N_2/CO_2 ratios.

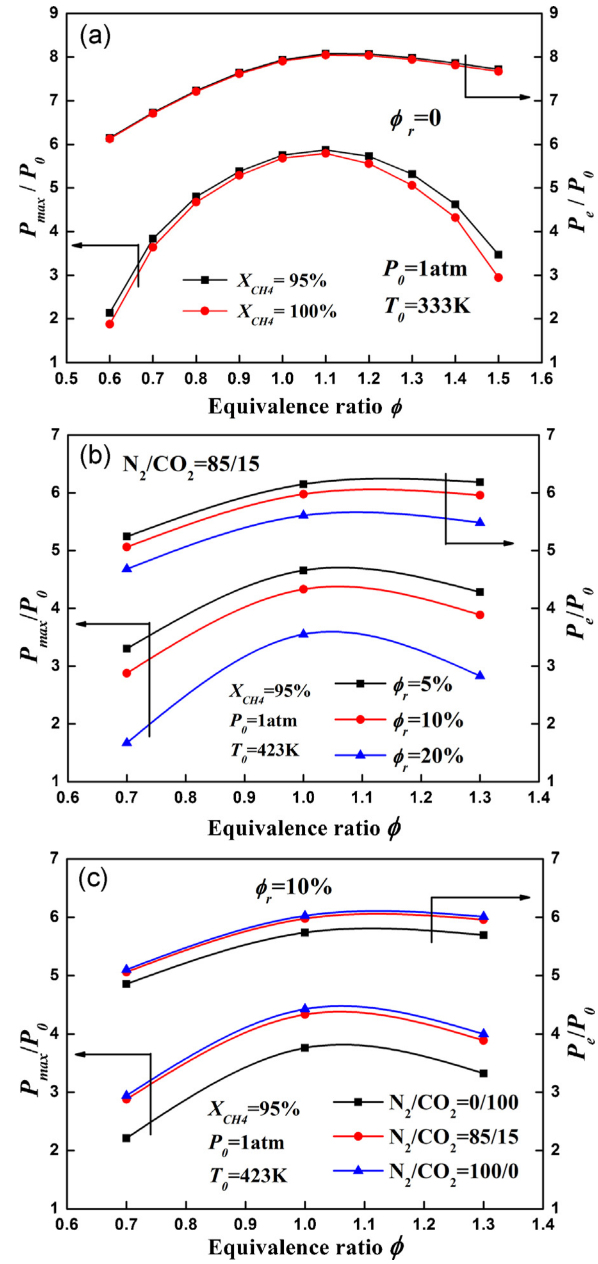

Fig. 8. P_{max}/P_0 and P_e/P_0 versus equivalence ratio for different initial conditions.

shows the effect of dilution gas on the maximum pressure when the initial pressure is 1 atm, the initial temperature is 423 K, the methane fraction is 95%, the equivalence ratio is 1.0 and the dilution gas is N_2(85%)/CO_2(15%). With the increase of dilution gas, the maximum combustion pressure is decreased significantly and the heat loss is increased. Obviously, the total amount of heating value and the flame propagation speed are both decreased with the increase of dilution ratio. Fig. 8(c) gives the effect of different kinds of dilute gas on the explosion pressure. With the increase of the proportion of nitrogen and carbon dioxide, the explosion pressure increases and the heat loss is decreased. This reveals the inhibition of carbon dioxide is stronger than nitrogen.

3.3. The combustion phasing

The combustion phasing, which is related to the heat release, is known to have a significant effect on the efficiency and emission characteristics of the internal combustion engines. The quantification of the combustion phasing in standard and well controlled experimental conditions is then important. Thus we adopt the definitions of the combustion duration (total period of combustion) [16], the flame development duration in Ref. [21].

Figs. 9 and 10 respectively give the combustion duration and the flame development time at various initial conditions. The two parameters illustrate the characteristics of the flame development and total period of combustion. Fig. 9 gives the combustion duration (t_R) at different initial conditions. Fig. 9(a) shows that for different methane fraction natural gas at the stoichiometric condition, the combustion duration increases significantly with the increase of the initial pressure. This is because the flame propagation speed is decreased with the increase of the initial pressure and thus the flame life within the combustion chamber is increased. However, because the density of the mixture is increased, the heat release is still significantly higher at higher pressures. With the increase of the initial temperature, as shown in Fig. 9(b), the combustion duration is decreased because of increased flame propagation speed. Additionally, the combustion duration is shortened as the methane fraction increases, but this behavior is very weak. The minimum value of t_R can be obtained when the equivalence ratio is around 1.1. Mixtures that have equivalence ratios far away from stoichiometric have significantly longer combustion duration. As shown in Fig. 9(c), the combustion duration is less than 100 ms for the equivalence ratio ranges between 0.9 and 1.3, however, for the equivalence ratio smaller than 0.7 or higher than 1.4, the combustion duration is higher than 200 ms. This is consistent with the flame propagation speed dependence on the equivalence ratio, as reported in Ref. [8]. Fig. 9(d) and (e) give the variation of t_R

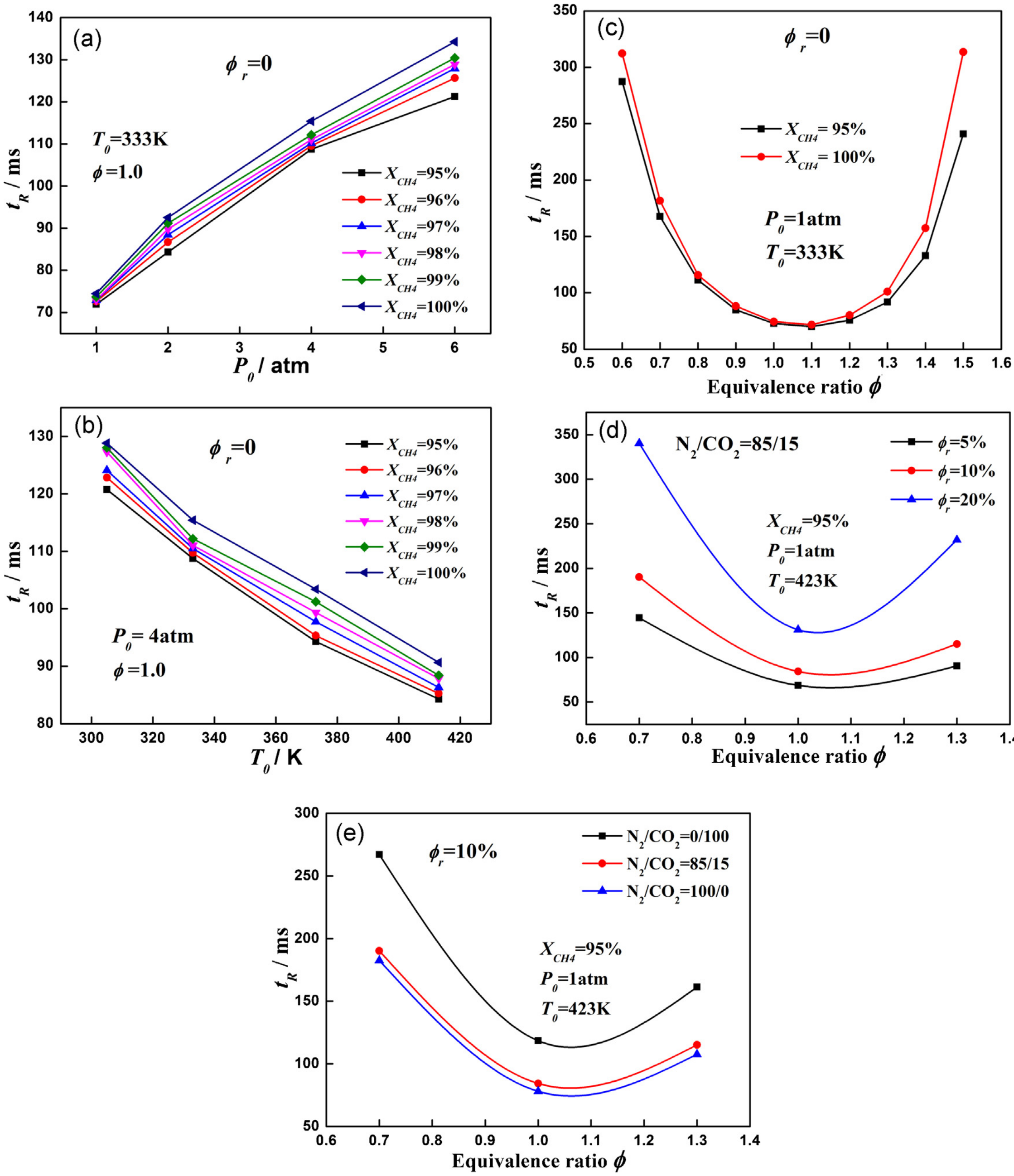

Fig. 9. Combustion duration for different initial conditions.

with different dilution gas addition. The combustion duration is significantly increased with the increase of dilution ratio because of the decreased flame propagation speed. Furthermore, the CO_2 addition results in further increase of the combustion duration.

The combustion duration decreases with the increase of the initial temperature and increases with the increase of the initial pressure. Meanwhile, it also exhibits a monotonic variation to the methane fraction. When the equivalence ratio is 1.0, a correlation between the combustion duration and the initial temperature, pressure as well as the methane fraction is taken the form of Eq. (2), and similar formations can be obtained for other equivalence ratios.

$$\frac{t_R}{145.965\,\mathrm{ms}} = \left(\frac{T_0}{305\,\mathrm{K}}\right)^{-1.174}\left(\frac{P_0}{6\,\mathrm{atm}}\right)^{0.314}\left(\frac{X_{CH_4}}{100\,\%}\right)^{1.339} \quad (2)$$

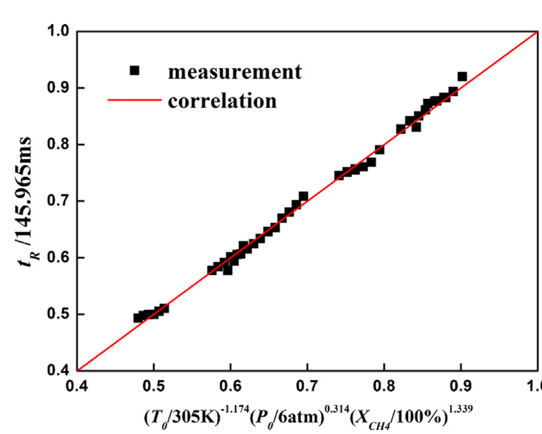

Fig. 10. Parity plot between experimental data and correlation calculation.

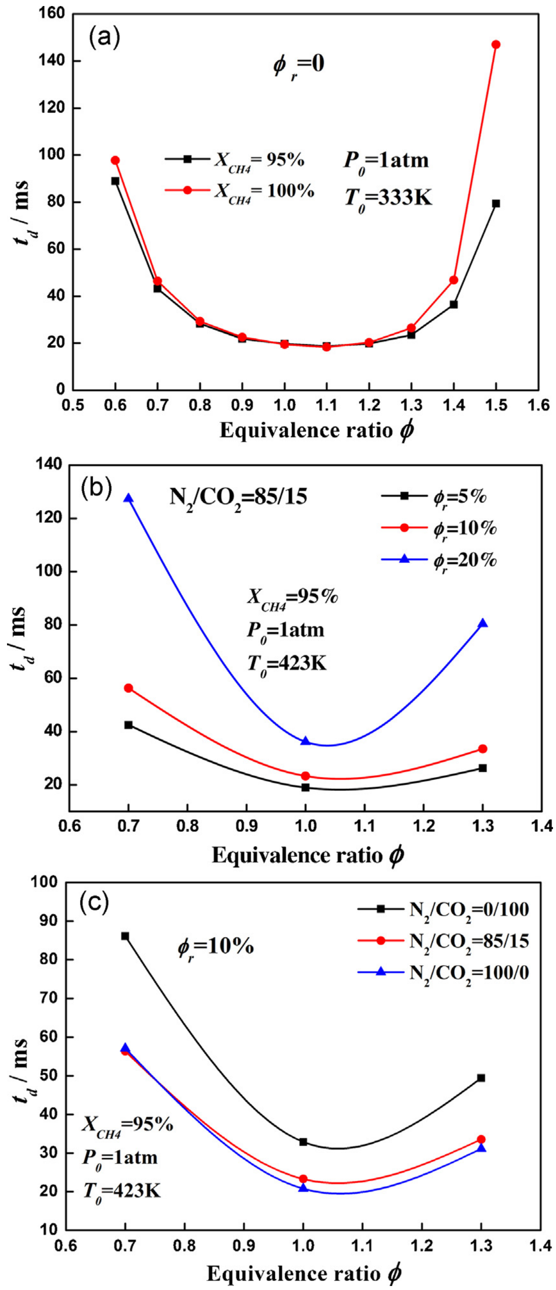

Fig. 11. Flame development time versus equivalence ratio for different initial conditions.

Fig. 10 shows that for all the test conditions, the measured combustion duration is normalized and plotted against the experimental conditions. The fitted curve by Eq. (2) is also shown for comparison. This empirical correlation is found to have a high R^2 (0.99553) thus it may be used to predict the combustion duration over the current experimental pressure and temperature range.

Fig. 11 gives the flame development time (t_d) as a function of the equivalence ratio at different initial conditions. The flame development time behavior as a function of the experimental condition is very similar to the total combustion duration: it is obviously seen that the flame development duration attains its minimum at around the equivalence ratio of 1.1, and the dilution gas addition increases the flame development duration. Additionally, the CO_2 dilution has a stronger effect in increasing the flame development duration, compared to N_2.

4. Conclusions

The explosion characteristics of high methane concentration natural gas were studied in a constant volume combustion vessel at different initial conditions. The main conclusions are summarized as follows.

1. For all tested methane fraction natural gases, with the increase of initial pressure, the peak explosion pressure, the maximum rate of pressure rise increase significantly. However, the pressure rise rate is decreased due to the increased charge density and reduced flame speed. This is confirmed by the increased total flame duration and flame development time. With the increase of the initial temperature, the peak explosion pressures decrease, but the pressure increase during combustion is accelerated, which indicates a faster flame speed and heat release rate, which is confirmed by reduced total flame duration and flame development time. The maximum value of the explosion pressure, the maximum rate of pressure rise, the minimum total combustion duration and the minimum flame development time were observed for the equivalence ratio is 1.1 mixtures.
2. With the increase of the methane fraction for a fixed initial pressure and temperature, the explosion pressure and the maximum rate of pressure rise are slightly decreased, while the combustion duration is postponed. The combustion phasing is empirically correlated with the experimental parameters.
3. The addition of dilute gas will strengthen the suppression on the explosion pressure, the maximum rate of pressure rise and the flame development. The suppression of carbon dioxide is stronger than that of nitrogen.

Acknowledgements

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