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The behavior of methane/hydrogen/air premixed flame in a closed channel with inhibition

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Abstract

Hydrogen enriched natural gas (HNG) is a promising alternative fuel. But the blended fuel will inevitably have different ignition and combustion characteristics as compared to natural gas. The extent of the resulting difference depends on the percentage of hydrogen addition. It may affect the compatibility of combustion systems and have safety implications. The present study was aimed at enhancing the safety of HNG through inhibition by inert gases. Laboratory tests were conducted for methane/hydrogen/air premixed flame propagating in a closed channel with either nitrogen (N₂) or carbon dioxide (CO₂) as the inhibitor. Mixtures with different hydrogen volumetric fractions in fuel, including 0%, 20%, 50% or 80% were investigated. The flame shape evolution and the overpressure in the channel were recorded by high-speed Schlieren

photography and pressure sensor, respectively. The flame shape was found to change in various ways depending on the inhibitor and hydrogen content. The pressure wave had remarkable impacts on flame and pressure dynamics. The effect of buoyancy on the flame deformation was observed and discussed. Both N_2 and CO_2 were found to have considerable inhibition effect on the flame speed and overpressure build-up in the channel while the inhibiting effect of CO_2 was stronger. The inhibition mechanisms of either N_2 or CO_2 were revealed from thermal and kinetic aspects.

Keywords: HNG, Premixed flame, Inhibition, Buoyancy, Mechanism

1 Introduction

Hydrogen enriched natural gas (HNG) is a promising alternative fuel. It was proved that the hydrogen addition (enrichment) can overcome the shortages of NG combustion system, such as high ignition energy, local flame extinction, low power output [1, 2] and significantly enhance the flame dynamics and combustion efficiency [3, 4]. Besides, due to its composition of only natural gas and hydrogen, the combustion of HNG mainly yields CO_2 and H_2O with constraint pollutant released. Given the series of environmental problems associated with fossil fuel, the HNG certainly has an excellent prospect as energy carrier in the near future. However, the mixed fuel will inevitably have different ignition and combustion characteristics as compared to natural gas. The extent of the resulting difference will depend on the percentage of hydrogen addition. It may affect the compatibility of the combustion systems and have safety implications.

From the safety perspective, both hydrogen and natural gas are highly flammable. Hydrogen is featured with low ignition energy, broad flammable range and large burning velocity [5, 6].

Therefore, the potential fire and explosion hazards related to HNG need to be evaluated. Particularly in confined spaces, once the HNG is exposed to air, a combustible mixture will be formed, which can be easily ignited, resulting in fire or explosion. Flame propagation in confined spaces is complicated by flame instability, pressure wave and boundary effect, etc. [7-9]. Confinement can generally promote flame acceleration and even induce deflagration to detonation transition (DDT) as a consequence [10]. At laboratory scale, flame propagation is usually investigated in channels simulating the confined spaces. For example, Mallard and Le Chatelier [11] reported their pioneering work in 1883, which captured a special flame structure in a long channel, namely, the flame front was irregular and would invert itself on the centreline. Thereafter, Ellis [12] took the first photograph of the inverted flame in the channel in 1928. Salamandra et al. [13] named this featured flame shape as “tulip flame”. Until 1990s, Clanet and Searby [14] distinguished four characteristic stages of flame deformation in channels, including spherical flame expansion, finger-shaped flame acceleration, flame-side wall contact and the tulip inversion. In recent years, the flame acceleration with shape changes attracted continuous attention in the area of combustion and explosions. For instance, Zhang et al. [15] found the varied structures of flame front during propagation in vented channel during their experimental and numerical investigations. Yu et al. [16-19] conducted a series of experiments in both closed and half-open channels to investigate the flame behaviour for methane, hydrogen and syngas in air. Xiao et al. [20] numerically investigated the influence of pressure wave on the stability of flame propagation in channels and found out that the Rayleigh-Taylor instability mainly causes the initiation of distorted tulip flame. Obviously from previous studies, the flame propagation in

confined spaces was very complex accompanied by drastic flame deformation and acceleration, which could result in violent pressure rise as well.

For safety consideration, it is beneficial to develop measures that can inhibit potential flame acceleration during combustion. The addition of some inhibitor is useful to constrain the flame and pressure dynamics in combustion. Many studies on the inhibiting effect of inert gas, water mist, powder and halogenated compounds have been carried out. For instance, Benedetto et al. [21] conducted explosion tests of $\text{CH}_4/\text{O}_2/\text{N}_2/\text{CO}_2$ and $\text{H}_2/\text{O}_2/\text{N}_2/\text{CO}_2$ in a closed cylindrical chamber. The results illustrated that the addition of CO_2 influenced the ignition, laminar burning velocity, flame temperature and combustion rate. Shen et al. [22] discussed the inhibiting effect and mechanism of N_2 or CO_2 on $\text{C}_2\text{H}_4/\text{N}_2\text{O}$ mixture explosion at various initial pressures. Explosion mitigation by water mist was examined for methane/air explosion in the tube by Wang et al. [23] in varied droplet sizes and pipe sizes. Yang et al. [24] built an inhibition model for water mist. Pei et al. [25] discussed the synergistic inhibition of inert gas and ultrafine water mist and proposed a promoted inhibiting effect of gas-liquid two phase medium. Yu et al. [26] compared the performance of Urea, $\text{Al}(\text{OH})_3$ and $\text{Mg}(\text{OH})_2$ on gas explosion. Dounia et al. [27] investigated the sodium bicarbonate powder on flame propagation inhibition by theoretical analysis and simulation. The combined effect of water/sodium chloride mist on the methane/air explosion was explored by Cao et al. [28]. Linteris and Truett [29] considered the inhibiting effect of fluoromethanes (CH_2F_2 , CF_3H or CF_4) on the burning velocity by calculations and measurements. The numerical study of the inhibiting efficiency of halogenated compounds (CF_3Br , CF_3I , CF_3H , C_2HF_5 , C_2F_6 or CF_4) was elucidated by Noto et al. [30]. These studies identified potential inhibitors and initiated the discussion about the underlying mechanism. Among above-mentioned

inhibitors, N₂ and CO₂ have the advantages of cleanliness, abundant in reserve and low cost [25]. Some previous studies on the inhibiting effect of N₂ or CO₂ for methane/air [31-35], hydrogen/air [36-39] combustion have been conducted. However, as mentioned before, the mixed fuel would have distinct combustion characteristics, and thus would probably give different response to either N₂ or CO₂ inhibition. Therefore, present study aimed to investigate their inhibiting effect on methane/hydrogen/air premixed flame, which is quite beneficial for quantitative assessment on safety measures for HNG utilization.

2 Experimental setup

The experimental setup is shown in Fig. 1, which mainly contains a rectangular channel with an inner cross-section of 25 mm × 25 mm and 300 mm long, a Schlieren system, a high-speed camera (FASTCAM SA2), a pressure sensor, a high-voltage igniter, an oscilloscope, a gas mixing system and a synchronization controller. The bottom and top walls of the channel were made of TP304 stainless steel, while the side walls were entirely replaced by two thick quartzs for optical path of Schlieren photography to capture the whole process of flame evolution from ignition.

The premixed gas mixture was firstly prepared in a gas mixing device then injected into the channel. The mixture was settled for at least 10 min. A pair of spark electrodes were located near the left end wall and the spark gap was rightly on the axis of the channel. The pressure sensor was placed on the top wall near the right end wall.

The testing gas in this study was methane/hydrogen/air mixture with various inhibitor contents (φ_{CO_2} or φ_{N_2}) and hydrogen fractions in fuel (φ_{H_2}), which were defined as follows:

$$\varphi_{N_2} = \frac{V_{N_2}}{V_{mixture}} \quad (1)$$

$$\varphi_{CO_2} = \frac{V_{CO_2}}{V_{mixture}} \quad (2)$$

$$\varphi_{H_2} = \frac{V_{H_2}}{V_{H_2} + V_{CH_4}} \quad (3)$$

where V_{N_2} , V_{CO_2} , V_{H_2} , V_{CH_4} and $V_{mixture}$ were the volumes of nitrogen, carbon dioxide, hydrogen, methane and the testing gas mixture, respectively. The purity of gases used in this work were 99.98% for CO_2 , 99.98% for N_2 , 99.90% for air, 99.99% for H_2 and 99.99% for CH_4 . The experiments were repeated at least three times to ensure the repeatability and the measurement uncertainty of the pressure sensor was $\pm 1\%$. The initial conditions were 298 K and 101325 Pa.

3 Flame shape evolution

Experiments were conducted for premixed methane/air flame inhibited by 0%-20% N_2 and 0%-20% CO_2 . Examples of the entire process of flame evolution recorded by the high-speed Schlieren photography are shown in Fig. 2, which captured the four-stage flame deformation defined by Clanet and Searby [14]. The flame expanded spherically right after ignition. After a short period, due to the confinement of the wall, the flame was gradually stretched into a finger shape. Then the skirt of the flame touched the sidewall. The flame-wall contact points moved along the wall fast catching the flame tip on the centreline. Eventually, the flame front turned into a plane shape and immediately after this, the flame front inverted forming a cusp towards the burnt gas. A classic tulip flame came into being. With the increase of inhibitor content, the moment when these characteristic structures were formed delayed.

Following the classic tulip inversion, some peculiar flame shapes appeared under certain conditions as shown in Figs. 3a and 3b. With the same inhibiting condition, the extent of flame deformation was stronger for mixtures with higher hydrogen content. For instance, the pure methane flame maintained its tulip shape until quenching in the absence of inhibition. When the hydrogen content, φ_{H_2} was increased to 20%, a few wrinkles appeared on the tulip flame lips. When φ_{H_2} was increased to 50%, some new cusps emerged near the sidewalls and they moved along the flame front towards the primary cusp. Such phenomenon was referred to as “cloudy flame” in our previous publication [40]. With φ_{H_2} reached 80% in the absence of inhibition, the flame shape underwent a more complicated deformation including elongated tulip shape, T-shape [5], and cellular flame [41] until the end. With the inhibition of N_2 or CO_2 , the flame structures were more stable. For example, for $\varphi_{H_2} = 50\%$ and 10% N_2 addition, the flame maintained the classic tulip flame until the end without changing to “cloudy flame”. In almost all cases, the flame propagated faster near the upper wall (e.g. at 61.90 ms with 10% N_2 and $\varphi_{H_2} = 0\%$ in Fig. 3a). This was thought to be due to the buoyancy effects: firstly, as the density of N_2 (or CO_2) is higher than that of CH_4 and H_2 mixtures, concentration gradients would be built up with time leading to relatively higher H_2 content near the top and higher N_2 fraction near the bottom. Thus, the inhibition was stronger on the flame near the bottom. Such effect was, however, less obvious for the cases with 80% H_2 within the range of N_2 fractions investigated, this was thought to be due to the higher flame speeds and relatively shorter physical times within the channel during which the concentration gradients were not sufficiently developed. Secondly, the flame with inhibition was slowed down, during which the movement of burnt gas affected by buoyancy

became more and more apparent. Therefore, the flame propagated obliquely with resultant force from buoyancy and combustion.

4. Flame dynamics

4.1 Flame propagation speed

Figure 4 shows the propagation speed of flame forefront versus time with varied hydrogen or inhibitor concentrations. All the flames accelerated at the early stage until reaching the maximum. With either increase of inhibitor content or decrease of φ_{H_2} , the occurrence of maximum flame propagation speed (v_{max}) delayed accordingly. After that, the propagation speed fluctuated with time accompanying the flame shape changes. At the condition of $\varphi_{H_2} = 80\%$ and 0% inhibitor, the propagation speed of the flame forefront fluctuated most drastically and even receded with negative propagation speed. As discussed in our previous study [40], at conditions with high hydrogen content, relatively stronger pressure waves which also propagated faster were reflected from the end of the channel, pushing back the unburnt gas with the effect of causing the flame to propagate backwards. Besides, the loss of the flame area caused by the flame-wall contacting was also an important factor affecting the flame propagation speed. It is noteworthy that the maximum flame propagation speed always appeared at the moment after the flame first touching the sidewall. This indicated that the flame speed did not decrease immediately upon touching the wall. But due to the loss of flame surface area, the acceleration was believed to decline immediately. The flame speed decrease caused by N_2 or CO_2 addition underwent different tendency. Take the condition of $\varphi_{H_2} = 20\%$ as example. The decrement of v_{max} with addition of N_2 or CO_2 is shown in Table 1. With the same addition of N_2 or CO_2 , the v_{max}

was always lower with CO₂ dilution. However, the decrement of flame speed at the condition of 10%-20% CO₂ was lower than that at the condition of 10%-20% N₂, though the value of v_{max} with CO₂ dilution was still smaller. This phenomenon was different from the linear relationship between flame speed and inhibitor content in several previous studies [42-46] on laminar flame, which may be attributed to the effect of wall interaction in this channel. However, the laminar flame speed calculated and discussed in the later section presented similar descending trend with those in previous studies [42-46].

Table 1. The decrement of v_{max} with addition of N₂ or CO₂.

Inhibitor content, %	Inhibited by N ₂ , $\varphi_{H_2} = 20\%$		Inhibited by CO ₂ , $\varphi_{H_2} = 20\%$	
	v_{max} , m/s	Decrement, m/s	v_{max} , m/s	Decrement, m/s
0	22.76	/	22.76	/
5	16.58	6.18	12.11	10.65
10	13.05	3.53	4.24	7.87
15	8.08	4.97	3.08	1.16
20	5.75	2.33	1.32	1.76

4.2 Overpressure dynamics

The overpressure in the channel is shown in Fig. 5. It rose rapidly following ignition before reaching its maximal value. With the increase of inhibitor content, the maximum overpressure decreased, and its arrival time was delayed. Analogous to the flame propagation speed, the overpressure rose and fluctuated more gently with increasing inhibitor content or decreasing

hydrogen fraction. As illustrated in Fig. 6, with CO₂ addition, the maximum overpressure was lower than that with N₂ addition for the same fuel composition.

5 Inhibiting mechanism of N₂ and CO₂

5.1 Thermal diffusivity

Thermal diffusivity is an important parameter to describe the heat transport in materials [47]. The decrease of thermal diffusivity would lead to the inhibition of flame propagation [48]. Figure 7 shows the thermal diffusivity versus inhibitor content with different hydrogen addition. It can be seen that the thermal diffusivity decreased with the increase of N₂ or CO₂, indicating that the addition of N₂ or CO₂ leads to reduction in the thermal diffusivity, resulting in the inhibition of flame propagation. The addition of CO₂ had stronger effect in reducing the thermal diffusivity and hence stronger inhibition effect. With the increase of φ_{H_2} and constant inhibitor content, the thermal diffusivity increased.

5.2 Thermal conductivity

On the other hand, the thermal conductivity is another property affecting the laminar flame speed of the premixed flame [49]. The computed thermal conductivity values for the mixtures considered are plotted in Fig. 8. As expected, with the increase of φ_{H_2} and constant inhibitor content, the thermal conductivity and laminar flame speed both increased. With the increase of inhibitor content, the thermal conductivity decreased resulting in lower laminar flame speed shown in Fig. 9. CO₂ had more profound effect on reducing the thermal conductivity and hence stronger inhibition effect.

5.3 Laminar flame speed

The laminar flame speed versus time is shown in Fig. 9. The laminar flame speed decreased almost linearly with N₂ addition. However, with CO₂ addition, it decreased monotonically in a curve shape with the decrement reduced with gradual increasing CO₂ content. But CO₂ dilution always led to lower laminar flame speed than N₂ dilution at the same content.

5.4 Flame thickness

Flame thickness has inhibiting effect on the hydrodynamic instability which influences the flame front stability [50, 51], i.e. the increase of flame thickness can lead to more stable flame evolution, i.e. presenting less flame shape changes. Flame thickness was calculated as follows [48, 52]:

$$\alpha = \frac{\lambda}{\rho_u \cdot C_p} \quad (4)$$

$$\delta = \frac{2\lambda}{\rho_u \cdot C_p \cdot S_L} \left(\frac{T_{ad}}{T_0} \right)^{0.7} \quad (5)$$

where the α , λ , ρ_u , C_p , δ , S_L , T_{ad} and T_0 were thermal diffusivity, thermal conductivity, unburnt mixture density, specific heat, flame thickness, laminar flame speed, adiabatic flame temperature and initial temperature of unburnt mixture, respectively. The calculation of laminar flame speed was based on HP-Mech [53] using CHEMKIN Pro software [54]. The other parameters were calculated by GASEQ [55] using chemical equilibrium method. As shown in Fig. 10, the flame thickness was enlarged with increasing inhibitor content, which would stabilize the flame front against instabilities, as can be seen from flame images above. The increase of flame thickness

was more prominent with higher inhibitor content. In general, the flame deformation would be attenuated due to the thickening of flame front by N₂ or CO₂ addition, especially with CO₂.

5.5 Kinetic effect and Adiabatic flame temperature

Firstly, due to the third-body collisional effect, the addition of N₂ or CO₂ would enhance two of the sensitive reactions for ignition ($H + O_2 + M = HO_2 + M$, $CH_3 + CH_3 + M = C_2H_6 + M$) and one of the sensitive reactions for laminar flame speed ($H + O_2 + M = HO_2 + M$) [56]. M is the third body and characterized by collisional efficiency. The reaction of $CH_3 + CH_3 + M$ is a chain-terminating step, which would inhibit the combustion by producing relatively stable compound of C₂H₆ in the radical pool. The reaction ($H + O_2 + M = HO_2 + M$) will compete for highly active H radical with the reaction ($H + O_2 = O + OH$) and produce less active HO₂ radical [57]. As a result, the inhibitor addition would result in weakened combustion and flame propagation. In both reaction paths, the collisional efficiency of CO₂ is greater than that of N₂ [53]. Besides, it is noteworthy that CO₂ also acted as a main reactant in the path of ($CO + OH = H + CO_2$) which is a very important reaction in combustion chemistry [45, 58]. Evidently, the addition of CO₂ would reverse the reaction ($CO + OH = H + CO_2$), which competed for extra H radical with ($H + O_2 = O + OH$) resulting in reductions of important radicals of O, H, and OH [58]. Therefore, compared to N₂, CO₂ had more pronounced inhibiting kinetics. Further, it could also infer that with higher hydrogen content, the advantage of CO₂ inhibition would be more enhanced since the pathways evolving H radical are of such great importance. The experimental results give some proofs in Fig. 6 that the inhibition discrepancy between N₂ and CO₂ on maximum overpressure was enlarged with 80% H₂ content.

The reduced adiabatic flame temperature can manifest the inhibited kinetics, which was calculated using the chemical equilibrium method [55] as shown in Fig. 11. The mixture with CO₂ addition at the same amount had lower flame temperature as expected. Nevertheless, it needs to mention that, besides the kinetic effect, both CO₂ and N₂ can reduce flame temperature with their high specific heat. Comparatively, CO₂ has larger specific heat than N₂ [42, 49]. As suggested by Law [49] and Hu et al. [44], the adiabatic flame temperature is closely related to laminar flame speed. Therefore, according to Eq. (5), the decreasing of adiabatic flame temperature would remarkably thicken the flame front.

6 Conclusions

The flame propagation of methane/hydrogen/air mixture was studied in a closed channel with inhibitor (N₂ or CO₂) addition. The hydrogen fractions in the fuel mixture were 0%, 20%, 50% and 80% by volume. For different φ_{H_2} and inhibitor contents, different flame evolution processes were observed.

Either increasing the inhibitor content or decreasing φ_{H_2} could remarkably decrease the flame propagation speed and the overpressure. At the same φ_{H_2} and inhibitor content, CO₂ had more profound inhibition effect on flame acceleration and overpressure rise.

Thermal analysis showed that the addition of N₂ and CO₂ would decrease thermal diffusivity and thermal conductivity, resulting in the reduction of laminar flame speed. The addition of the inhibitor resulted in the increase of flame thickness, which attenuated the flame deformation and consequently suppressed the flame acceleration. The inhibitor could also decrease the adiabatic flame temperature by inhibited kinetics and high specific heat. Compared to N₂, the

mixture with CO₂ addition has lower thermal diffusivity, conductivity and laminar flame speed, smaller adiabatic flame temperature and larger flame thickness.

Kinetic analysis showed that both N₂ and CO₂ addition could inhibit the chemical process due to their collisional effect on key elementary reactions including $H + O_2 + M = HO_2 + M$, $CH_3 + CH_3 + M = C_2H_6 + M$. The collisional efficiency of CO₂ is higher than N₂. Additionally, CO₂ could reverse the reaction ($CO + OH = H + CO_2$) effectively consuming extra H atom. Therefore, CO₂ has more pronounced inhibition kinetics than that of N₂.

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Figure captions:

Fig. 1 Experimental apparatus.

Fig. 2 Flame evolution in the channel (a: inhibited by N₂; b: inhibited by CO₂).

Fig. 3 The flame evolution in the channel at later stages with hydrogen addition (a: inhibited by N₂; b: inhibited by CO₂).

Fig. 4 Flame propagation velocity versus time with varied φ_{H_2} or inhibitor addition (a, b: $\varphi_{H_2} = 0\%$; c, d: $\varphi_{H_2} = 20\%$; e, f: $\varphi_{H_2} = 50\%$; g, h: $\varphi_{H_2} = 80\%$; a, c, e, g: inhibited by N_2 ; b, d, f, h: inhibited by CO_2).

Fig. 5 Overpressure in the channel versus time with varied φ_{H_2} and inhibitor content (a, b: $\varphi_{H_2} = 0\%$; c, d: $\varphi_{H_2} = 20\%$; e, f: $\varphi_{H_2} = 50\%$; g, h: $\varphi_{H_2} = 80\%$; a, c, e, g: inhibited by N_2 ; b, d, f, h: inhibited by CO_2).

Fig. 6 Maximum overpressure versus inhibitor content (a: $\varphi_{H_2} = 0\%$; b: $\varphi_{H_2} = 20\%$; c: $\varphi_{H_2} = 50\%$; d: $\varphi_{H_2} = 80\%$).

Fig. 7 Thermal diffusivity versus inhibitor content with varying φ_{H_2} .

Fig. 8 Thermal conductivity versus inhibitor content with varying φ_{H_2} .

Fig. 9 Laminar flame speed versus inhibitor content with varying φ_{H_2} .

Fig. 10 Flame thickness versus inhibitor content with varying φ_{H_2} .

Fig. 11 Adiabatic temperature versus inhibitor content with varying φ_{H_2} .

Fig. 1

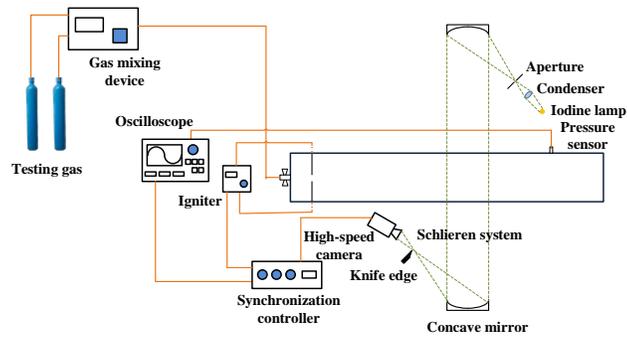
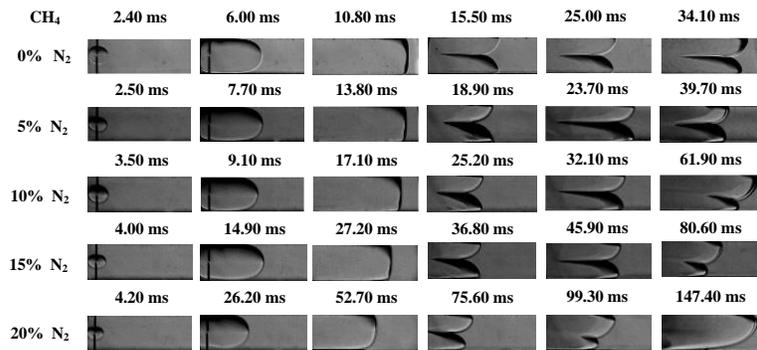
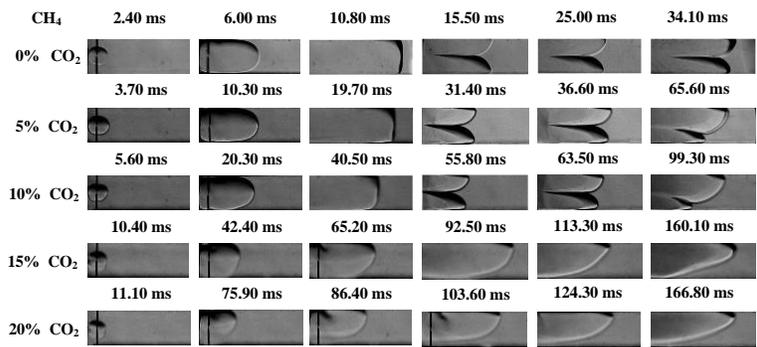


Fig. 2

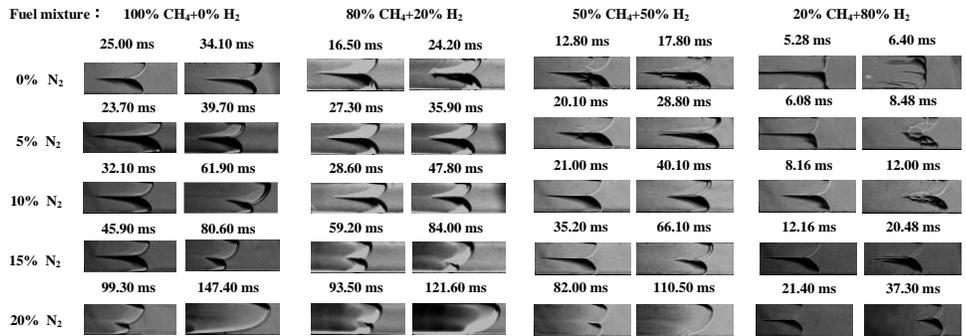


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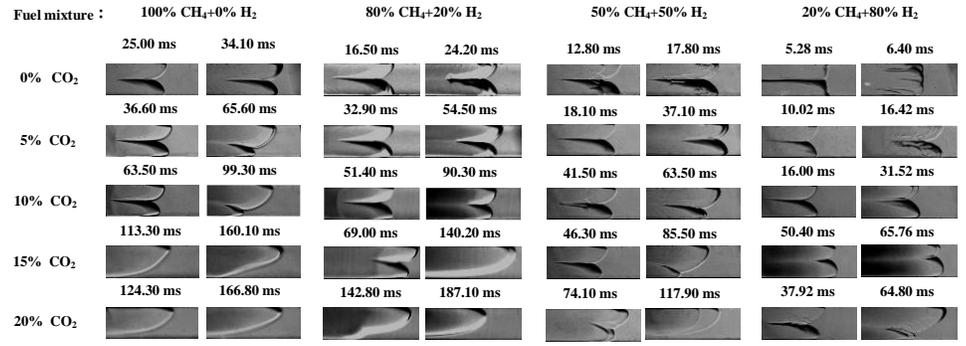


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Fig. 3

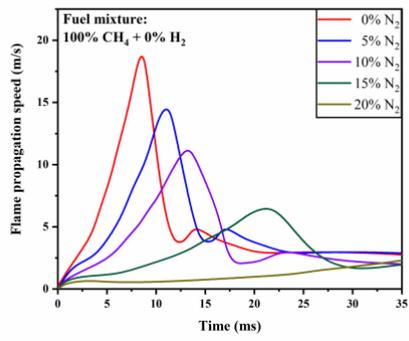


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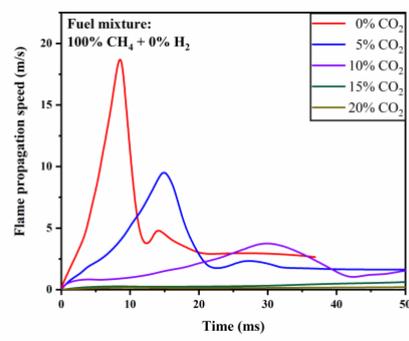


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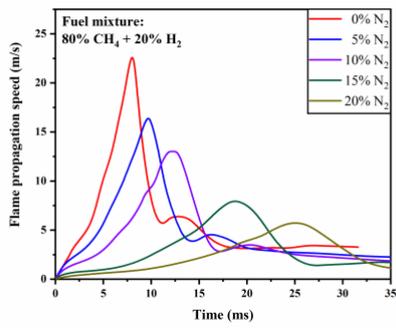
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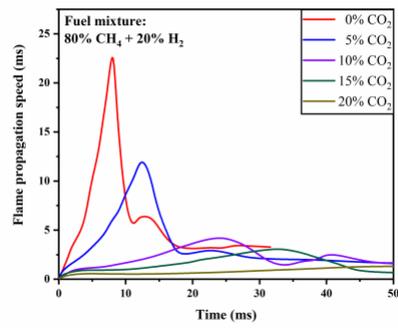
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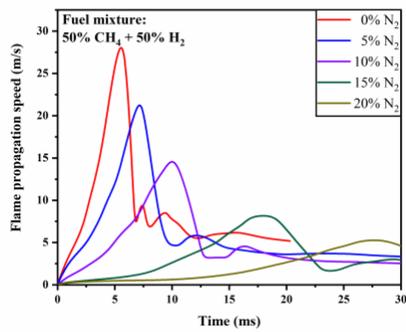
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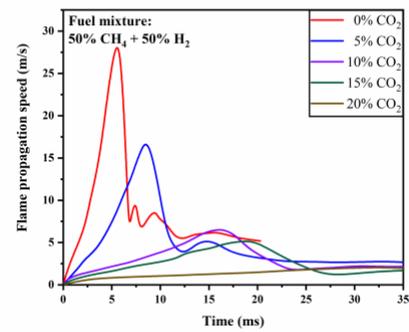
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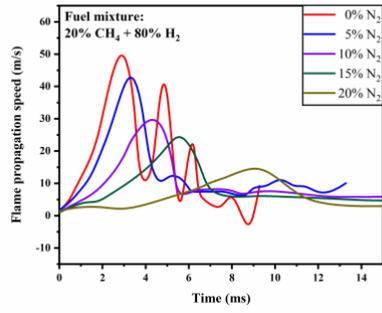
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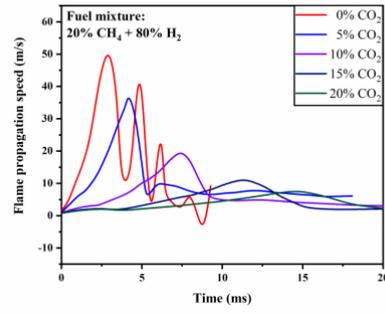
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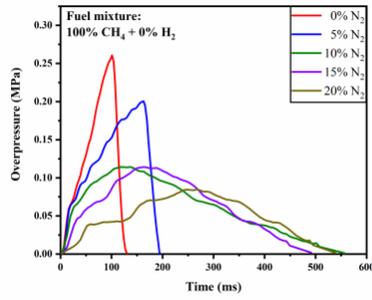


(g)

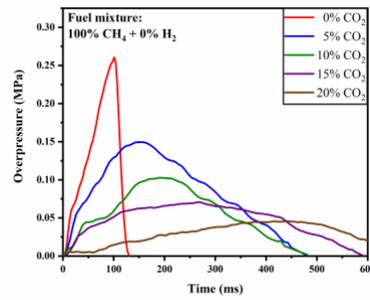


(h)

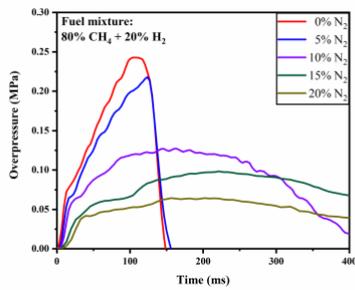
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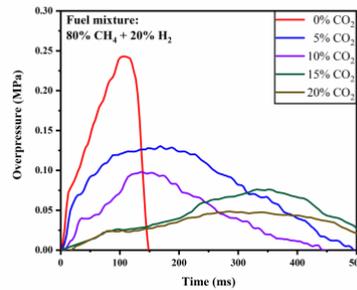
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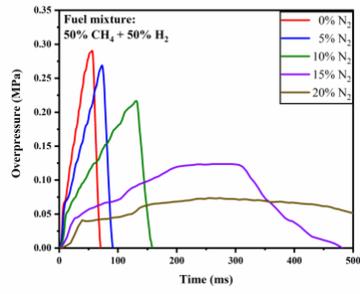
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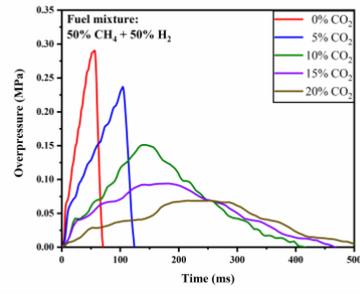
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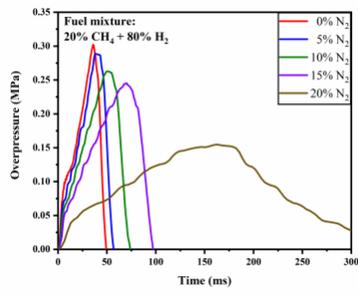
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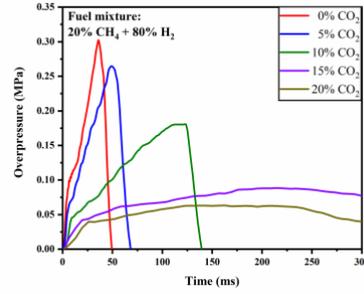
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(f)

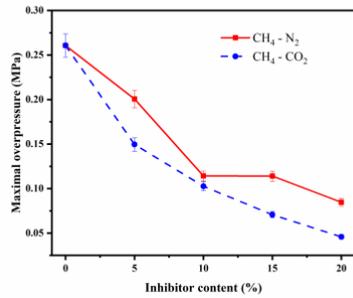


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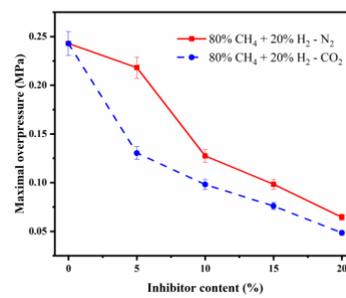


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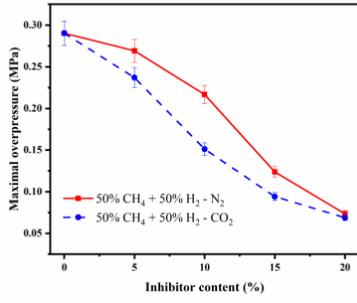
Fig. 6



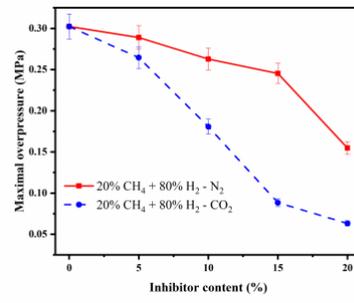
(a)



(b)



(c)



(d)

Fig. 7

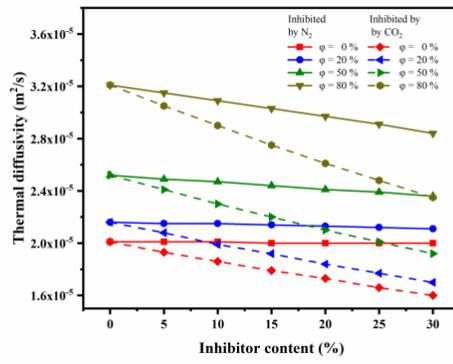


Fig. 8

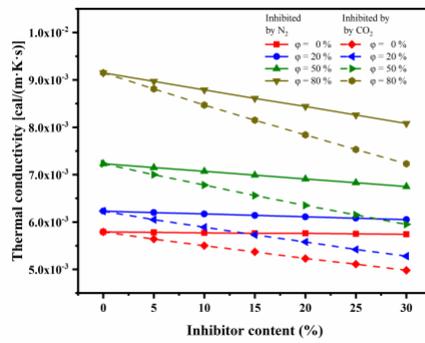


Fig. 9

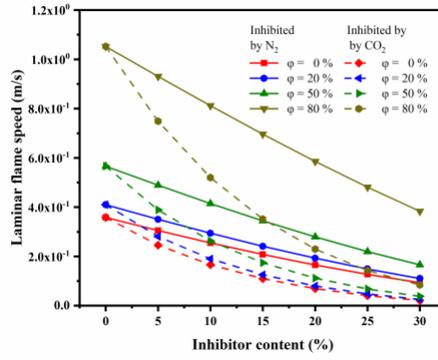


Fig. 10

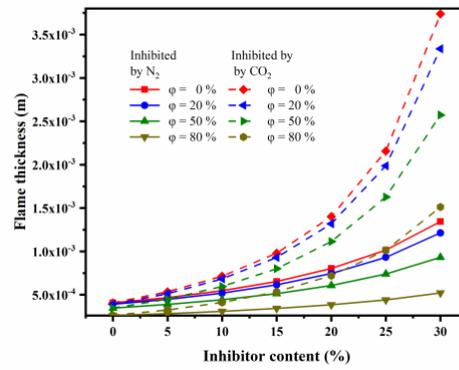


Fig. 11

