



Effect of CO₂ dilution on laminar burning velocities, combustion characteristics and NO_x emissions of CH₄/air mixtures

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Abstract

The laminar combustion characteristics of CH₄/air premixed flames with CO₂ addition are systemically studied. Experimental measurements and numerical simulations of the laminar burning velocity (LBV) are performed in CH₄/CO₂/Air flames with various CO₂ doping ratio under equivalence ratios of 1.0–1.4. GRI 3.0 mech and Aramco mech are employed for predicting LBV, adiabatic flame temperature (AFT), important intermediate radicals (CH₃, H, OH, O) and NO_x emissions (NO, NO₂, N₂O), as well as the sensitivity analysis is also conducted. The detail analysis of experiment and simulation reveals that as the CO₂ addition increases from 0% to 40%, the LBVs and AFTs decrease monotonously. Under the same CO₂ doping ratio, the LBVs and AFTs increase first and then decrease with the increase of equivalence ratio, and the maximum of LBV is reached at equivalence ratio of 1.05. The mole fraction tendency of important intermediates and NO_x with equivalence ratio and CO₂ doping ratio are similar to the LBVs and AFTs. Reaction $H + O_2 \rightleftharpoons O + OH$ is found to be responsible for the promotion of the generation of important intermediates and NO_x under the equivalence ratios and CO₂ addition through sensitivity analysis. The sensitivity coefficients of elementary reactions that the increasing of CO₂ doping ratio promotes or inhibits formation of intermediate radicals and NO_x decreases.

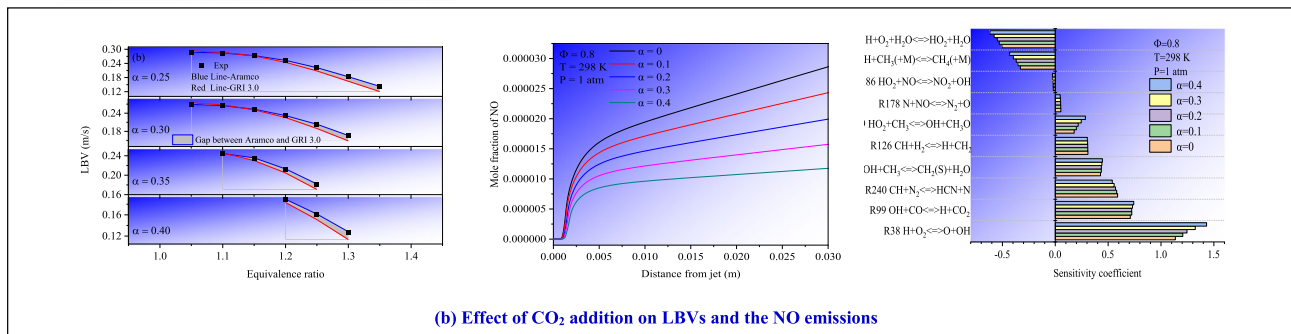
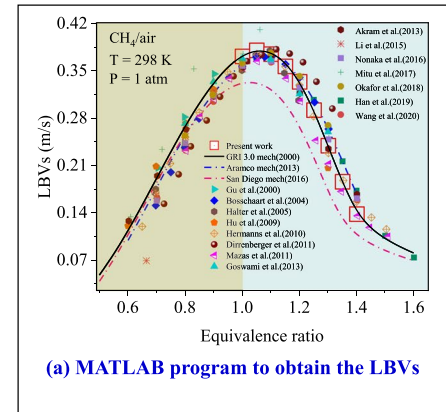
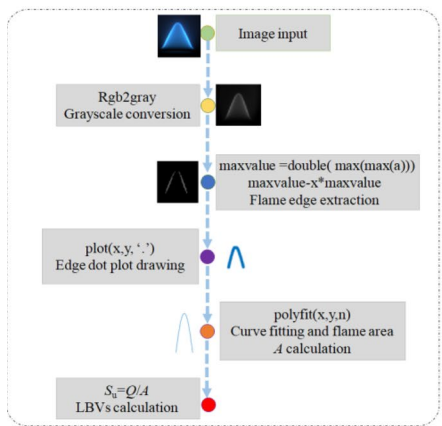
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Graphical abstract



Keywords CO₂ dilution · Laminar burning velocity · Adiabatic flame temperature · Sensitivity analysis

List of symbols

| | |
|----------------------|-----------------------------------|
| AFT | Adiabatic flame temperature |
| CH ₄ | Methane |
| CO ₂ | Carbon dioxide |
| LBV | Laminar burning velocity |
| MFC | Mass flow controller |
| N ₂ | Nitrogen |
| O ₂ | Oxygen |
| V _{diluent} | Volume fractions of dilution gas |
| V _{fuel} | Volume fractions of dilution fuel |
| ϕ | Equivalence ratio |
| α | CO ₂ doping ratio |
| \dot{m}_{fuel} | Mass flow rates of fuel |
| \dot{m}_{air} | Mass flow rates of oxidant |

1 Introduction

Carbon dioxide (CO₂) and methane are responsible for global warming and climate change. Human beings must solve a series of problems related to global warming, and greenhouse gas emissions is the main factor leading to this problem (Wang et al. 2021; Li and Fang 2014). In order to

cope with the high CO₂ concentration in the atmosphere, it is necessary to reduce carbon emissions through various methods. CO₂ capture and utilization have attracted more and more attention, such as CO₂ capture (Talapaneni et al. 2019; Huang et al. 2019) and conversion of CO₂ to hydrocarbons (Ao et al. 2020; Guo et al. 2019; Zhang et al. 2021; Shamiri et al. 2016). Besides, exploring the effect of CO₂ dilution on the fuel combustion (e.g., hydrocarbons, synthesis gas, biogas, etc.) is also one of the important research fields. Exhaust gas recirculation (EGR) has been shown to be an effective method to reduce NO_x emissions and improve burst resistance. EGR contains gases consisting a large amount of CO₂, and EGR affects the combustion process through three pathways: (1) thermal, (2) dilution, and (3) chemical effects (Wang et al. 2022). CO₂ is also required as a diluent for both moderate and intense low oxygen dilution combustion and oxyfuel combustion (Liu et al. 2020). Methane, as the simplest hydrocarbon fuel, is of great theoretical and practical importance for the study of its blending and combustion with CO₂. Laminar combustion is the basis of turbulent combustion and the cornerstone of further study of combustion and it can reflect various combustion characteristic parameters such as laminar burning velocity, ignition energy, maximum

flame temperature, ignition delay, ignition temperature and concentration of fuel (Movileanu et al. 2011). Laminar burning velocity (LBV) is an inherent characteristic of fuel and an important parameter for laminar flame propagation and stability. Laminar burning velocity depends on the type and composition of fuel and initial conditions such as equivalence ratio, pressure and temperature, and contains important information such as reaction, diffusion, heat release, tempering, instability, etc. (Chu et al. 2020; Nonaka and Pereira 2016; Zhang et al. 2015). It's a very vital parameter in the design effective control of combustion systems, such as the design and manufacture of combustors and explosion suppression devices, and the optimization of internal combustion engines (Hu et al. 2009a, b; Dirrenberger et al. 2011; Razus et al. 2010; Ren et al. 2019a, b; Chu et al. 2019).

Sampath et al. (2023) investigated the effect of CO₂ on the laminar burning velocity of CH₄/air at high temperatures by using the externally heated diverging channel (EHDC) method. It was shown that the dilution of CO₂ enhances the competition for H-atom consumption, and the LBV increased with the increase of the mixture and decreased with the proportion of doped CO₂. Ghabi et al. (2023) studied the effect of microsecond pulsed plasma on non-premixed biogas. Shang et al. (2022) systematically investigated the effect of N₂/CO₂ on H₂/CH₄/air laminar flame velocity. Ueda et al. (2021) explored the effect of CO₂ on the premixed combustion characteristics of methane/air using the spherical expansion method. Jithin et al. (2020) investigated the combined effect of CO₂/N₂ dilution on the laminar combustion rate of methane/oxygen by heat flux method and numerical simulation. The results showed that the laminar burning velocity decreased with increasing proportion of blended CO₂ and it is found that the laminar burning velocity decreased more in the fuel-rich combustion case than in the fuel-lean combustion and stoichiometric ratio. Anggono et al. (2020) investigated the effect of CO₂ concentration on the laminar burning velocity and Markstein length of CH₄/air premixed flames at high pressure constant volume combustion. It was shown that there is a monotonic relationship between the Markstein length and the CO₂ dilution ratio and that the unstretched laminar burning velocity of the mixture decreased with increasing CO₂ concentration. Azatyan et al. (2010) reported the effect of CH₄, N₂, CO₂ and steam addition on the LBVs of H₂. The predictions showed that with the increase of H₂ doping ratio, the LBVs of H₂/Air monotonically decreased, but did not exceed 1.5 times, and the AFTs changed slowly. The effect of CO₂ and N₂ on the flame stability and LBVs of syngas/air was successfully performed by Burbano et al. (2011). Through experimental and numerical simulations, Burbano et al. (2011) showed that with the increase of CO₂ dilution doping ratio, the LBVs decreased obviously due to the decrease of heat release and the increase of heat capacity. This conclusion was consistent

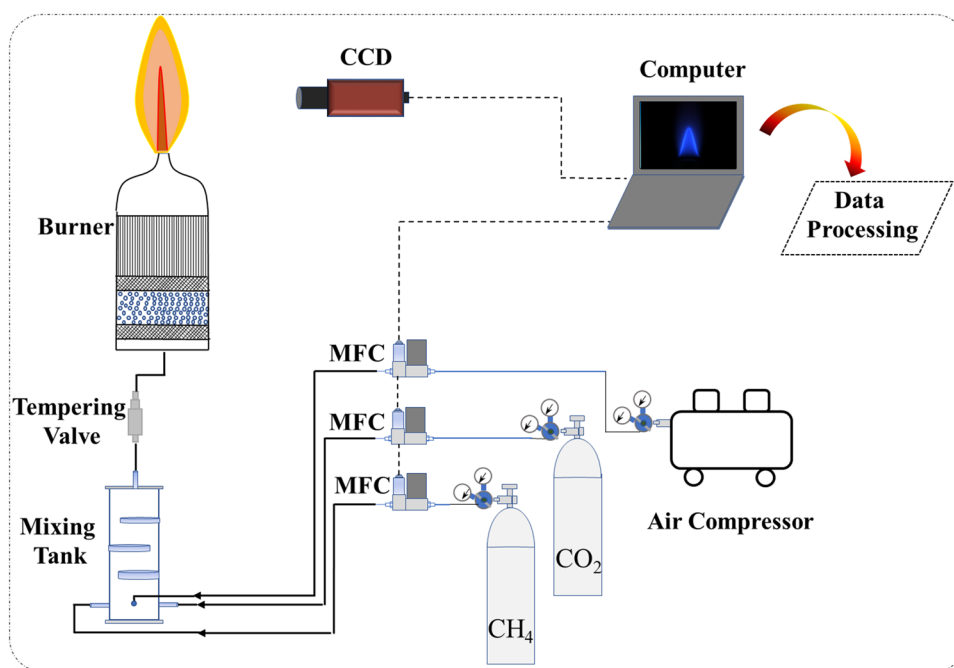
with the conclusion of Ref. Wang et al. (2012) measured the LBVs of CO/H₂/CO₂/O₂ by using spherical flame. The experimental results proved that H₂ and CO₂ have opposite effects on the LBV of syngas. Sun and Xu (2020) conducted the relationship between turbulent burning velocity and H₂ content in synthesis gas and revealed that turbulent burning velocity was a second-order polynomial of hydrogen volume fraction. Based on heat flux method and Bunsen burner, Wang et al. (2015) measured the LBVs of CO/H₂/N₂/CO₂ and numerically analyzed the effect of H and OH radicals on the LBV of syngas. The results showed that H had a linear relationship with LBVs. Nonaka and Pereira (2016) studied the effect of CO₂ addition in biogas on LBVs. The LBVs and other combustion characteristics of the natural gas blended with N₂, CO₂ and H₂O were carried out by Ren et al. (2019a, b, 2020). In addition, the effects of CO₂ on NO_x formation of methane combustion products were also conducted. Xiang et al. (2019, 2020) numerically investigated the chemical and physical effects of CO₂ and H₂ on LBVs and other laminar combustion characteristics of CH₄.

In view of the above considerations, the first objective is to investigate the effect of blending a wide range of CO₂ concentrations on methane combustion characteristics through a combination of experimental and numerical simulations. The second objective is to analyze the effect of CO₂ on NO_x generation during methane combustion, from the molar amount of NO_x to the temperature sensitivity of NO_x. There have been many studies on the effect of CO₂ on methane combustion characteristics, but few have addressed the effect of CO₂ blending into methane in terms of temperature sensitivity and NO_x generation. Therefore, this work conducts a study on the laminar combustion characteristics and NO_x emission of methane blended with CO₂.

2 Experiment setup

The laminar premixed flame experimental system is shown in Fig. 1. The experimental systems were mainly divided into: Bunsen burner, gas delivery system, gas supply system and safety system. The Bunsen burner mainly consists of a lamp body section and a lamp nozzle connected. The inner cavity of the lamp body section is equipped with a packing layer, a flow equalizing plate and a sintered net from bottom to top. The device plays a role in stabilizing the flow of the gas mixture flowing through it, and the flame ignited from the lamp nozzle is close to being conical, so that the detection accuracy of the flame propagation speed can be improved. The gas delivery system is completed by mass flow controller (MFC) and mixing tank. The brand of MFC is Sevenstar Huachuang CS200 series with an accuracy of 0.35% of full scale. The gas supply system contains N₂, CO₂, O₂ and CH₄ and the purity of

Fig. 1 Experimental apparatus for the combustion of CH₄/air mixtures with diluted CO₂



CH₄, N₂, CO₂ and O₂ are 99.99%. The safety system is tempering valve, which is to prevent the flame from flowing back to the mixing tank to explode. After the gas through the flowmeter and mixing tank, it passes the channel at the bottom of the burner, glass beads, flow equalization plates and multi-layer metal sintering net to form a stable conical flame at the nozzle. Besides, an industrial camera is used to photograph the flame image. The flame images are obtained by a CCD camera, model MANTA G-504C manufactured in Germany. The CCD camera chip is the Sony ICX655, the response frequency range is visible light. The image acquisition is realized by connecting the CCD camera port with the computer port through a Gigabit Ethernet cable. This model of CCD camera can be very good completion of the experimental process in the acquisition of flame images.

2.1 CO₂ doping ratio

CH₄/CO₂/N₂/O₂ is studied under premixed combustion at 298 K and 1 atm. The sum of CH₄ and dilution is 1, that is, the content of methane in mixtures decreases with increasing dilution gas content. Therefore, the general formula for calculating the doping ratio is:

$$\alpha = \frac{V_{\text{diluent}}}{V_{\text{diluent}} + V_{\text{fuels}}} \quad (1)$$

where V_{diluent} and V_{fuels} are volume fractions of dilution gas and fuel, respectively. α represents the proportion of dilution gas in the mixture.

Therefore, in the mixture, α is defined as the CO₂ doping ratio, and the formula is as follow:

$$\alpha = \frac{V_{\text{CO}_2}}{V_{\text{CO}_2} + V_{\text{CH}_4}} \quad (2)$$

where V_{CO_2} is the volume fraction of CO₂. V_{CH_4} denotes the volume fraction of methane in the mixture. In this paper, CH₄/O₂/N₂ mixed with CO₂ content of 0%–40% ($\alpha = 0, 5\%, 10\%, 15\%, 20\%, 25\%, 30\%, 35\%, 40\%$) is systematically analyzed.

The effect of CO₂ on the laminar combustion of methane is different, because of the different properties of CO₂ and N₂. During the experiment of mixing carbon dioxide with methane, we found that when the flow rates of methane were set at 280 mL/min and 260 mL/min, more experiments could be carried out. Therefore, in the experiments of mixing methane with carbon dioxide, the flow rates of methane are 280 mL/min and 260 mL/min. Table S1 in Supplementary information presents the experimental operation of CH₄/Air/CO₂ flame.

2.2 Laminar burning velocity calculation

Figure 2 shows the images obtained by CCD camera. The work conditions of $\Phi = 1.2$ are selected to show the images of methane mixed with carbon dioxide with doping ratios.

After capturing the images of premixed flame, the MATLAB code is used to calculate the LBVs of mixtures. The main steps to obtain the LBVs of CH₄ can be found in Ref

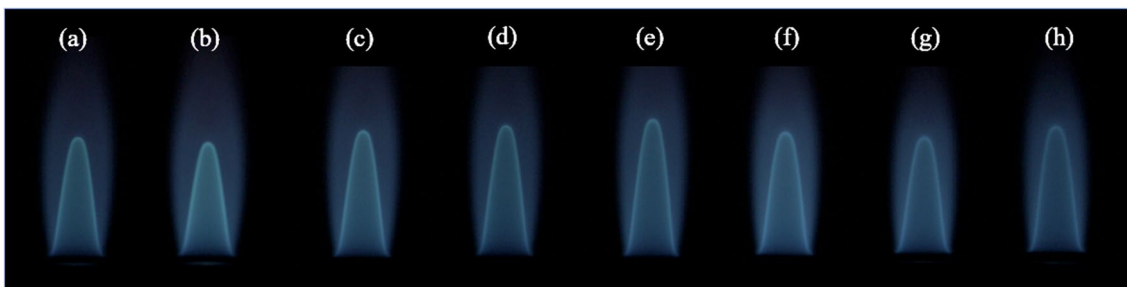


Fig. 2 Images obtained from CCD camera under methane/air with different CO₂ doping ratios. **a** $\alpha=5\%$. **b** $\alpha=10\%$. **c** $\alpha=15\%$. **d** $\alpha=20\%$. **e** $\alpha=25\%$. **f** $\alpha=30\%$. **g** $\alpha=35\%$. **h** $\alpha=40\%$

(Chu et al. 2021). Figure 3 gives the main steps to obtain the LBVs of CH₄ through the MATLAB code.

2.3 Error analysis

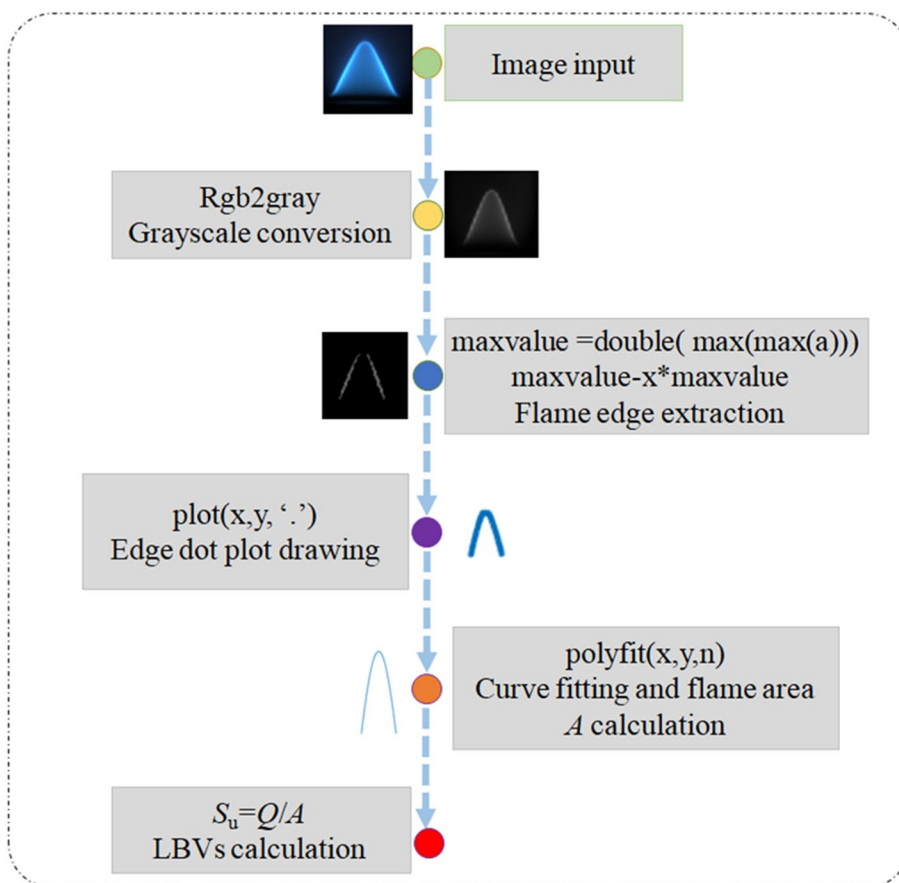
In the process of measuring the LBVs with the Bunsen burner, the measurement error is mainly caused by the error of the total gas flow rate (δ_Q) and the error generated when calculating the flame area (δ_A). The error of the total flow of the premixed gas is determined by the error of methane flow rate (δ_{CH_4}), and the accuracy of the methane flowmeter

is $\pm 2\%$ of the upset point, the error of the air flowmeter (δ_{air}), and the error of CO₂ flowmeter (δ_{CO_2}). The accuracy is $\pm 1\%$ of the set point. According to the error transfer principle, the error calculation method in Refs (Chu et al. 2021; Moffat 1988) can be used to calculate the total gas flow error:

$$\delta_Q = \sqrt{\delta_{CH_4}^2 + \delta_{air}^2 + \delta_{CO_2}^2} = \sqrt{2^2 + 1^2 + 1^2} = \sqrt{6} \quad (3)$$

The error of flame area is mainly determined by the resolution of the camera and the method of experimental photo processing. By calculating the adjacent points of the

Fig. 3 The major steps of calculating laminar burning velocity by MATLAB code



maximum gradient point in the inner boundary of the flame, the error is 4.5% approximately.

The total calculation error of LBV is:

$$\delta_L = \sqrt{\delta_Q^2 + \delta_A^2} = \sqrt{(\sqrt{6})^2 + (4.5)^2} \approx 5.1\% \quad (4)$$

The calculation error of laminar burning velocity is approximately 5.1% by calculating.

2.4 Modeling details

In the simulation, we choose the CHEMKIN-Pro/PRE-MIXED code to simulate the effect of CO₂ on methane laminar combustion. Two different reaction mechanisms are used for the calculations, the Aramco mech (Metcalf et al. 2013) and the GRI 3.0 mech (Smith et al. 2000) and these mechanisms are widely used for the calculations of small molecule hydrocarbon fuels. The Aramco mech contains 493 species and 2716 reactions and the GRI 3.0 mech contains 53 species and 325 reactions. Since the Aramco model does not include the NO_x component, the GRI 3.0 model is used to analyze the effect of CO₂ dilution on NO_x formation. The chemical reactions, thermal properties and transport properties database are imported using CHEMKIN format. The maximum grid number is set to 500, and the parameters chosen for this study ensured grid independence. GRAD and CURV are set to 0.04, and the iteration interval are -0.002 to 0.06 m. In addition, the Soret effect is added to the simulation. Multicomponent transport is incorporated into the reactor. Since the Aramco model does not include the NO_x component, the GRI 3.0 mech is used to analyze the effect of CO₂ dilution on NO_x formation.

3 Results and discussion

3.1 Experiment and mechanism verification

Figure 4 shows the LBVs comparison between GRI 3.0, Aramco and San Diego mech (Prince et al. 2017) under different equivalence ratios. GRI 3.0 mech and Aramco mech are in good agreement with the measured LBVs of this paper and Refs. (Gu et al. 2000; Bosschaart and Goey, 2004; Halter et al. 2005; Hu et al. 2009a, b; Hermanns et al. 2010; Dirrenberger et al. 2011; Mazas et al. 2011; Goswami et al. 2013; Akram et al. 2013; Li et al. 2015; Nonaka and Pereira 2016; Mitu et al. 2017; Okafor et al. 2018; Han et al. 2019; Wang et al. 2020). The yellow–brown represents lean mixture and light-cyan represents rich mixture. The predictions of San Diego mechanism (Prince et al. 2017) are in good agreement with the experimental results in fuel-lean combustion. However, when equivalence ratio $\Phi \geq 1$, the predictions of LBVs

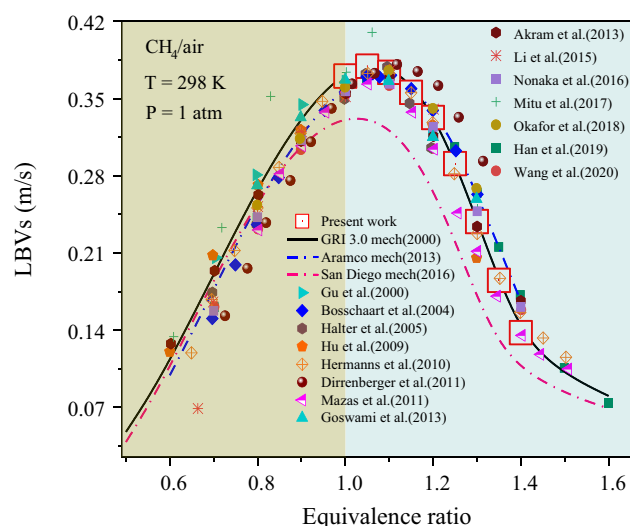


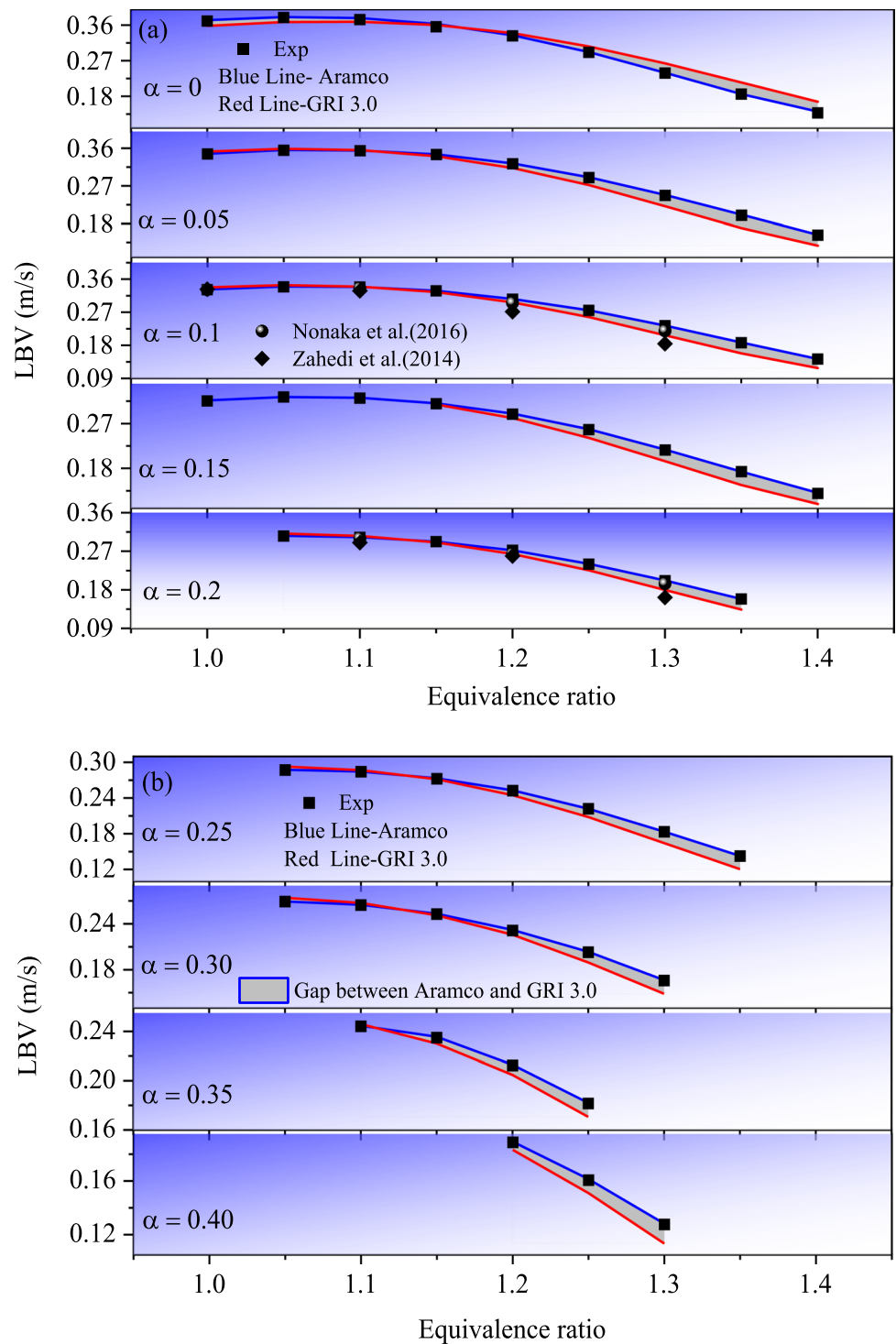
Fig. 4 Laminar burning velocities profile for CH₄/air mixture at 1 atm and 298 K for about 20 years

are lower than the experimental results. Therefore, the San Diego mechanism is not selected for prediction the combustion characteristics of methane in present work.

3.2 Effect of CO₂ addition on laminar burning velocity

In Fig. 5, the solid red line and blue line represent the simulated results of GRI 3.0 and Aramco mech individually. Black dots indicate the LBVs of CH₄ measured experimentally, and gray areas represent the prediction difference between the two mechanisms. From the results of experiments and mechanism predictions, it can be seen that the predicted LBVs of the two mechanisms are not much different when $\Phi \leq 1.15$, and the simulated values of GRI 3.0 mech are slightly larger than that of Aramco mech. However, when the $\Phi \geq 1.15$, the predictions of Aramco mech are higher than that of GRI 3.0 mech. Compared the predicted results of GRI 3.0 and Aramco mech with the experimental measured LBVs, it is found that the gap between experimental results and Aramco mechanism prediction results, and the errors between experiment and simulation are within 5% at same initial condition. This reveals that the process of measuring and calculating LBVs of CH₄ is accurate. As can be seen from Fig. 5, under the same CO₂ doping ratio, the LBVs ascend first and then descend with the increase of equivalence ratio, and when $\Phi = 1.05$, the LBVs reach the maximum. The LBVs became lower linearly as the CO₂ doping ratio increases. This is because the doping ratio of CO₂ increased, reducing the relative percentage of CH₄ in the CH₄ + CO₂ mixture. Another reason is that the specific heat capacity of CO₂ is relatively large, which will absorb part of the reaction heat (Burbano

Fig. 5 Comparison of the measured laminar burning velocity with the simulated values



et al. 2011), resulting in the reaction rate and temperature decreased. With the increase of CO₂ doping ratio, the percentage of activated molecules (CH₄ + O₂) in reactant decreases, which causes the effective collision times per unit time decrease. Besides, the movement of molecules is decelerated, the number of collisions between reactant

molecules per unit time presents a downward trend, and the reaction rate is decelerated. Thus, temperature and LBVs of CH₄ decrease. In addition, the Aramco mechanism simulation results are in good agreement with the other experiments (Nonaka and Pereira 2016; Zahedi and Yousefi 2014).

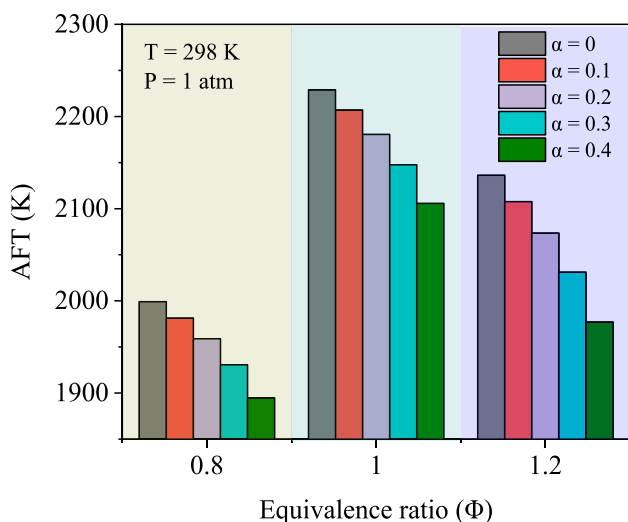


Fig. 6 Peak values of adiabatic flame temperatures change with different CO_2 doping ratios

3.3 Adiabatic flame temperature

Figure 6 shows the effect of CO_2 addition on peak values of AFTs under various equivalence ratios. It can be seen that at the same CO_2 doping ratio, the change trend of AFTs with equivalence ratio is similar to that of LBVs. For example, when CO_2 doping ratio is 0.1, the AFTs are 1981.30 K, 2207.15 K and 2107.74 K at equivalence ratios of 0.8, 1.0 and 1.2, respectively.

At the same CO_2 addition, the AFTs decrease with the increasing of equivalence ratio, and the change trend is the same as that of LBVs. The AFTs are descended with the increase of the CO_2 doping ratio because the content of methane decreases, which leads to lower combustion intensity between fuel and air.

3.4 Intermediates radicals

Combustion is a quite complex process, and a series of intermediates generated in the combustion processing of methane. Based on the above view, Aramco mech was selected for predicting the effect of CO_2 addition on intermediate radicals under different conditions. In order to study the changes of the intermediate radicals with different working conditions, the equivalence ratios of 0.8 (fuel-lean combustion), 1.0 (stoichiometric ratio) and 1.2 (fuel-rich combustion) are chosen for simulation, respectively.

3.4.1 Effect of CO_2 addition on CH_3

Figure 7 shows the change trend of mole fraction of intermediate radical CH_3 with various CO_2 contents under different equivalence ratios. CH_3 , as an important intermediate, the

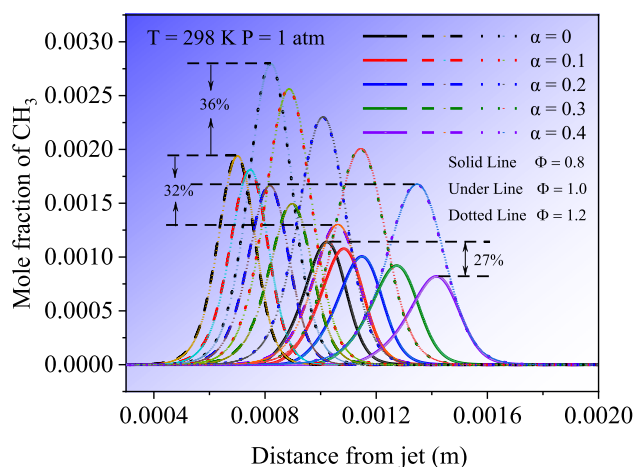


Fig. 7 Intermediate radical CH_3 changes with different CO_2 doping ratios

maximum mole concentration is reached in the beginning of the reaction, and completely reacts in a relatively short period of time and is no longer generated.

As shown in Fig. 7, firstly, keeping CO_2 doping ratio unchanged, the mole fraction of CH_3 showed a same trend with of the change of equivalence ratio. This is because the proportion of methane gradually increases with the increasing of equivalence ratio. Therefore, CH_3 generated in the oxidation process is gradually increasing. Because the oxidation reaction is the most intense in the slightly fuel-rich combustion ($\Phi = 1.05$), and the AFT is the highest and the LBV is the fastest, the time of CH_3 mole fraction reaching maximum is first advanced and gradually delayed with the increase of the equivalence ratio.

3.4.2 Effect of CO_2 addition on H

Figure 8 shows the effect of CO_2 addition on the H under different equivalence ratios. As the basic intermediate radical, H has an indispensable relationship with LBV and AFT. It reached the maximum mole concentration at beginning of the reaction and existed in the subsequent reaction process.

Compared with CH_3 , under the same equivalence ratio and CO_2 doping ratio, the maximum mole fraction of H appeared later than CH_3 . This illustrates that the reaction for H formation is later than CH_3 . Moreover, the mole fraction of H is much larger than that of CH_3 .

As can be seen from Fig. 8, mole fraction of H gradually increases with equivalence ratio becomes larger under the same CO_2 content. The maximum mole fraction shows a trend of advancing first and then postponing from fuel-rich to fuel-lean combustion, which is consistent with that of CH_3 . The existence of free radical H lasts longer than CH_3 . There maybe two reasons accounting for this. One is that H is decomposed by CH_4 continuously in the oxidation

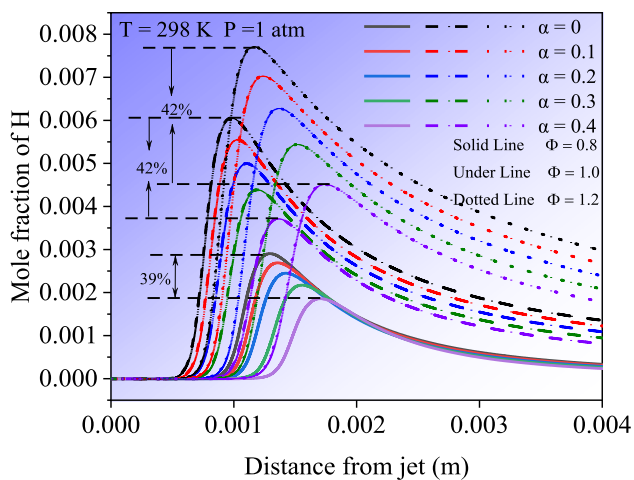


Fig. 8 Intermediate radical H changes with different CO₂ doping ratios

process. The other is that CH₃ also decomposes free radical H in the oxidation process, which makes radical H existed all the time in the whole oxidation process of CH₄.

3.4.3 Effect of CO₂ addition on OH

Figure 9 depicts the intermediate radical OH changes with different CO₂ doping ratios. OH exists all the time in the whole combustion process and gradually decreases, with a smaller decrease than that of H. It is speculated that O₂ continuously participate in the reaction with CH₄ during the combustion process, and oxidative decomposition to generate OH. Under the same CO₂ doping ratio, OH generation time is advanced and then delayed as the equivalence ratio increasing.

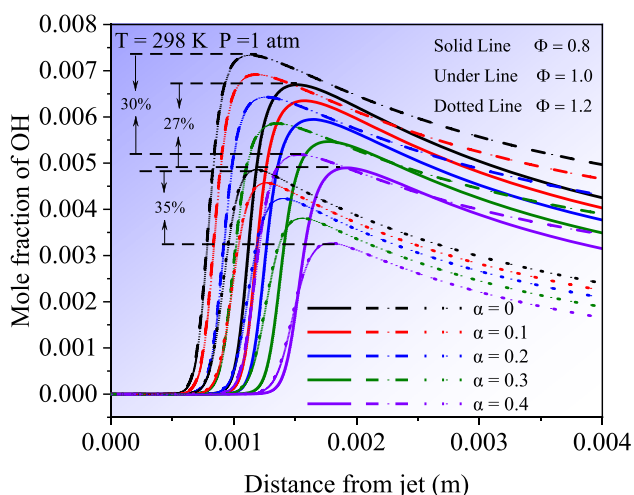


Fig. 9 Intermediate radical OH changes with different CO₂ doping ratios

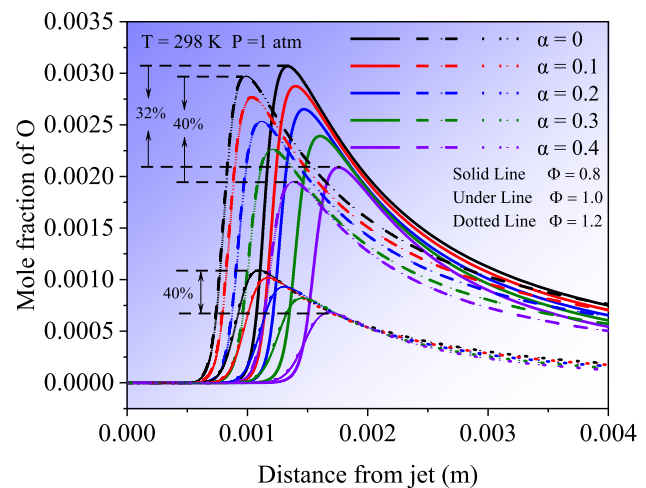


Fig. 10 Intermediate radical O changes with different CO₂ doping ratios

3.4.4 Effect of CO₂ addition on O

Figure 10 shows the changes of O at different CO₂ doping ratios and equivalence ratios. O as one of important intermediates, the time from the generation to the maximum mole fraction is very short, which is existed in the whole combustion process. From Fig. 10, it can also be found that at the same CO₂ blending ratio, the maximum mole fraction of O drops down slightly with the increasing of equivalence ratio. This is because as the equivalence ratio changes from 0.8 to 1.2, the methane keeps unchanged and the O₂ decreases gradually. Thus, the proportion of O in the combustion process decreases.

3.5 Effect of CO₂ addition on generation of NO_x

NO_x is main pollutant in the process of fuel combustion in internal combustion engine. The majority of nitrogen oxides emitted in combustion are NO, followed by NO₂ and N₂O. NO and NO₂ can be involved in the formation of acid rain and photochemical smog in the atmosphere. N₂O is also a pollutant of interest in recent years, as it is considered a typical greenhouse gas. Therefore, it is great significance to investigate the effect of CO₂ on NO_x formation during methane combustion for high efficiency and low pollution combustion. Because Aramco mech does not contain NO_x, the influence of CO₂ on methane NO_x formation is still predicted by GRI 3.0 mech. Fig. S1 in Supplementary information depicts the NO generation and AFTs of the article by using GRI 3.0 mech compared with the results of other simulation mechanisms. Fig. S2 in Supplementary information shows the NO_x generation of methane mixed with different CO₂ doping ratio when $\Phi = 0.8, 1.0$ and 1.2.

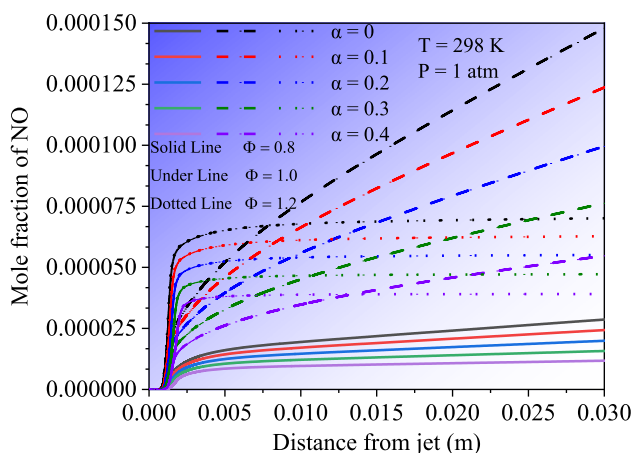


Fig. 11 NO emissions changes with different CO₂ doping ratios

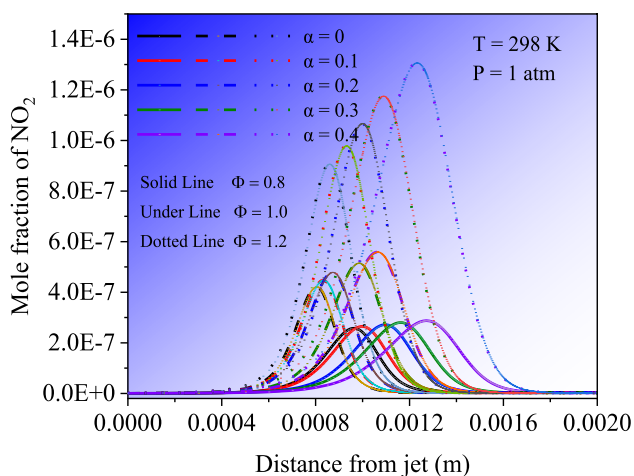


Fig. 12 NO₂ emissions changes with different CO₂ doping ratios

3.5.1 Effect of CO₂ addition on generation of NO

Figure 11 depicts the NO generation under equivalence ratios of 0.8, 1.0 and 1.2 with various CO₂ addition. According to Fig. 11, it can be analyzed that the total NO generation decreases with the increase of CO₂ addition. It is presumed that the generated NO_x is thermal NO_x.

As can be seen from Fig. 11, the equivalence ratio increases from 0.8 to 1.0, and the NO generation amount increases by 5 times. It is speculated that the equivalence ratio increases from 0.8 to 1.0, and the adiabatic flame temperature gradually increases, which results in an increase in thermal NO_x.

3.5.2 Effect of CO₂ addition on generation of NO₂

Figure 12 shows the effect of various CO₂ doping ratio on NO₂ emissions at different equivalence ratios. NO₂, as an

intermediate, its variation trend is similar to CH₃, but the difference is that the amount of NO₂ increases monotonously with the increase of the amount of CO₂. Keeping equivalence ratio unchanged, the maximum mole fraction of NO₂ increases and the corresponding time of reaching maximum mole fraction later with the increase of CO₂. When $\Phi = 0.8$, maximum mole fraction of NO₂ and the time of reaching the maximum mole fraction do not change much. The highest concentration and the time of reaching highest concentration is changed obviously while $\Phi = 1.2$.

3.5.3 Effect of CO₂ addition on generation of N₂O

N₂O is a typical greenhouse gas, but the combustion rate can increase by the efficient use of N₂O. Figure 13 depicts the changes of N₂O generation at different equivalence ratios and CO₂ doping ratios. It can be seen that N₂O is an intermediate with a low mole fraction. The generation time and the time of reaching the highest mole fraction is delayed by blending N₂O, and the highest mole fraction increases.

At any equivalence ratio, the maximum N₂O mole fraction has a similar trend with the increase of CO₂ dilution. However, in the subsequent combustion process, the mole fraction decreases as the increasing of blending CO₂.

3.6 Sensitivity analysis

In order to further reveal the impact of blending CO₂ on intermediate radicals and NO_x emissions, the maximum gradient point of temperature is chosen for sensitivity analysis of the effect of CO₂ addition on CH₄ combustion (Dong et al. 2020, 2021). Armaco mech is used to analyze the effect of CO₂ addition on intermediate radicals and GRI 3.0 mech is used to analyze the effect of CO₂ addition on NO_x.

3.6.1 The sensitivity analysis of NO

Figure 14 describes the effect of CO₂ blending on NO production of methane combustion products under different equivalence ratios. The starting path of thermal NO_x is R178 N + NO = N₂ + O and N free radicals are quickly consumed. From the sensitivity analysis of NO_x, it can be seen that the effect on NO production is greater than that of inhibition. R38 H + O₂ ⇌ O + OH and R240 CH + N₂ ⇌ HCN + N mainly promote the generation of NO. R35 H + O₂ + H₂ ⇌ HO₂ + H₂O, R125 CH + O₂ ⇌ O + HCO and R52 CH₃ + H (+M) ⇌ CH₄ (+M) have strong inhibitory effect on NO production at fuel-lean, stoichiometric ratio and fuel-rich combustion.

For the same elementary reaction, such as R38 H + O₂ ⇌ O + OH, the change trend of sensitivity coefficient is consistent with the equivalence ratio. When $\Phi = 1$, the sensitivity coefficient of R125 CH + O₂ ⇌ O + HCO decreases with

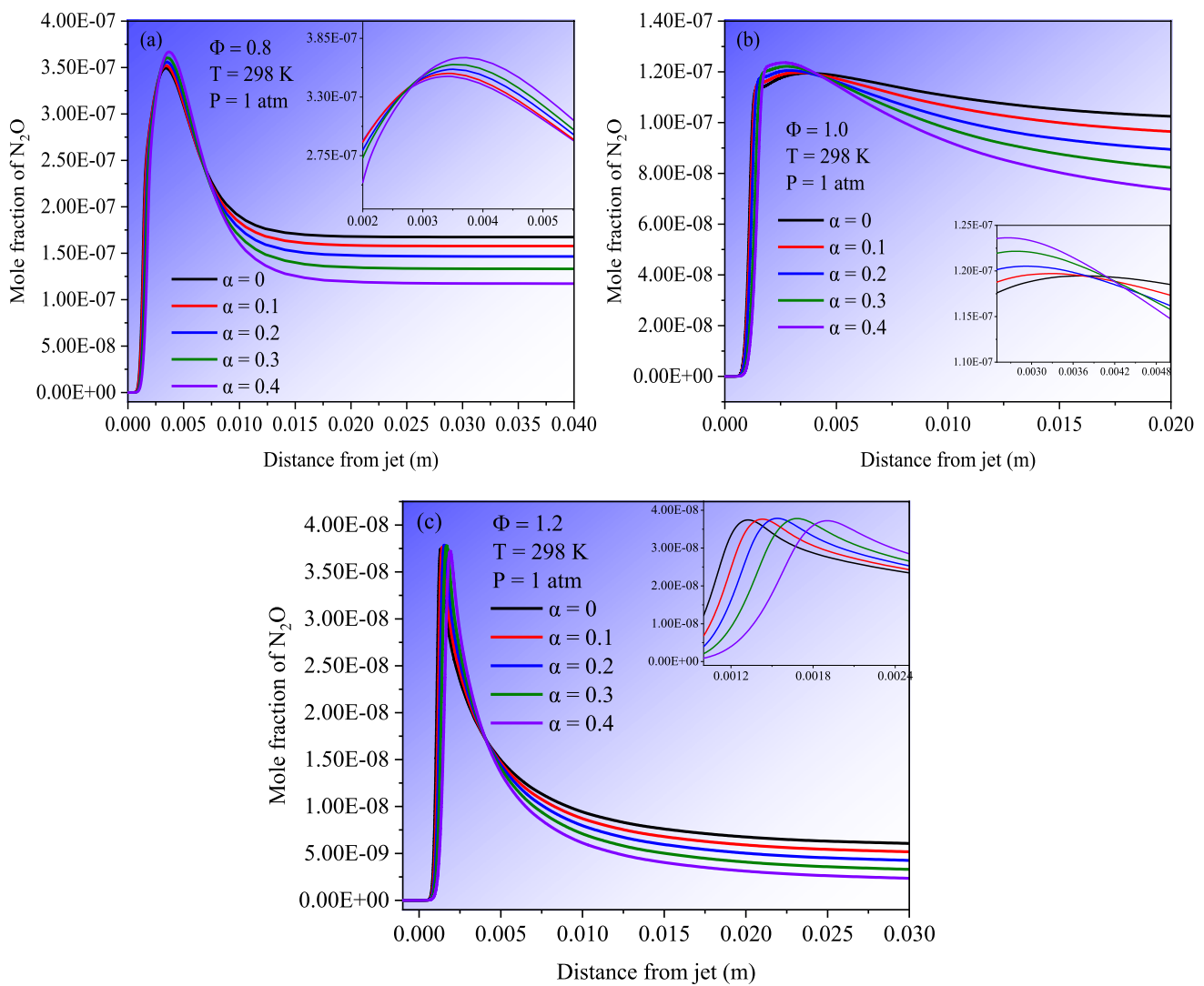


Fig. 13 N₂O emissions changes with different CO₂ doping ratios

the CO₂ doping ratio increases. Different from the change of intermediate radicals, for the same elementary reaction, taking R38 $\text{H} + \text{O}_2 \rightleftharpoons \text{O} + \text{OH}$ as an example, the sensitivity coefficient increases continuously with the increase of equivalence ratio, rather than a trend of first increasing and then decreasing.

3.6.2 The sensitivity analysis of NO₂

As can be seen from Fig. 15, different from the effect on NO, R38 $\text{H} + \text{O}_2 \rightleftharpoons \text{O} + \text{OH}$ is to inhibit NO₂ generation. R52 $\text{CH}_3 + \text{H} (+\text{M}) \rightleftharpoons \text{CH}_4 (+\text{M})$ has dominant effect on promoting NO production at all equivalence ratio. In the top 10 elementary reactions, the number of elementary reactions promoting NO₂ generation increases first and then decreases with the increase of equivalence ratio.

At any equivalence ratio, the sensitivity coefficients of R38 $\text{H} + \text{O}_2 \rightleftharpoons \text{O} + \text{OH}$ and R52 $\text{CH}_3 + \text{H} (+\text{M}) \rightleftharpoons \text{CH}_4 (+\text{M})$ increases with the increase of CO₂ doping ratio. And the CO₂ addition increases, the increase amplitude is also enhanced.

3.6.3 The sensitivity analysis of N₂O

As can be seen from Fig. 16, R38 $\text{H} + \text{O}_2 \rightleftharpoons \text{O} + \text{OH}$ promotes N₂O generation at all equivalence ratios, which is the reaction with the largest sensitivity coefficient among the top 10 elementary reactions. When the Φ is less than or equal to 1.0, R183 $\text{N}_2\text{O} + \text{H} \rightleftharpoons \text{N}_2 + \text{OH}$ mainly inhibits N₂O formation, and the sensitivity coefficient does not change much with the increase of the CO₂ ratio. R52 $\text{CH}_3 + \text{H} (+\text{M}) \rightleftharpoons \text{CH}_4 (+\text{M})$ is inhibited the generation of N₂O at $\Phi = 0.8$,

Fig. 14 Sensitivity coefficients with respect to the elementary reactions for NO under different CO₂ doping ratios

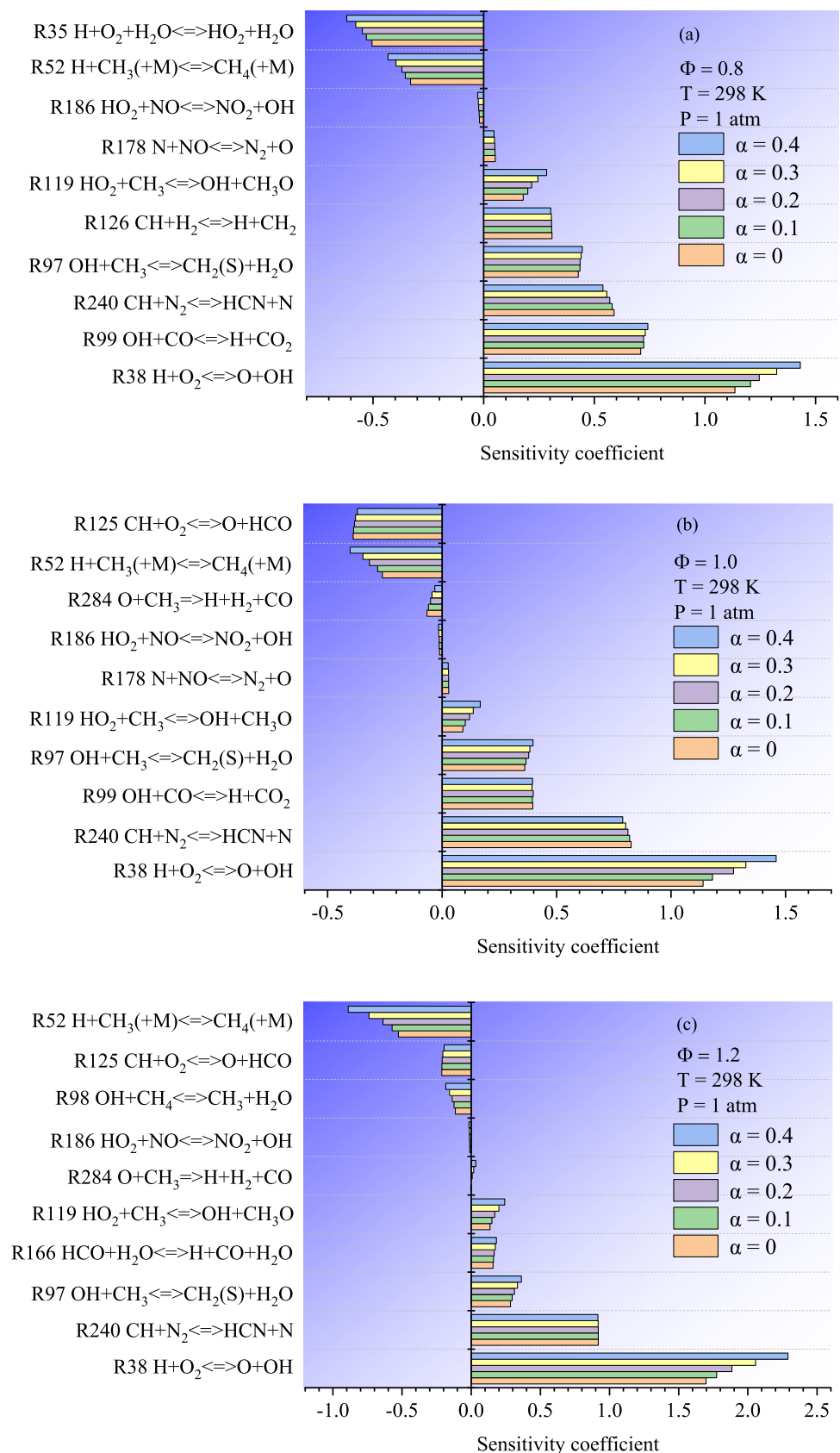


Fig. 15 Sensitivity coefficients with respect to the elementary reactions for NO₂ under different CO₂ doping ratios

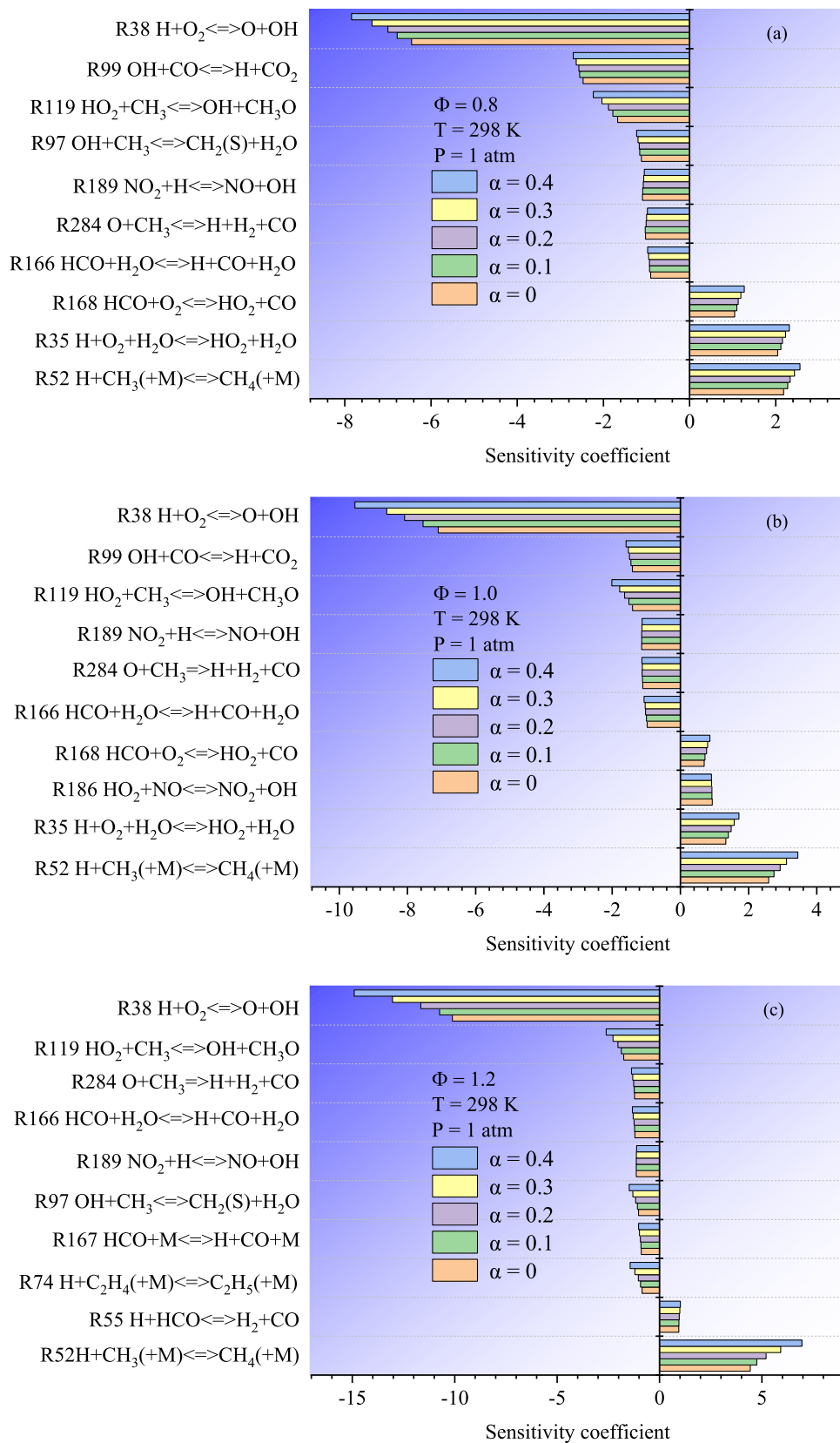


Fig. 16 Sensitivity coefficients with respect to the elementary reactions for N_2O under different CO_2 doping ratios

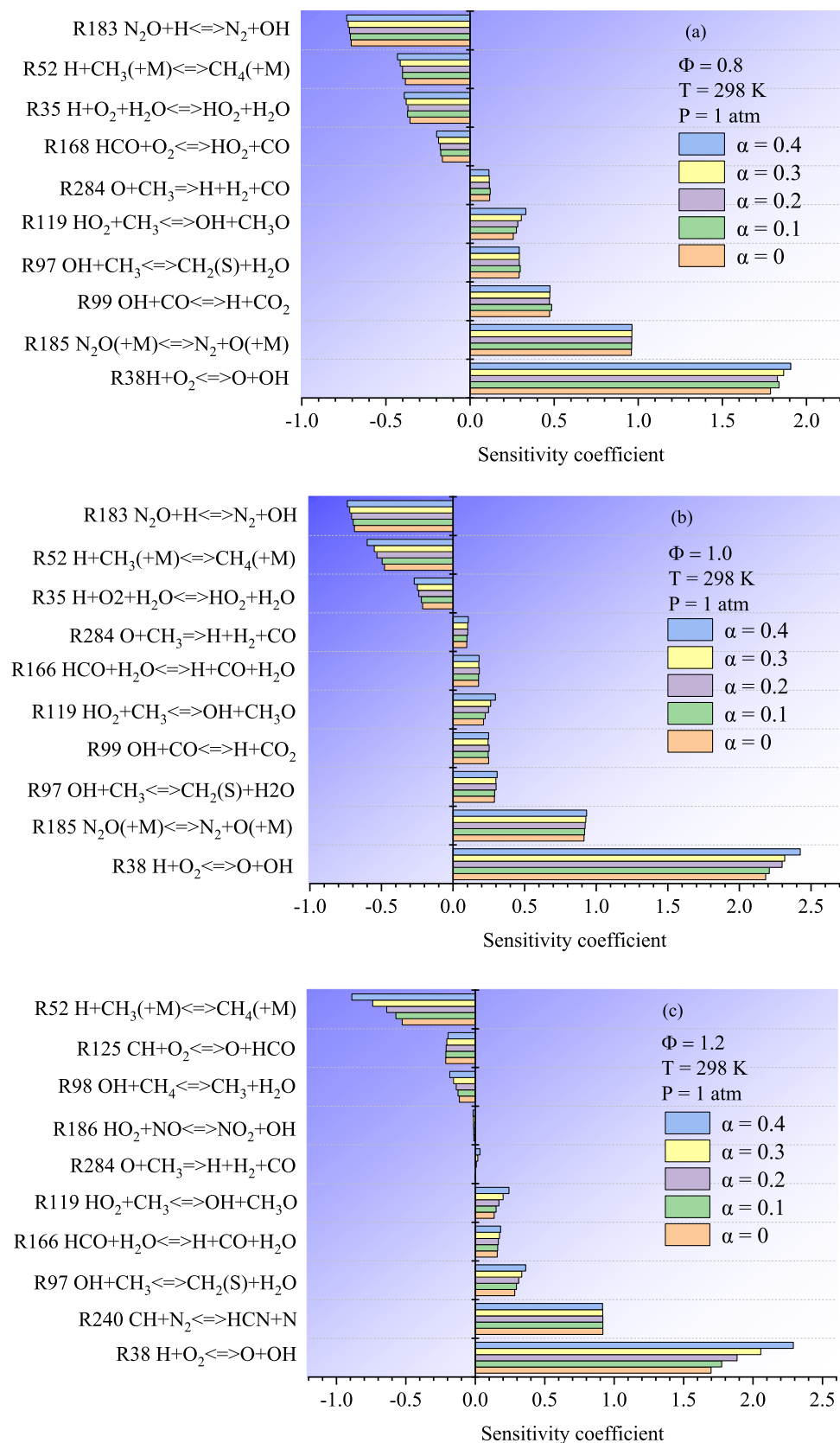
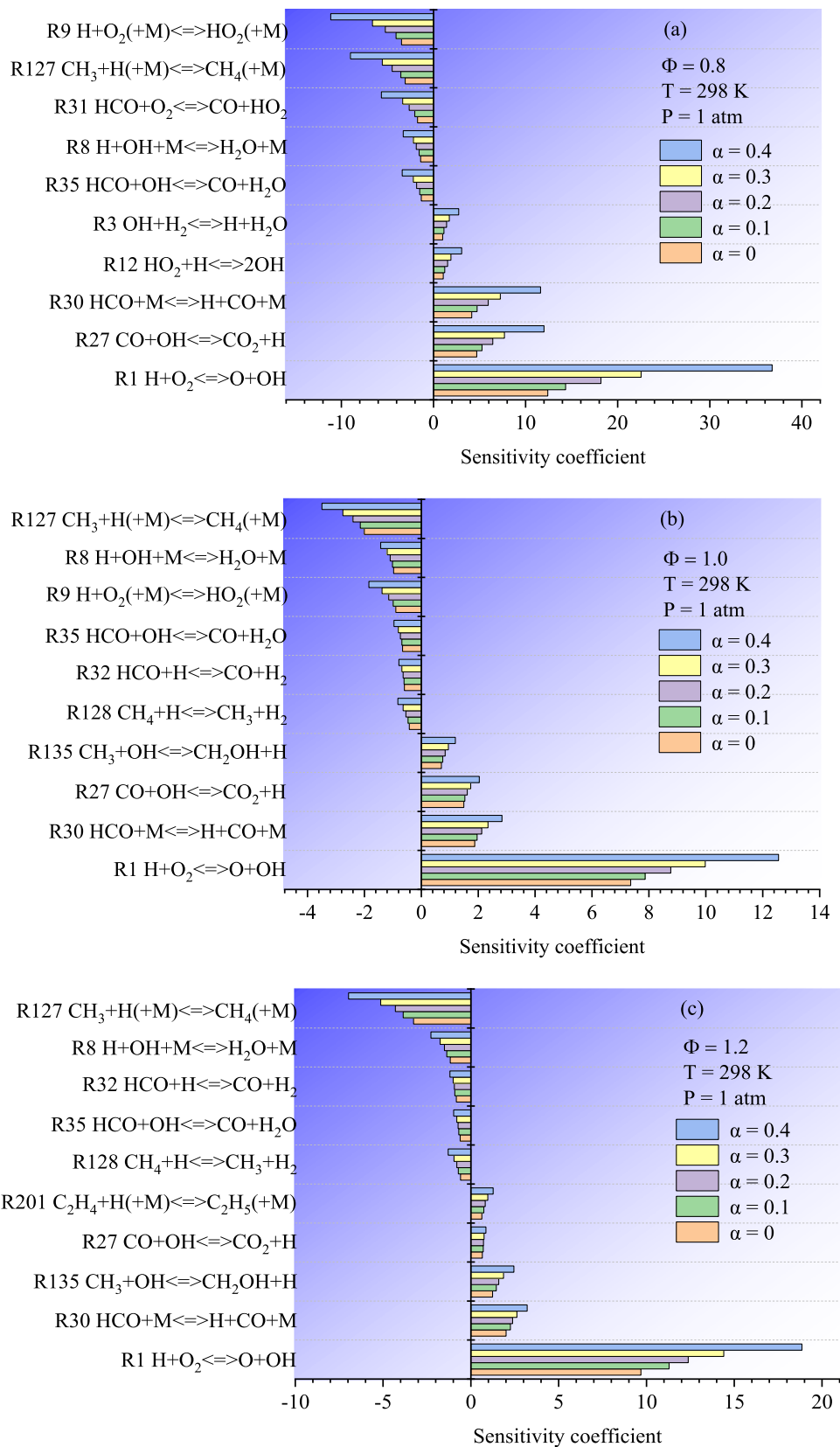


Fig. 17 Sensitivity coefficients with respect to the elementary reactions for CH₃ under different CO₂ doping ratios



1.0 and 1.2, and the sensitivity coefficient increases with increasing CO₂ addition.

3.6.4 The sensitivity analysis of CH₃, H, OH and O

According to the results, when the CO₂ doping ratio stays unchanged, the sensitivity coefficient decreases first and then increases as the increasing of equivalence ratio. In the top 10 reactions that have a great influence on formation of CH₃, the sensitivity coefficient of reactions that promote the generation of CH₃ with the increase of the equivalence ratio decreases and then increases. The change trend is opposite to that of LBVs, AFTs, intermediate radicals.

At the same equivalence ratio, it can be seen in Fig. 17 that the CH₃ sensitivity coefficient increases with the increase of the CO₂ addition, and the increase in sensitivity coefficient becomes larger with the increase of CO₂ addition. This is because with the increasing of CO₂ addition, the decrease in AFT gradually increasing and the sensitivity coefficient becomes larger, which means the reactions become more sensitive with temperature change. The generation of CH₃ is promoted by R1 $H + O_2 \rightleftharpoons O + OH$. When $\Phi = 0.8$, R9 $H + O_2(+M) \rightleftharpoons HO_2(+M)$ plays the main role of inhibition. When Φ is greater than or equal to 1, R127 $CH_3 + H(+M) \rightleftharpoons CH_4(+M)$ has a negative effect on generation of CH₃.

Figures S3–S5 in Supplementary information report the H, OH and O sensitivity coefficients with respect to the elementary reactions, respectively.

4 Conclusions

In this paper, CO₂ is used as an additive to study its blending on the methane laminar combustion characteristics and the NO_x emission characteristics. The study starts from studying the effect of blending a wide range of CO₂ concentrations on the methane combustion characteristics through a combination of experiments and numerical simulations. The second mission is to analyze the effect of CO₂ on NO_x production during methane combustion from the molar amount of NO_x to the temperature sensitivity of NO_x. The reliability of the simulation results is verified by comparing the laminar burning velocity measured by Bunsen burner with CHEMKIN simulations, with the main parameter being the laminar combustion velocity. With the increase of equivalence ratio, laminar burning velocity increases first and then decreases. The laminar burning velocity reaches the maximum when the equivalence ratio is 1.05. GRI 3.0 mech and experimental results have good agreement in measuring the laminar burning velocity of pure CH₄, but after blending CO₂, the laminar burning velocity predicted by the Aramco

mechanism is highly fitted to the experimental results. The predictions of the two mechanisms are different. When Φ is less than or equal to 1.15, the prediction of GRI 3.0 mech is greater than that of Aramco mech, but when $\Phi > 1.15$, the predicted result by Aramco mech is greater than the simulated value of GRI 3.0 mech. At any equivalence ratio, the laminar burning velocity of CH₄ decreases with the increase of CO₂ addition. Blending carbon dioxide absorbs the heat of reaction in the combustion process, resulting in a decrease in the adiabatic flame temperature. Aramco mech is selected to analyze the effect of CO₂ addition to the adiabatic flame temperature, intermediate radicals, and the sensitivity analysis of the intermediate radicals, it was found that adiabatic flame temperature and the maximum mole fraction of the intermediate radicals decreases after blending CO₂. Through sensitivity analysis, it is found that the elementary reaction $H + O_2 \rightleftharpoons O + OH$ always promotes the generation of free radicals, and the sensitivity coefficient of the elementary reaction gradually decreases with the increase of CO₂ doping ratio. The effect of CO₂ on the NO_x emissions (NO, NO₂ and N₂O), and sensitivity analysis of NO_x formation is predicted by GRI 3.0 mech. The results of numerical simulation show that the amount of NO_x generates or the maximum mole fraction point decreases by blending CO₂. The elementary reaction $H + O_2 \rightleftharpoons O + OH$ also always promotes NO_x emission. The results of this study show the effect of CO₂ on methane combustion characteristics and NO_x emission, which provides insight for further effective utilization of CO₂. However, subsequent studies can be conducted by only verifying the effect of CO₂ on methane, and subsequent studies should be conducted for the blending of CO₂ with macromolecular hydrocarbon fuels and oxygenated fuels.

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Declarations

Competing interest The authors declare that they have no conflict of interests regarding the publication of this paper.

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