



# Chemical effect of hydrogen peroxide addition on characteristics of methane–air combustion



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## ABSTRACT

The effects of hydrogen peroxide addition on the reaction pathway of premixed methane/air flames are numerically investigated using the PREMIX code with the GRI-Mech 3.0 chemical kinetic mechanisms and detailed transport properties. Hydrogen peroxide is used as the oxidizer substituent of air. Results show that the laminar burning velocity and adiabatic flame temperature of premixed methane–air flame are significantly increased with H<sub>2</sub>O<sub>2</sub> addition. The addition of hydrogen peroxide increases not only all the reaction rates of intermediate species, but also the concentrations of intermediate species. The traditional reaction pathways of CH<sub>4</sub>/air flame are altered by the addition of hydrogen peroxide, due to the enhanced production of OH and HO<sub>2</sub>. The enhanced OH radicals promote HO<sub>2</sub> productions through reaction (R89). The increased HO<sub>2</sub> accelerates the progressive reaction of CH<sub>3</sub> to form CH<sub>3</sub>O and then CH<sub>2</sub>O.

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## 1. Introduction

Plenty of flame-stabilized strategies have been investigated and applied in macro- and micro-scale combustion systems for decades [1]. For example, coaxial dump combustor [2] and swirler [3,4] are extensively utilized in combustion systems to increase flow residence time as well as fuel–air mixing, and hence to improve flame stability by means of flow structure modification. Besides, heat exchanger [5], flue gas recirculation mechanism [6] and turbo-charger [7] are generally applied in engines and furnaces to increase temperature and pressure in the flow field and to extend flammability limits. On the other hand, by employing catalyst to accelerate chemical reaction [8,9] or by imposing microwave [10] or plasma [11] to induce active chemical radicals (H, OH) in flames are also the well-known strategies to improve combustion behavior. Nevertheless, these approaches result in extra expenses for assembling additional mechanical auxiliaries in combustion system and retrofitting the mechanical design of combustor configuration. Besides, the operational procedure of the retrofitted combustion

system and the optimum operating range of the combustor are also needed to be re-modified.

In order to minimize the modification of combustion system, an attractive alternative is to employ active fuels such as hydrogen or to add strong oxidants such as hydrogen peroxide to fuels to enhance combustion. Accordingly, the addition of hydrogen [12–14], hydrogen peroxide [15,16] or nitrous oxide [17,18] in hydro-carbon fuels has received increasing attention recently. Wang et al. [12] numerically studied the effect of hydrogen addition on methane–air mixture combustion. They found that the mole fractions of major species, CH<sub>4</sub>, CO, and CO<sub>2</sub>, are decreased and the production rates of the dominant reactions contributing to these major species are increased remarkably as hydrogen is added. Wang et al. [13,14] experimentally investigated the effects of hydrogen addition on combustion and emission performance of a spark-ignited gasoline engine. They found that flame development and propagation periods of the gasoline engine are shortened with the increase of hydrogen blending ratio. The measured tailpipe HC, CO, and CO<sub>2</sub> emissions are reduced whereas the NO<sub>x</sub> emissions are increased with hydrogen addition. Among these energetic additives, hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) is particularly an environmentally friendly oxidant with strong oxidability and it appears as liquid state in normal temperature with benefits of easy-to-store and recharging.

A few recent studies have indicated that hydrogen peroxide is a promising additive for improving practical combustion process

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[19–22]. Golovitchev et al. [19,20] found that the improvement of methane auto-ignition with hydrogen peroxide addition is much stronger than that with hydrogen. Kim et al. [21] and Martinez et al. [22] discovered that the presence of hydrogen peroxide could evidently reduce the concentration of unburnt hydrocarbons, CO and NO<sub>x</sub>. Furthermore, Ting and Reader [15] as well as Chen et al. [16] numerically investigated the effects of hydrogen peroxide on the premixed methane–air flame under various concentrations of H<sub>2</sub>O<sub>2</sub> additions. They found that the adiabatic temperature and laminar burning velocity of the premixed methane–air flames are increased due to the increase of OH, H, and O radical concentrations. It is conjectured that the existence of hydrogen peroxide leads to the shift of chemical reaction pathways, and to further produce chemical radicals. In order to further understand the flame structure of methane–air flame with hydrogen peroxide addition and the roles of hydrogen peroxide on methane reactions, the reaction pathways under the condition of partial replacement of air with hydrogen peroxide are investigated.

## 2. Numerical model and chemical mechanism

In this study, the adiabatic, unstrained, free propagation velocities of the laminar premixed CH<sub>4</sub>/air/H<sub>2</sub>O<sub>2</sub> flames are calculated by the PREMIX code of CHEMKIN Collection. The PREMIX code solves the equations governing steady, isobaric, quasi-one-dimensional flame propagation. For a freely propagating flame, the mass flow rate is an eigenvalue and must be determined as part of the solution. An additional constraint is required and a flame-fixed coordinate system is established by fixing the temperature at 500 K. Temperature and species gradients at the boundaries should be avoided to obtain the accurate flame speed. It means that the boundaries should be sufficiently far from the flame. In this work, the adiabatic flame temperature is calculated by the EQUIL code of CHEMKIN Collection. An initial reactant mixture is specified with the constraints of constant enthalpy and constant pressure. To obtain accurate adiabatic flame temperature, besides reactants and products, all radical species that might occur in the flame are also included.

The GRI-Mech 3.0 mechanism composing of 53 chemical species and 325 reaction steps is used for the CH<sub>4</sub>/air/H<sub>2</sub>O<sub>2</sub> flame calculations. Detailed thermal and transport properties are also included. This mechanism has been used satisfactorily to simulate non-catalytic H<sub>2</sub>O<sub>2</sub> decomposition [23]. The reaction rate constant is represented by the modified Arrhenius expression,

$$k = \bar{A}T^b \exp\left(\frac{-E_a}{RT}\right) \quad (1)$$

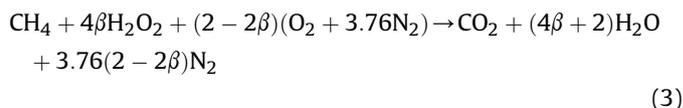
where  $\bar{A}$  is the pre-exponential factor,  $b$  is the temperature exponent, and  $E_a$  is the activation energy. The chemical kinetics with CHEMKIN format is used in the code. Details of the chemical reaction rate formulation and CHEMKIN format can be found in the user's manual [24].

At the inlet boundary, the unburned reactants are supplied at 423 K and 1 atm. This selected temperature is the boiling temperature of H<sub>2</sub>O<sub>2</sub> and it is assumed that all reactants at this temperature are in gas phase. Hydrogen peroxide is used as the oxidizer substituent by partial replacement of air. One mole H<sub>2</sub>O<sub>2</sub> can be decomposed to produce half mole O<sub>2</sub> and 1 mol H<sub>2</sub>O. Therefore, the global reaction for the stoichiometric CH<sub>4</sub>/H<sub>2</sub>O<sub>2</sub> mixture is defined as:



In the case of using H<sub>2</sub>O<sub>2</sub> to replace partial air, the total amount of O<sub>2</sub> is maintained to keep the constant fuel/oxidizer ratio.

Therefore, the reduced O<sub>2</sub> from air is supplied from the decomposed H<sub>2</sub>O<sub>2</sub>. For the stoichiometric condition, the global reaction is defined as:



where  $\beta$  is the replacement percentage of air by H<sub>2</sub>O<sub>2</sub>. The reduction of air also leads to reduction of N<sub>2</sub> in the oxidizer stream.

In order to validate the setting of boundary conditions and the chemical mechanisms used in the present study, we calculated the laminar burning velocity of premixed methane–air flame with 20% additions of hydrogen peroxide in the range of equivalence ratios from 0.6 to 1.1. The results are compared with those calculated by Ting and Reader [15] as shown in Fig. 1. We found that the agreements between both results are very good. These results verify the correct setting of boundary conditions and the suitable chemical mechanisms used in the present study.

## 3. Results and discussion

### 3.1. Characteristics of CH<sub>4</sub>/air/H<sub>2</sub>O<sub>2</sub> flame

To examine the effects of H<sub>2</sub>O<sub>2</sub> on premixed methane flames, the numerical results of premixed stoichiometric CH<sub>4</sub>/air and CH<sub>4</sub>/50% air + 50% H<sub>2</sub>O<sub>2</sub> are compared for illustration. Fig. 2 shows the resultant temperature and some of the important species concentration profiles. With 50% of air replaced by hydrogen peroxide, the adiabatic flame temperature increases approximately by 400 K due to the reduction of nitrogen dilution and heat release from thermal decomposition of hydrogen peroxide. For pure air case, the reactant CH<sub>4</sub> is completely consumed within 1.0 mm of the spatial coordinate but 0.78 mm for the 50% H<sub>2</sub>O<sub>2</sub> replacement case. Hydrogen peroxide enhances methane consumption. In addition, the H<sub>2</sub>O<sub>2</sub> decomposition results in an increase of H<sub>2</sub>O production. The addition of hydrogen peroxide significantly increases CO formation, but produces similar amount of CO<sub>2</sub>. Fig. 2 also shows that the mole fraction of OH, H and O increases with increasing hydrogen peroxide addition. Especially, the increasing trends of HO<sub>2</sub>, HCO,

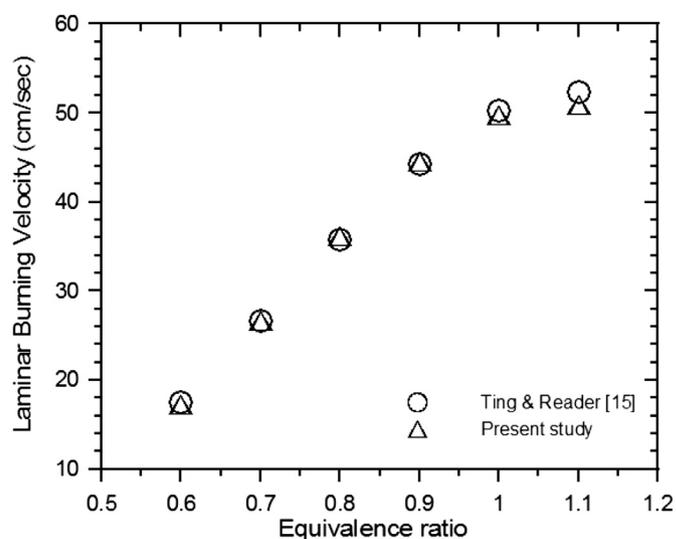


Fig. 1. Comparison of the calculated laminar burning velocity of premixed methane/air flames with 20% addition of H<sub>2</sub>O<sub>2</sub> in the range of equivalence ratios from 0.6 to 1.1 with those predicted by Ting and Reader [15].

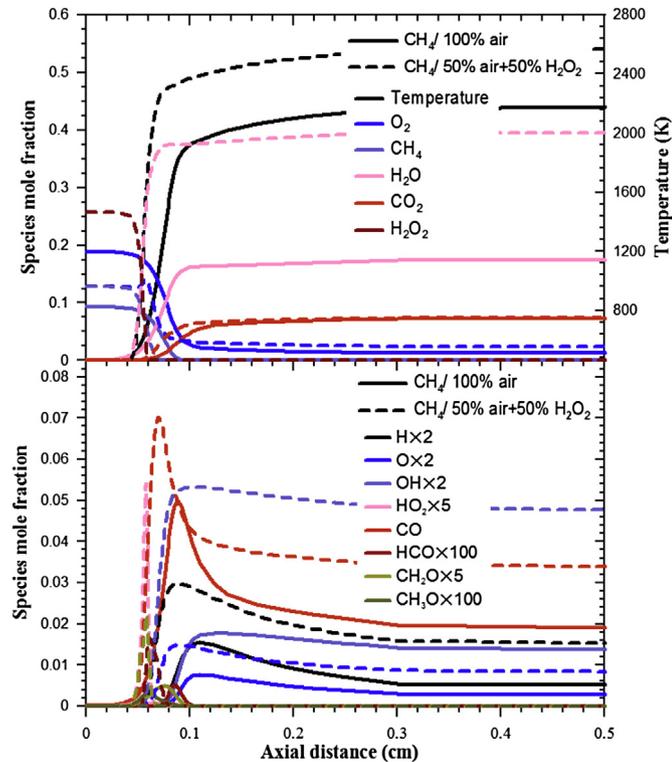


Fig. 2. Effect of hydrogen peroxide addition on temperature and species concentration of stoichiometric  $\text{CH}_4/\text{air}/\text{H}_2\text{O}_2$  flames. The solid lines are for  $\text{CH}_4/100\%$  air and the dashed lines represent  $\text{CH}_4/50\%$  air +  $50\%$   $\text{H}_2\text{O}_2$ .

$\text{CH}_2\text{O}$  and  $\text{CH}_3\text{O}$  are more significant. These facts suggest that the dominant reactions of methane combustion are altered by  $\text{H}_2\text{O}_2$  addition. Hydrogen peroxide decomposition increases the active radicals, enhances the reaction rate, and then accelerates the laminar burning velocity.

### 3.2. Burning velocity of $\text{CH}_4/\text{air}/\text{H}_2\text{O}_2$ flame

Fig. 3 shows the effect of replacement of air with  $\text{H}_2\text{O}_2$  on the laminar burning velocity and adiabatic flame temperature at the stoichiometric condition. It can be seen that the laminar burning velocity is increased with increasing the percentage of hydrogen peroxide addition. The laminar burning velocity is about  $0.71$  m/s

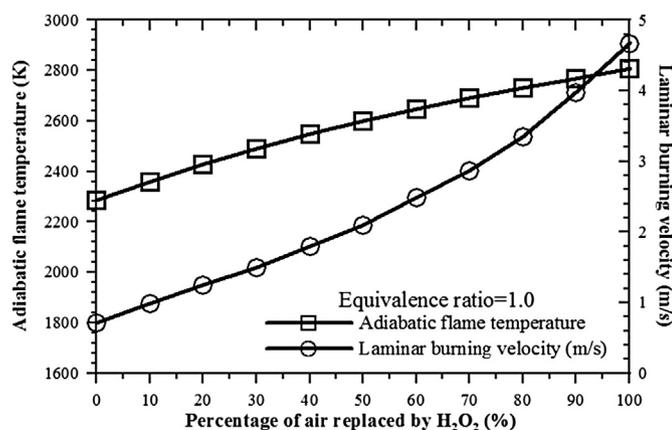


Fig. 3. Computed laminar burning velocity and adiabatic flame temperature of methane/air flames with different percentages of air replaced by  $\text{H}_2\text{O}_2$ .

for the stoichiometric  $\text{CH}_4/\text{air}$  flame at an inlet temperature of  $423$  K and it is increased to about  $2.1$  m/s with  $50\%$  of air replaced by  $\text{H}_2\text{O}_2$ . When air is completely replaced by  $\text{H}_2\text{O}_2$ , the laminar burning velocity approaches  $4.7$  m/s. This significant increase is because that the oxidizer is completely provided from decomposition of hydrogen peroxide and hydrogen peroxide dominates the reaction rate of methane oxidation. Fig. 3 also shows that the adiabatic flame temperature increases with increasing the percentage of  $\text{H}_2\text{O}_2$  addition. The maximum temperature increase is up to  $520$  K when air is completely replaced by  $\text{H}_2\text{O}_2$ . The increase of adiabatic temperature is primarily due to heat release from hydrogen peroxide decomposition. The maximum heat release rate is about  $6.09 \times 10^9$  J/m<sup>3</sup> s for stoichiometric  $\text{CH}_4/\text{air}$  flame. For the case of stoichiometric  $\text{CH}_4/50\%$  air +  $50\%$   $\text{H}_2\text{O}_2$  flame, the maximum heat release rate is about  $5.42 \times 10^{10}$  J/m<sup>3</sup> s which is almost 9 times higher than that of stoichiometric  $\text{CH}_4/\text{air}$  flame. When the air is completely replaced by  $\text{H}_2\text{O}_2$ , the maximum heat release rate can even reach to  $4.49 \times 10^{11}$  J/m<sup>3</sup> s which is about two-order of magnitude higher than that for stoichiometric  $\text{CH}_4/\text{air}$  flame. It is apparent that the decomposition of  $\text{H}_2\text{O}_2$  dominates the heat release and then increases the adiabatic flame temperature.

In order to understand the effect of chemical reaction on the flame speed of  $\text{CH}_4/\text{air}/\text{H}_2\text{O}_2$  flames, the first-order sensitivity analysis of laminar burning velocity is shown in Fig. 4 for different reactant compositions at stoichiometric condition. In the case of pure air, the dominant reactions for laminar burning velocity are,



For the hydrogen peroxide replacement cases, the dominant reactions shift to the following reaction steps:

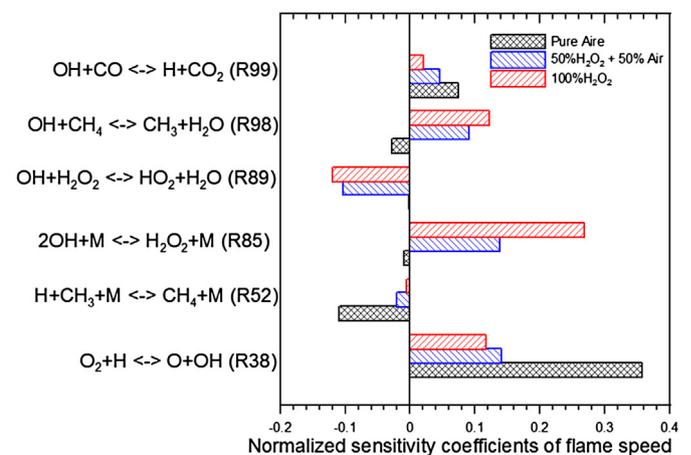


Fig. 4. Sensitivity analysis of laminar burning velocity for stoichiometric  $\text{CH}_4/\text{air}/\text{H}_2\text{O}_2$  flames.

Among these reactions, (R85) and (R89) are the most important chemical reactions. Hydrogen peroxide promotes the production of OH radicals, modifies the reaction pathway, and enhances the reaction rate leading to the increase of flame speed.

### 3.3. Production rate of intermediate species

The effects of  $H_2O_2$  on the major species and OH radical of the  $CH_4/air/H_2O_2$  premixed flames have been discussed in our previous paper [16]. In the present study, the dominant reactions for the intermediate species (O, H,  $HO_2$ , HCO,  $CH_3O$  and  $CH_2O$ ) in  $CH_4/air/H_2O_2$  premixed flames are further analyzed. These species are crucial to clearly reveal the reaction pathway in  $CH_4/air/H_2O_2$  flames.

Fig. 5 shows the production rate of H for  $CH_4/air$  and  $CH_4/50\%$  air + 50%  $H_2O_2$  flames at stoichiometric condition. With hydrogen peroxide addition, some of the main reactions are enhanced and shifted in axial position. Reactions (R46) and (R57) are evidently increased. The dominated reaction for H production is



Since OH is significantly increased by  $H_2O_2$ , it then assists in yielding H from reaction (R84). However, the dominant reaction for H consumption is



In Fig. 5 it is obvious that all the main reactions are enhanced by hydrogen peroxide addition. Fig. 6 shows the production rate of O for two different flames at stoichiometric condition. With hydrogen peroxide addition, the main reactions of O are not altered. Nonetheless, the rates of dominant reaction steps are enhanced and the total production rate of O is also increased. The dominant reactions for O production is



And the main O consumption reactions are

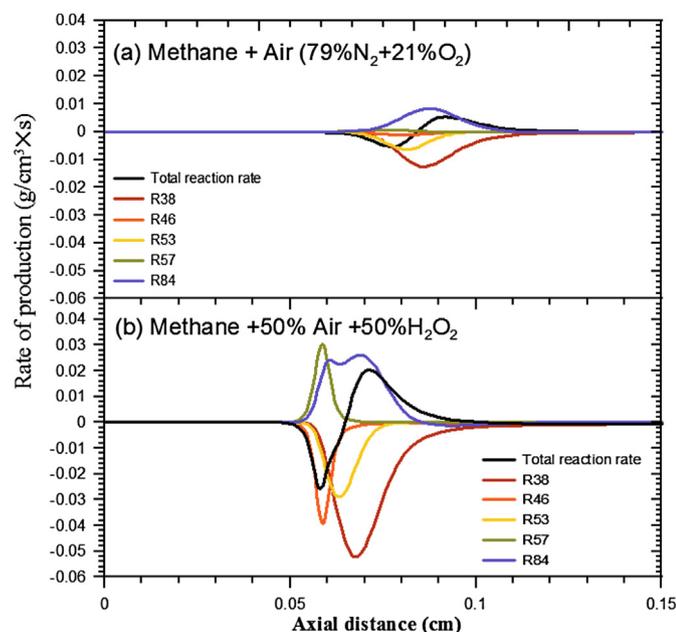


Fig. 5. Effect of  $H_2O_2$  on H production rate for  $CH_4/air$  and  $CH_4/50\%$  air + 50%  $H_2O_2$  flames at stoichiometric condition.

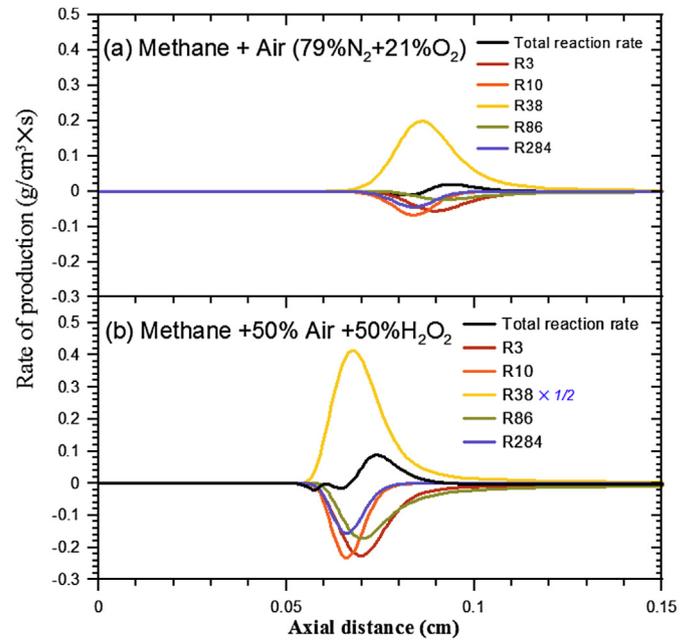


Fig. 6. Effect of  $H_2O_2$  on O production rate for  $CH_4/air$  and  $CH_4/50\%$  air + 50%  $H_2O_2$  flames at stoichiometric condition.



The chain branching reaction (R38) is one of the most important reactions in combustion process.

Fig. 7 shows the  $HO_2$  production rate for two different flames at stoichiometric condition. For the  $CH_4/air$  flames, the dominant reactions for the production and consumption of  $HO_2$  are (R168) and (R46), respectively.

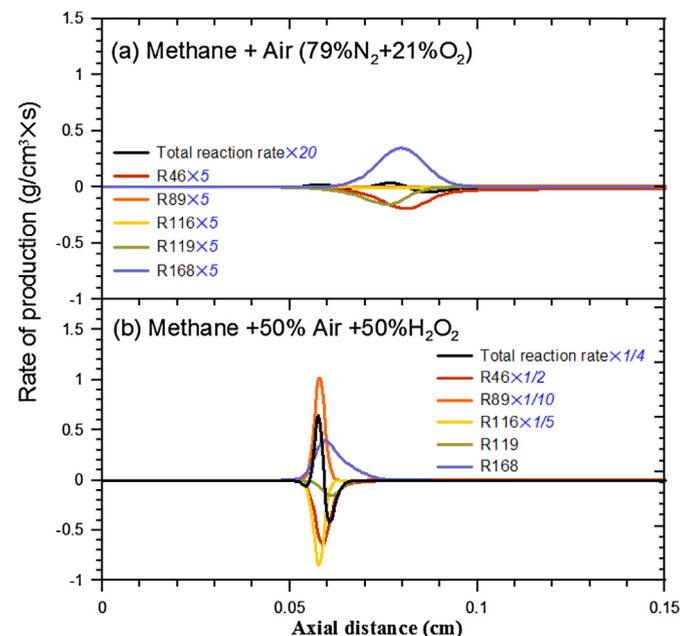


Fig. 7. Effect of  $H_2O_2$  on  $HO_2$  production rate for  $CH_4/air$  and  $CH_4/50\%$  air + 50%  $H_2O_2$  flames at stoichiometric condition.



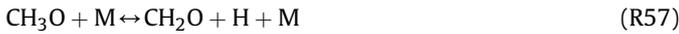
When 50% of air is replaced by hydrogen peroxide, all the major reactions and the total production rate are greatly increased. The dominant reaction for  $\text{HO}_2$  production is shifted from (R168) to (R89).



Hydrogen peroxide promotes the production of  $\text{HO}_2$ . In addition to reaction (R46), the other dominant reaction for  $\text{HO}_2$  consumption is reaction (R116).



Fig. 8 compares the  $\text{CH}_3\text{O}$  production rate for two different flames at stoichiometric condition. Similarly to the previous intermediate species, the existence of  $\text{H}_2\text{O}_2$  promotes the main reactions and the total reaction rates substantially. The dominant reactions for  $\text{CH}_3\text{O}$  production and consumption are (R119) and (R57), respectively,



Reaction (R57) is also the dominant reaction for  $\text{CH}_2\text{O}$  production when hydrogen peroxide is used to replace air (see Fig. 9).

Fig. 9 shows the  $\text{CH}_2\text{O}$  production rate for two different flames at stoichiometric condition. For the  $\text{CH}_4/\text{air}$  flame, the dominant reactions of  $\text{CH}_2\text{O}$  consumption and production are (R58) and (R10), respectively,



Reaction (R10) is the most important step for  $\text{CH}_3$  oxidation to

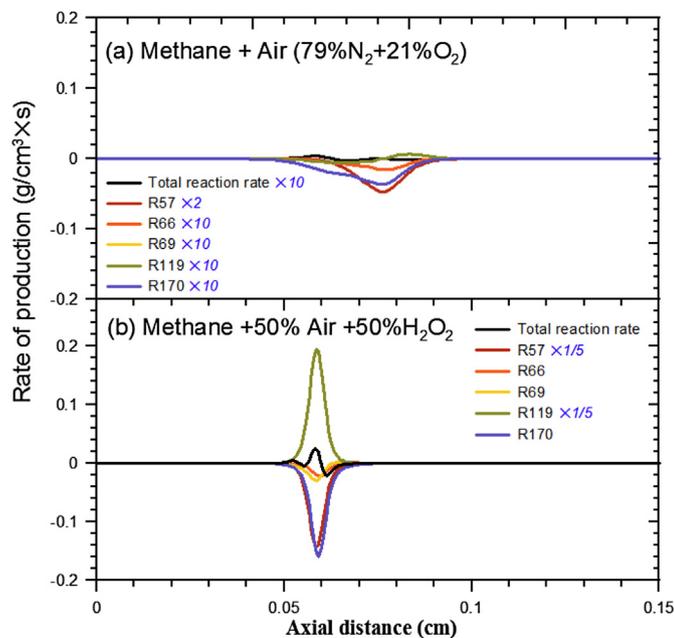


Fig. 8. Effect of  $\text{H}_2\text{O}_2$  on  $\text{CH}_3\text{O}$  production rate for  $\text{CH}_4/\text{air}$  and  $\text{CH}_4/50\% \text{ air} + 50\% \text{ H}_2\text{O}_2$  flames at stoichiometric condition.

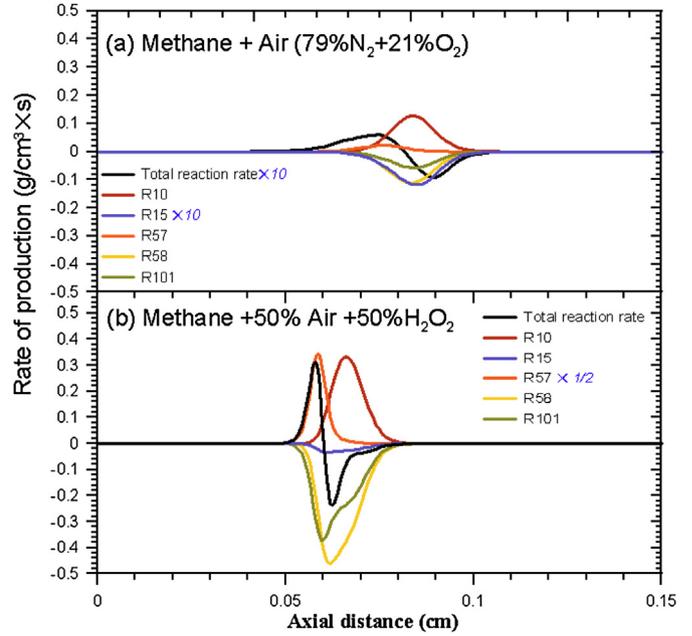
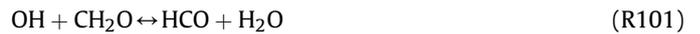


Fig. 9. Effect of  $\text{H}_2\text{O}_2$  on  $\text{CH}_2\text{O}$  production rate for  $\text{CH}_4/\text{air}$  and  $\text{CH}_4/50\% \text{ air} + 50\% \text{ H}_2\text{O}_2$  flames at stoichiometric condition.

form  $\text{CH}_2\text{O}$ , which belongs to progressive dehydrogenation of  $\text{CH}_4$  to  $\text{CH}_3$  and then  $\text{CH}_3\text{O}$ ,  $\text{CH}_2\text{O}$ ,  $\text{HCO}$ ,  $\text{CO}$  and finally to  $\text{CO}_2$ . For the  $\text{CH}_4/50\% \text{ air} + 50\% \text{ H}_2\text{O}_2$  flame, hydrogen peroxide enhances reactions (R10) and (R57), but the enhancement of reaction (R57) is more significant than that of reaction (R10). Therefore, the reaction pathway of  $\text{CH}_3$  to  $\text{CH}_2\text{O}$  is changed. The primary path is from  $\text{CH}_3$  to  $\text{CH}_3\text{O}$  and then to  $\text{CH}_2\text{O}$ . With 50% of  $\text{H}_2\text{O}_2$  replacement, the reactions (R10), (R15), (R57), (R58), and (R101) are all enhanced. In addition to reaction (R58), the reaction (R101) is enhanced to become the second important reaction for  $\text{CH}_2\text{O}$  consumption.

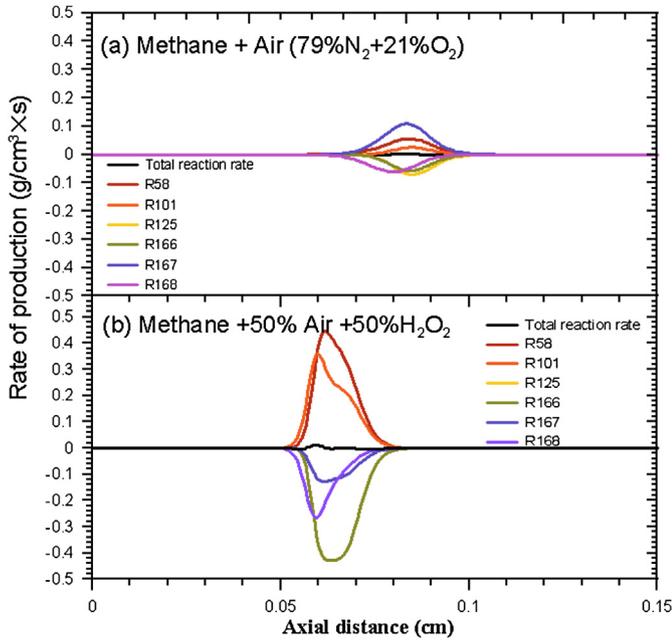


The  $\text{HCO}$  production rate is shown in Fig. 10. For the  $\text{CH}_4/\text{air}$  flames, the dominant reactions for  $\text{HCO}$  production and consumption are



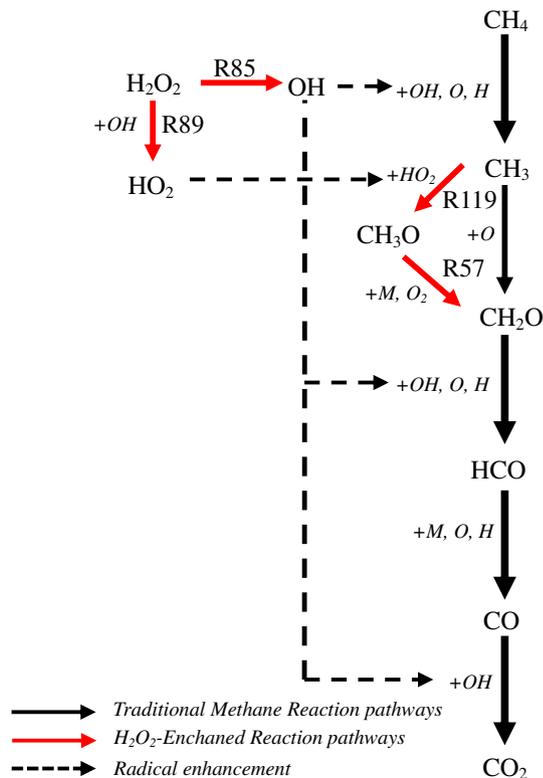
When air is partially replaced by hydrogen peroxide, all the main reactions and the total production rate are increased. Reactions (R166), (R167), and (R168) are also the dominant reactions for  $\text{CO}$  production. The produced  $\text{CO}$  is then reacted with  $\text{OH}$  to form  $\text{CO}_2$ .

Finally, the effect of hydrogen peroxide addition on reaction pathways is shown in Fig. 11. The black arrows indicate the traditional dominant  $\text{CH}_4/\text{air}$  combustion pathways. The red arrows represent the enhanced reaction steps by  $\text{H}_2\text{O}_2$  addition and the blue dashed lines represent the increased radicals to enhance the



**Fig. 10.** Effect of  $\text{H}_2\text{O}_2$  on  $\text{HCO}$  production rate for  $\text{CH}_4/\text{air}$  and  $\text{CH}_4/50\% \text{air} + 50\% \text{H}_2\text{O}_2$  flames at stoichiometric condition.

dominant reaction steps. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.) It has been shown that the addition of hydrogen peroxide increases the productions of  $\text{OH}$  and  $\text{HO}_2$  in  $\text{CH}_4/\text{air}$  flames [21]. The increased  $\text{OH}$  radicals promote progressive dehydrogenations of  $\text{CH}_4$  to  $\text{CH}_3$  and of  $\text{CH}_2\text{O}$  to  $\text{HCO}$ , and finally oxidation of  $\text{CO}$  to  $\text{CO}_2$ . In traditional  $\text{CH}_4/\text{air}$  combustion,  $\text{CH}_2\text{O}$  is primarily produced from



**Fig. 11.** Effect of  $\text{H}_2\text{O}_2$  addition on reaction pathways.

oxidation of  $\text{CH}_3$  through reaction (R10). However, with the addition of  $\text{H}_2\text{O}_2$ ,  $\text{CH}_2\text{O}$  is primarily produced from  $\text{CH}_3\text{O}$  through reaction (R57). In addition, the increased  $\text{HO}_2$  also accelerates  $\text{CH}_3\text{O}$  production through reaction (R119).

#### 4. Conclusions

In the present study, the effects of hydrogen peroxide addition on premixed methane/air flames are numerically investigated with GRI-Mech 3.0 chemical kinetic mechanisms and detailed transport properties. Hydrogen peroxide is used as an oxidizer to replace partial air. The characteristics of laminar burning velocity, adiabatic flame temperature, and production rate of intermediate species ( $\text{O}$ ,  $\text{H}$ ,  $\text{HO}_2$ ,  $\text{HCO}$ ,  $\text{CH}_2\text{O}$  and  $\text{CH}_3\text{O}$ ) are studied. The following findings are obtained from this study.

1. The laminar burning velocity and adiabatic temperature of premixed  $\text{CH}_4/\text{air}$  flames are obviously increased with the addition of  $\text{H}_2\text{O}_2$ . The dominant reactions for laminar burning velocity are shifted from (R38), (R52), and (R99) to (R38), (R85), (R89), and (R98). In addition, the decomposition of  $\text{H}_2\text{O}_2$  dominates the net heat release rate and then affects the adiabatic flame temperature.
2. Hydrogen peroxide increases not only the reaction rates, but also the concentrations of intermediate species. The increase of intermediate species then alters the reaction pathway of methane flames.

In  $\text{CH}_4/\text{air}$  flame,  $\text{CH}_2\text{O}$  is primarily produced by the oxidation of  $\text{CH}_3$  with  $\text{O}$ . With the addition of  $\text{H}_2\text{O}_2$ , the primary reaction pathway is shifted from the progressive reaction of  $\text{CH}_3$  to  $\text{CH}_3\text{O}$  and then to  $\text{CH}_2\text{O}$ . This difference is primarily due to the substantial increase of  $\text{HO}_2$ .

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