Effects of Hydrogen Peroxide on Methane Premixed Flames

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1 Introduction

Lean combustion is generally considered as a timely solution for the more stringent environmental regulations and global weather concerns in the new century. However, lean combustion is suffered to combustion instability, such as flame pulsation, flame blowout and flickering, due to low heat release rate and high local extinction. It turns out to release high CO and UHC (unburned hydrocarbon) emissions in lean combustion application. Some strategies for stabilizing lean premixed flames and extending lean flammability limit have been proposed in decades [1-3]. Altering chemical characteristics in flames is one of successful manners to sustain lean combustion. For example, utilizing catalyst in combustors [4] and adding strong oxidant to fuels [5] are common approaches. Employing active fuel or adding strong oxidant to fuels is subject to increase flame temperature and reactive radicals in flames, and then to further stabilize lean combustion. Nevertheless, hydrogen peroxide (H_2O_2) is an environmentally friendly oxidant with strong oxidability. Even after chemical dissociation, hydrogen peroxide produces only oxygen and steam.

A few recent studies have indicated some potential promises in utilizing hydrogen peroxide for improving practical combustion process. Golovitchev et al. [6] examined the possibility of promoting methane auto-ignition in air using 5-10% hydrogen peroxide. Ting and Reader [7] used PREMIX code to investigate the effects of hydrogen peroxide on the premixed methane-air flame under atmospheric conditions. Hydrogen peroxide was found to be effective in enhancing the burning velocity, and this was particularly true for the richer mixtures considered. Kim et al. [8] discovered that hydrogen peroxide assisted the conversion of harmful nitric oxide to nitrogen dioxide in diesel exhaust gas. In addition, Born and Peters [9] found that proper injection of hydrogen peroxide into a diesel engine reduced soot and NOx drastically. Martinez et al. [10] reported that the concentrations of UHC, CO and NOx from their industrial pilot plant scale combustion chamber fueled with natural gas were lowered significantly by the injection of a few hundred ppm of hydrogen peroxide. Based on the above studies, promotion to stable lean combustion is more effective with the addition of hydrogen peroxide

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than that with hydrogen addition. Hydrogen is considered as an easy-ignited fuel and helpful for increasing flame temperature. But, hydrogen peroxide is as a reactive stimulator or an oxidizer to shift chemical pathways and to further enhance chemical radicals.

This study numerically examines the effects of hydrogen peroxide on premixed methane-air flames using CHEMIKIN Collection with GRI-Mech 3.0 chemical reaction mechanisms [11]. Numerical results indicate the addition of hydrogen peroxide affects the reaction pathway, laminar burning velocity, flame temperature, and pollutant formation.

2 Numerical Method

In this work, the PREMIX code of Chemkin collection 3.7 is used to calculate the adiabatic, unstrained, free propagation velocities of the laminar premixed CH_4/air/H_2O_2 flames. It solves the steady-state comprehensive mass, species and energy conservation equations for the planar flame. In addition, the adiabatic flame temperature is calculated by using the EQUIL code. The GRI-Mech 3.0 chemical kinetic mechanisms and detailed transport properties are used without any modifications. The unburned reactants are supplied at 423 K and 1 atm. This temperature is the boilng temperature of H_2O_2 and it will make reactant to be in gas phase.

Simulations were performed for three different conditions: (1) replacing partial air by H_2O_2 (2) adding H_2O_2 as fuel additive (3) using H_2O_2 as an oxidizer but with different concentrations. The decompostion of one mole H_2O_2 will produce 0.5 mole O_2 and one mole H_2O. Therefore, the stochiometric CH_4/H_2O_2 ratio is 0.25 and the global reaction is defined as:

$$CH_4 + 4H_2O_2 \rightarrow CO_2 + 6H_2O \quad (1)$$

In the case of replacing partial air by H_2O_2, the total O_2 is maintained to keep the equivalence ratio constant but N_2 will be decreased. In the case of adding H_2O_2 as fuel additive, the maximum ratio of H_2O_2 to methane is 100%. In the last case of using H_2O_2 as an oxidizer, the concentration of H_2O_2 is varied from 30 to 100%.

3 Results and Discussion

The temperature and species concentration profiles of the premixed stoichiometric CH_4/air and CH_4/85% air /15% H_2O_2 are shown in Fig.1. Results indicate that the permixe flame with 15% air replaced by hydrogen peroxide has a higher adiabatic flame temperature due to heat-released from thermal decomposition of hydrogen peroxide. In addition, the increase of H_2O_2 content in air results in an increase of water production, but decreases the carbon dioxide formation. On the other hand, some intermediate radicals, such as CO, OH, H, O, and HO_2 etc, show the increasing trends. Thess facts suggest that the dominate reaction path of methane combustion is altered by H_2O_2 effect. Figure 2 shows the effect of replacing partial air by H_2O_2 on laminar burning velocity and adiabatic flame temperature for three different equivalence ratios (ER). It can be seen that the laminar burning velocity increases with increasing the replacement of air with H_2O_2. As air is completely repalced wih H_2O_2, the laminar burning velocity approaches to 4.7 m/s for ER = 0.6, 0.8, and 1.0. This is due to that the oxidizer is completely provided from hydrogen peroxide decomposition and hydrogen peroxide dominates the reaction rate of methane. The adiabatic flame temperature is also increased with increasing the replacement of air with H_2O_2. The maximum increased temperature is about 900 K for ER = 0.6 flame, which is higher than the increased temperature for ER = 0.8 and 1.0 flames. The increased temperature is due to that the heat released from hydrogen peroxide decomposition is much higher than that released from methane reactions.

Figure 3 indicates the effect of H_2O_2 concentrations on laminar burning velocity and adiabatic flame temperature of stiochoimetric CH_4/H_2O_2 flames. Results show that with 30 vol.% of H_2O_2 the laminar burning velociy is slightly higher than that of the stoichiometric CH_4/air flame, but the adiabatic flame temperature is lower by about 250 K. When the H_2O_2 concentration is increased to 40 vol.%, the adiabatic flame temperatue is about the same as that of the stoichiometric CH_4/air flame. Nevertheless, the laminar burning velocity is increased to 1.57 m/s, which is twice higher than that in stoichiometric condition (0.72 m/s). It means that the existence of hydrogen peroxide in premixed methane flame not only generates thermal effect, but also results in chemical effect. Figure 4 shows the effects of H_2O_2 additions to methane fuel on the laminar burning velocity and adiabatic flame temperature of the CH_4/H_2O_2/air flames with ER = 0.6, 0.8, and 1.0. It can be seen that the laminar burning velocity increases with increasing the H_2O_2 concentrations, especially for the case of ER = 1.0. However, the adiabatic flame temperature remains relatively constant for three different equivalence ratios studied. It is noted that the increase of H_2O_2 concentrations from 0 to 100% increases the laminar burning velocity by 42% for ER = 1.0 flame. When ER = 0.6, the laminar burning velocity is increased only by 26%. Comparisons of Figs. 4 and 2 suggest that the addition of small amount of H_2O_2 to fuel has lesser effect on the laminar burning velocity and adiabatic flame temperature than the replacement of air with H_2O_2. The effects of H_2O_2 on the OH, CO, NO and NO_2 formations in three different hydrogen peroxide adding approaches are investigated as shown in Figs. 5-8. The increase of OH concentration in flames manifests the severe chemical reactions, especially for the case of H_2O_2 as an oxidizer. OH concentration increases significantly when the H_2O_2 as an oxidizer is increased to 60% (see Fig. 5c). It elucidates that the hydrogen peroxide effect consists of thermal and chemical effects. The addition of extra H_2O_2 to fuel results in insignificant effect on OH formation (Fig. 5b).

Figure 6 shows that the CO concentration increases drastically in the preheat zone and then decreases in the reaction zone. The replacement of partial air with H_2O_2 increases CO formation (Fig. 6a). Whereas additing extra H_2O_2 to fuel decreases CO formation (Fig. 6b). CO concentration increases significantly when the H_2O_2 as an oxidizer is increased to 60% (Fig. 6c). As to NO_x emission, Figs. 7 and 8 show that both NO and NO_2 concentrations increase with either replacing partial air by H_2O_2 or adding extra H_2O_2 to fuel. The enhancement of NO_x production is due to the increased flame temperature.

4 Conclusions

The effect of hydrogen peroxide on methane/air premixed flames are numerically investigated under the atmosphere condition. Hydrogen peroxide is used as fuel additive or oxidizer for three different conditions: (1) replacing partial air by H_2O_2, (2) adding H_2O_2 as fuel additive, (3) using H_2O_2 as an oxidizer but with different concentrations. Results show that the laminar burning velocities are obviously increased no matter H_2O_2 is used as fuel additive or oxidizer. The adibatic temperatures are also increased with the addition of H_2O_2. However, when H_2O_2 is used as fuel additive, the adiabatic flame temperature is not increased obviously, especial in the fuel-lean conditions.

The effects of H_2O_2 on the OH, CO, NO and NO_2 are also investigated. OH concentration is always increased unless lower H_2O_2 concentration is used as an oxdizer (30% in the study). The increase of OH concentration enhances the reaction rate and then affects the laminar burning velocity. For CO emission, the CO emission is decreased with adding H_2O_2 as fuel additive. However, using H_2O_2 to replace air will increase CO emission. It is due to the coversion of CO_2 to CO in the high temperature condition. If H_2O_2 is used as an oxidizer with lower concentrations (30% and 40%), CO emission is decreased. As to NO_x emission, using H_2O_2 as an oxidizer will never produce NO_x. For the other two cases, the NO_x emission is always increased due to the increased flame temperature.

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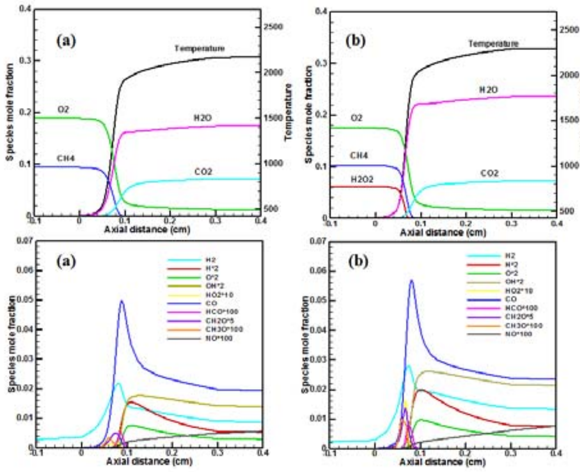

Fig. 1 Profiles of temperature and species concentration of stoichiometric CH_4/air/H_2O_2 flames. (a) CH_4 and 100% air, (b)CH_4, 85% air and 15% H_2O_2.

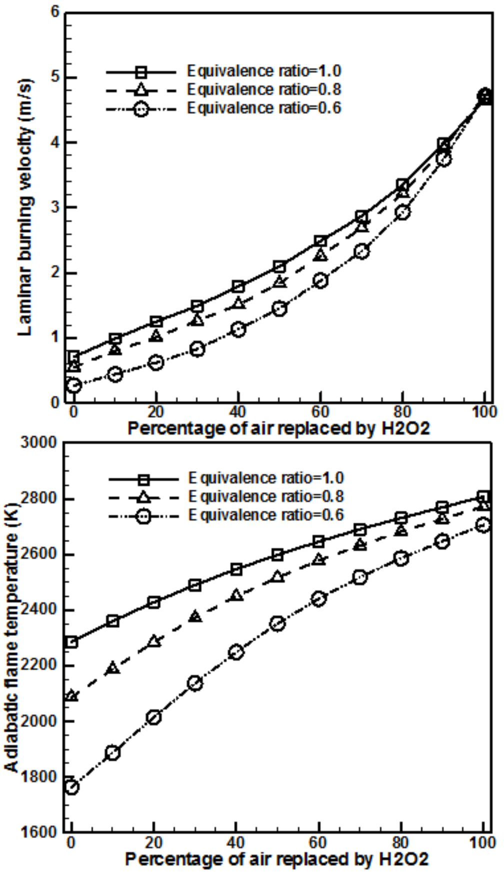

Fig. 2 The laminar flame speed and adiabatic flame temperature of methane/air flames with different percentages of air replaced by H_2O_2.

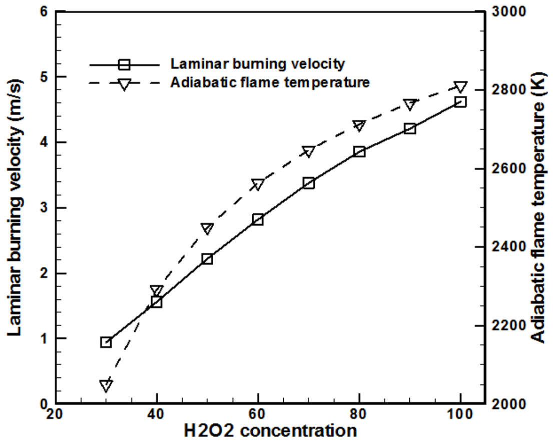

Fig. 3 The laminar flame speed and adiabatic flame temperature of stoichiometric methane/ H_2O_2 flames with different H_2O_2 concentration.

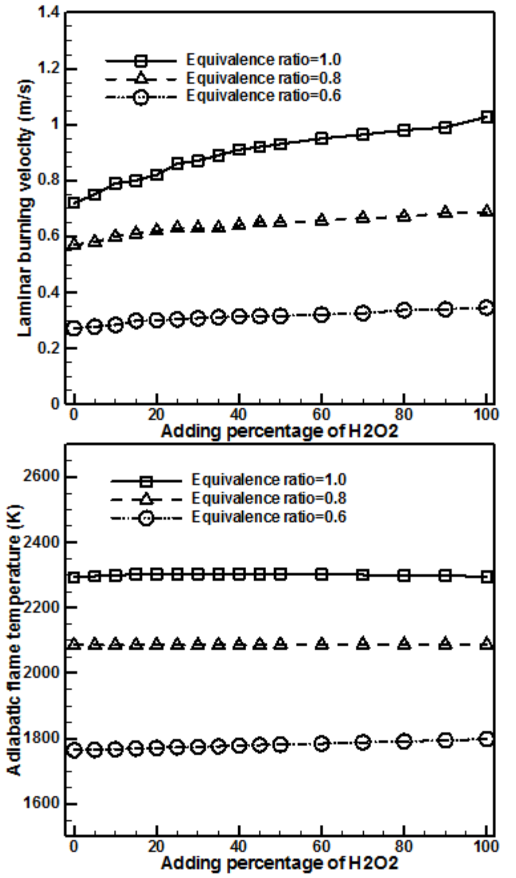

Fig. 4 The laminar flame speed and adiabatic flame temperature of methane/air/H_2O_2 flames with different percentages of H_2O_2 additions.

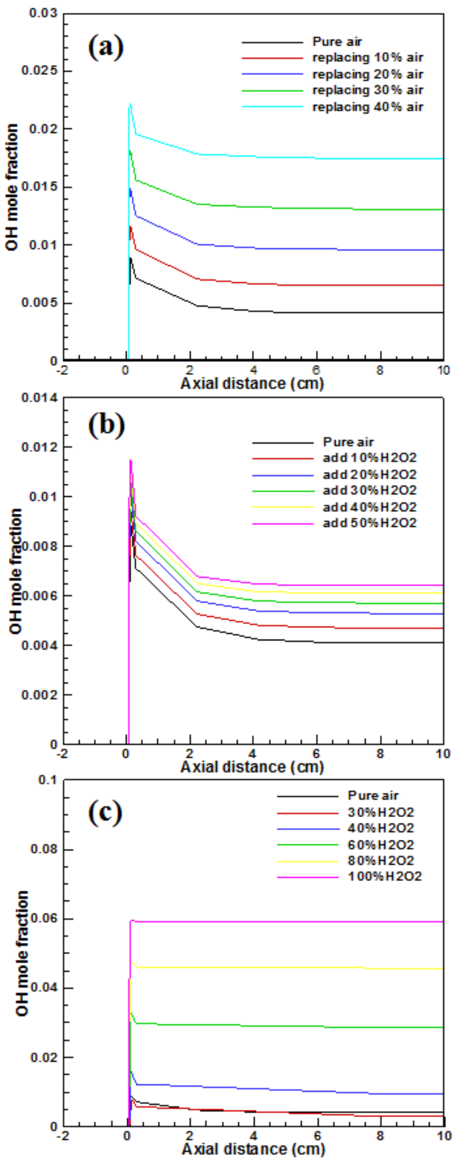

Fig. 5. The effect of H_2O_2 on OH mole fraction for three cases: (a) replacing partial air by H_2O_2 (b) adding extra H_2O_2 (c) H_2O_2 as an oxidizer with different concentrations.

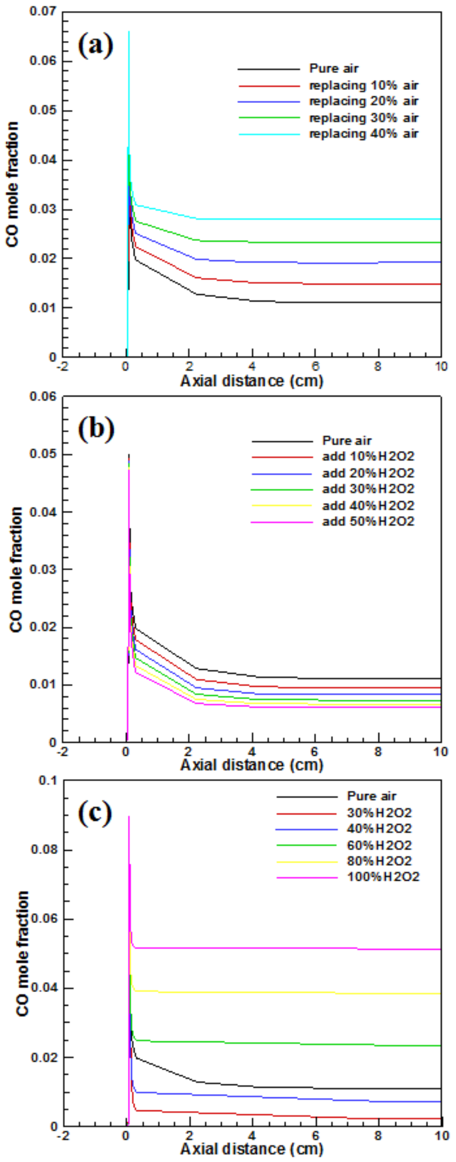

Fig. 6 The effect of H_2O_2 on CO mole fraction for three cases: (a) replacing partial air by H_2O_2 (b) adding extra H_2O_2 (c) H_2O_2 as oxidizer with different concentrations.

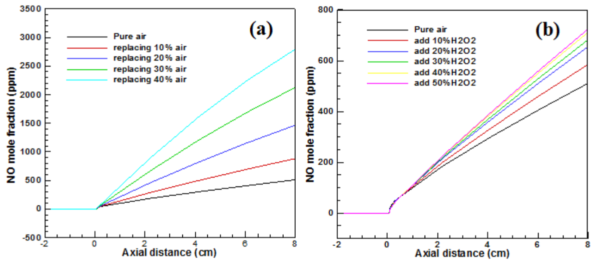

Fig. 7 The effect of H_2O_2 on NO mole fraction for two cases: (a) replacing partial air by H_2O_2 (b) adding extra H_2O_2.

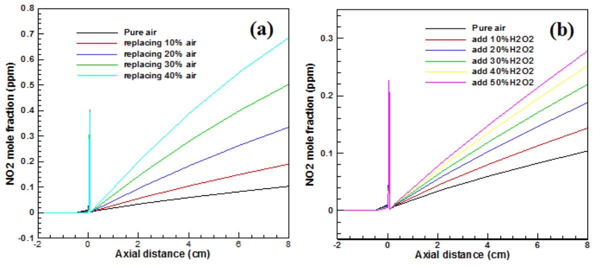

Fig. 8. The effect of H_2O_2 on NO_2 mole fraction for two cases: (a) replacing partial air by H_2O_2 (b) adding extra H_2O_2.