

添加氢气对 LPG/空气预混火焰结构的影响

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摘要: 针对氢气添加的 LPG(液化石油气)+空气预混火焰结构进行了数值研究, 详细计算了在含氢比 a 为 0% 到 45%、稀释因子 D 为 21% 到 16% 条件下的自由蔓延火焰, 得到了不同燃烧条件($\phi=0.7-1.4$)下的绝热燃烧速率变化规律. 由于 LPG 中的主要成分为丙烷和丁烷, 作者针对 C3 和 C4 物质提出了详细化学反应动力学系统, 并针对氢气添加的丁烷燃烧过程进行了数值计算, 得到了与实验相一致的结果, 验证了改进的详细化学机理的有效性. 此外, 进一步计算了对撞双火焰的加氢 LPG 火焰, 更加深入地探讨了火焰拉伸对燃烧稳定性和温度的影响, 重点研究了 ϕ 在 0.5 到 0.7 的稀薄燃烧, 验证了氢气添加可以有效提高稀薄燃烧条件下熄火拉伸率, 扩大稀薄燃烧的极限, 增加火焰的稳定性.

关键词: 液化石油气; 氢气添加; 绝热燃烧速度; 熄火拉伸率

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Effects of Hydrogen Enhancement in LPG/Air Premixed Flame

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Abstract: A numerical study of hydrogen-enhanced liquefied petroleum gas (LPG)+air flames was presented. The variations of the adiabatic burning velocity in different conditions of combustion ($\phi=0.7-1.4$) were studied extensively. The hydrogen content in the fuel was varied from 0% to 45% and the dilution factor was from 21% to 16%. Since the major components of LPG are butane and propane, an appropriate chemical kinetic model must be chosen to solve the chemical reaction of C3 and C4 species. Validation of the chemical kinetic model against the fundamental combustion data was performed to insure accuracy. In addition, independent simulations were conducted in the opposed-jet, symmetric, twin-flame configuration. The effects of fluid mechanics, as manifested by the induced strain rate, were also considered. The effects of extinction strain rate on flame temperature and the flammability limits were calculated and the results showed that hydrogen-enhanced LPG/air premixed flames were more stable at high flame strain. The lean flammability limits were extended by the H₂ addition.

Key Words: Liquefied petroleum gas; Hydrogen-enhanced; Adiabatic burning velocity; Extinction strain rate

LPG fuel consists mainly of propane, butane, propylene, butylene in various proportions according to its state or origin. The composition of LPG fuel varies very widely from one country to another. As one of clean fuel, LPG fuel has attracted increased interest in the recent years. Lee *et al.*^[1] investigated experimentally the flame propagation and combustion characteristics of LPG fuel by using a CVCC (constant volume combustion cham-

ber) and a port injection type heavy duty LPLi (liquefied petroleum liquid injection) engine system; the effects of ϕ on both the flame propagation and the combustion characteristics are proved greater than that of the ambient conditions; ϕ is defined as the ratio of the fuel-to-oxidizer ratio to the stoichiometric fuel-to-oxidizer ratio. The experimental study of laminar burning velocity and explosion index of LPG/air and propane/air

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mixtures was performed by Huzayyin *et al.*^[2]; the maximum laminar burning velocity found for propane is nearly $45.5 \text{ cm} \cdot \text{s}^{-1}$ at $\phi=1.1$, while that for LPG fuel is nearly $43.2 \text{ cm} \cdot \text{s}^{-1}$ at 4.5% fuel ($\phi \approx 1.5$). Experimental determination of the flammability limits of LPG-air mixture was concerned by Mishra *et al.*^[3], the lower flammability limits were found to be 1.81% and the upper flammability limits were 8.86% of LPG for upward propagation of flame.

In addition, gas addition to hydrocarbon has been studied more and more recently. Coppens *et al.*^[4,5] studied non-stretched methane/hydrogen/air flame; the adiabatic burning velocity and NO formation were measured experimentally, and it was found that in lean flames, enrichment by hydrogen has little effect on NO, while in rich flames, the concentration of NO decreases significantly. Some numerical studies were performed in hydrogen-enhanced methane-air premixed flames. The same results as the experimental study were presented using the numerical study by Gauducheau *et al.*^[6]. However, there are some limitations in this field as follows. (1) The studies of gas addition to hydrocarbon are almost focused on methane and some low carbon species. The study of C3 and C4 species is difficult to be found in literature. (2) There are limited data in the literature about the numerical study of LPG flame, and also lack of experimental data on the LPG study. (3) The data of LPG flame in the literature is not good enough to be provided as reference, because the compositions of LPG fuel are remarkably different from each other. (4) In hydrogen-enhanced hydrocarbon/air premixed flames, there is lack of data of the temperature as a function of the strain rate. (5) There is no study about hydrogen-enhanced LPG/air premixed flames so far.

Thus, for the purpose of prediction of hydrogen-enhanced premixed LPG/air flame structure, the contents of the present study are as follows. (1) The detailed chemical reaction mechanism was obtained to emphasize the important roles of the C3 and C4 species, and the comparison between the experimental results and the numerical simulation of the detailed chemical reaction mechanism is required. The results of the comparison proved that the detailed mechanism was good for the study of LPG flames. (2) Adiabatic burning velocities with different mole fraction of hydrogen in the LPG fuel and with different dilution factor were numerically determined. (3) Additionally, another judgment, extinction strain rate was simulated to investigate the effect of hydrogen addition to LPG fuel flames.

1 Numerical modeling details

1.1 Chemical kinetic model

The original kinetic model employed in this study is taken from literature^[7]; the oxidation kinetics of C1 and C2 species are based on the GRI-Mech 1.2^[8]. The original kinetic model consists of 619 reactions and 92 chemical species and is compiled to predict the combustion characteristics of methane, ethylene, acetylene, propane, butane, and isobutene, and the validation tests were conducted in literatures^[7,9]. Sung *et al.*^[10] computationally

explored the feasibility of extending the dilution limit of an engine during cold-start conditions. By adding H_2 or CO to the inlet manifold of the engine, they modified the kinetic mechanism consisting of 69 species and 515 reactions, which reduced the central processing unit time by 40%, because C5, C6, and the higher order hydrocarbons were not expected to be important in the flame speed calculations of butane flames. Thus, in the LPG/air flames, we used the same kinetic mechanism very similarly. It also enhanced some intermediate reactions to protect the hierarchical structure of combustion mechanism of higher hydrocarbons. Finally, in the present study, we used Sung's modified chemical kinetic mechanism, enhanced N kinetics from GRI-Mech, which consists of 129 species and 877 reactions.

First, we numerically determined the adiabatic flame temperature of premixed propane/air flame and *n*-butane/air flame at $\phi=1$ at NPT (normal pressure and normal temperature conditions); the numerical results were 2266 and 2270 K, while the experimental values are 2273 and 2276 K^[11]. More importantly, we obtained the adiabatic burning velocities of premixed *n*-butane/air flames, which are shown in Fig.1. The comparison reveals the success of our modified chemical kinetic model, and our numerical values are higher than the experimental values^[11], the same as Sung's^[10]. It has been mentioned that the convergence is more difficult in fuel-rich condition. Therefore, in our study, we used a good method to help convergence in LPG/air flames.

1.2 Computational specification

There were two different flame modes employed in the present study. One was freely propagation flame to obtain the adiabatic burning velocities, and the other was opposed-jet, symmetric, twin-flame to obtain the extinction strain rate. First, we chose the premixed laminar flame-speed calculation model in the CHEMKIN program^[12] from Sandia National Laboratories to study the one-dimensional flame. This model needs the measured temperature profile as an input. As already known, a good temperature profile can give a good convergence. Coppens *et al.*^[4,5] used $100 \text{ K} \cdot \text{cm}^{-1}$ as the temperature gradient on the calculation of flame structure, while Dyakov *et al.*^[13] used $50 \text{ K} \cdot \text{cm}^{-1}$. In our

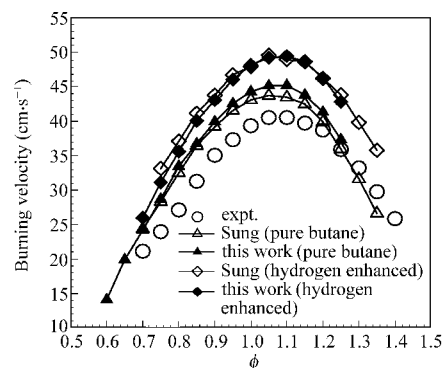


Fig.1 Comparison of experimental data^[11] and computed adiabatic flame speeds by Sung *et al.*^[10] and this work for premixed *n*-butane /air flame

study, $100 \text{ K} \cdot \text{cm}^{-1}$ was used as the temperature gradient on the calculation at $\phi=0.7-1.15$, and then used the temperature solution of the last calculation to model the next temperature profile; we found that convergence was easier than fixed temperature profile initial guess. The calculations were performed with an unburned mixture, at 300 K and 101.325 kPa. The adaptive mesh parameters were GRAD=0.1 and GURV=0.5. The relative and absolute error criteria were RTOL= 1×10^{-4} and ATOL= 1×10^{-9} , respectively; the total number of grid points was typically 110–140.

2 Results and discussion

2.1 Flame structure of freely propagation premixed LPG/air flame

In the present study, LPG used as fuel consists of 50% propane and 50% *n*-butane typically. The profiles of reactant mole fractions and temperature for premixed LPG/air flame are presented in Fig.2. Some parameters are as follows: fuel and oxidizer are mixed before combustion at $\phi=1$; there is no hydrogen addition to fuel ($a=0$); dilution factor $D=0.21$. The parameters are defined as follows^[4]:

$$a = \frac{X_{\text{H}_2}}{X_{\text{H}_2} + X_{\text{LPG}}} = \frac{X_{\text{H}_2}}{X_{\text{H}_2} + X_{\text{C}_3\text{H}_8} + X_{\text{C}_4\text{H}_{10}}}$$

$$D = \frac{X_{\text{O}_2}}{X_{\text{O}_2} + X_{\text{N}_2}}$$

$$\phi = \left(\frac{\sum_i X_{F_i}}{X_{\text{O}}} \right) \left(\frac{\sum_i X_{F_i}}{X_{\text{O}}} \right)_{st}$$

where X_k is the mole fraction of species k .

As seen in Fig.2, the temperature increases to the maximum of 2050 K rapidly at 0.05–0.16 cm; the reactants O_2 , C_3H_8 and C_4H_{10} are almost consumed completely, and some N_2 is also consumed in the reaction zone. The profiles of the mole fractions of some of the intermediate products, O, H, OH, H_2 , and HO_2 , are presented in Fig.3. The concentrations of H_2 and HO_2 increase at first and then decrease in the reaction zone, H, OH, and O are obtained in the high temperature zone. Fig.4 shows the concentration profiles of CO, CO_2 , and H_2O ; the formation of major products CO_2 and H_2O occurs in the reaction zone rapidly, and

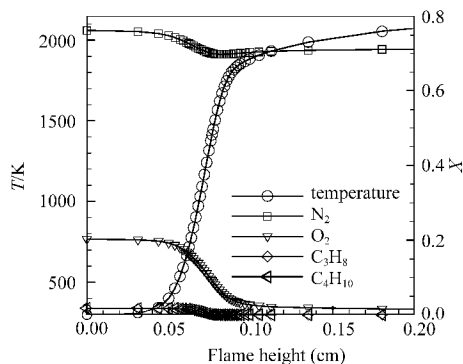


Fig.2 Profiles of reactant mole fraction and temperature for freely propagation premixed LPG/air flame

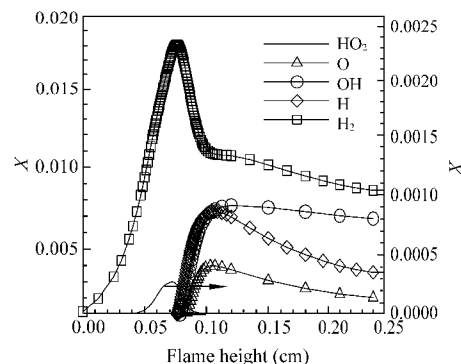


Fig.3 Profiles of mole fractions of O, H, H_2 , OH, HO_2 for freely propagation premixed LPG/air flame

the formation of intermediate products and some free radicals are earlier than the major products. The NO_x mole fraction profiles are shown in Fig.5. We can see that the formations of NO and NO_2 are very different. The formation of NO occurs mainly in the high temperature zone in which the reactions are very violent, but the formation of NO_2 is mainly in the low temperature zone. The NO_2 is then consumed in the high temperature zone, which is controlled by the formation mechanism of NO and NO_2 , and the formation of NO_x is more sensitive to temperature and some important free radicals.

2.2 Adiabatic burning velocity

The speed of a laminar unstretched freely propagating one-dimensional flame is the reference speed for all combustion studies. Some different combustion velocities are defined during flame propagation^[14]. In the present study, the adiabatic burning velocities of premixed LPG/air flame with various hydrogen fractions in fuel ($a=0-0.45$) and various dilution factors ($D=0.21-0.16$) were numerically determined. Fig.6(a, b) shows the results at $a=0.05$ and $a=0.45$. (1) Adiabatic burning velocities increase with the increase of dilution factor at the same hydrogen fraction in fuel. (2) Adiabatic burning velocities also increase with the increase of hydrogen fraction in fuel at the same dilution factor. The max value at $a=0.05$ is about $45 \text{ cm} \cdot \text{s}^{-1}$, while the value is about $50 \text{ cm} \cdot \text{s}^{-1}$ at $a=0.45$. (3) The maximum value of adiabatic burning velocity is at $\phi > 1$. As seen in Fig.6(a, b), the maximum adiabatic burning velocity is close to $\phi=1.1$; similar results for

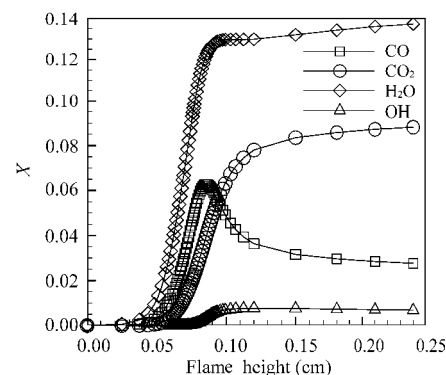


Fig.4 Profiles of mole fractions of CO, CO_2 , H_2O , and OH for freely propagation premixed LPG/air flame

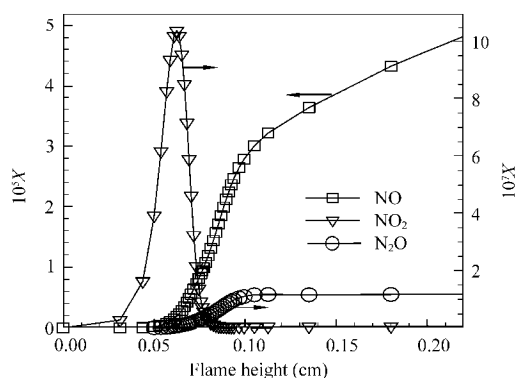


Fig. 5 Profiles of mole fractions of NO, NO₂, and N₂O for freely propagation premixed LPG/air flame

the premixed methane + hydrogen + air flame were presented by Coppens *et al.*^[4], but Hermanns *et al.*^[15] showed different results that the maximum adiabatic burning velocities shifted from $\phi=1.05$ –1.1 for methane/air flame when the hydrogen concentration was increased. Law *et al.*^[16] explained that the maximum laminar burning velocity of hydrogen flame is around $\phi=1.8$, and for methane or propane flame, it is around $\phi=1.1$.

The effects of H₂ addition were determined because the reaction zone moved near the inlet with the increase of the fraction of H₂ in fuel. Early oxidation of H₂ causes an increase in O, H, and OH radicals. It can be seen in Fig. 7 that primary radicals O, H, and OH, which are active species and react within the radical

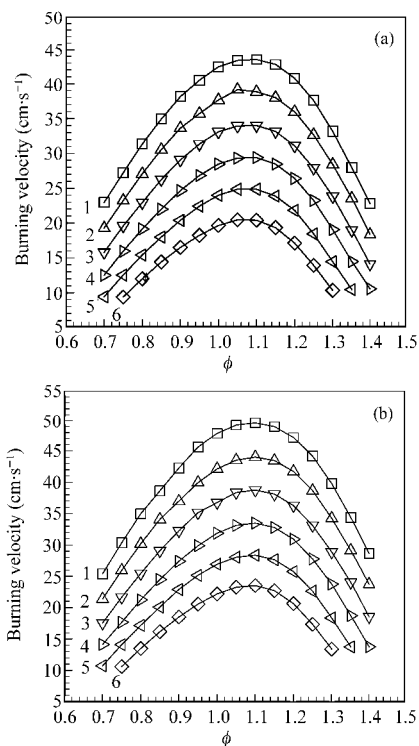


Fig. 6 Adiabatic flame speeds of hydrogen-enhanced premixed LPG/air flame with various dilution factors (D) and hydrogen fractions (a) as a function of ϕ

(a) $a=0.05$, (b) $a=0.45$; (1) $D=0.21$, (2) $D=0.20$, (3) $D=0.19$, (4) $D=0.18$, (5) $D=0.17$, (6) $D=0.16$

pool, have great effect on combustion. The reactions $H+O_2=O+OH$, $O+H_2=H+OH$, and $OH+OH=O+H_2O$, are chain branching in the controlling reactions of higher order hydrocarbons combustion, provide the essential radical pool of H, O, and OH, and are of great importance in the oxidation of hydrogen and hydrocarbons^[10]. The higher temperature and larger size of the radical pool with H₂ addition in turn lead to larger adiabatic burning velocity. Additionally, some reaction rates of the intermediate reactions were varied because of H₂ addition^[17], which can be a great factor for NO_x emission.

We can also see in Fig. 6(a, b) that the maximum adiabatic burning velocity increased from 43.11 to 49.65 cm·s⁻¹ at $D=0.21$ when the hydrogen fraction in fuel increased from 0.05 to 0.45 ($a=0.05$ to $a=0.45$). Lean burning at small ϕ is more attractive; the adiabatic burning velocity increased from 22.57 to 25.4 cm·s⁻¹ at $D=0.21$ when a increased from 0.05 to 0.45, by amounts of the order of 12.54%. The results from $a=0.05$ to $a=0.45$ (data available freely from www.whxb.pku.edu.cn) also show that the percent increase in the adiabatic burning velocities at various ϕ is linear with the amount of hydrogen addition. As expected, the hydrogen addition results in the extension of the lean flammability limit towards lower value and enhancement of the combustion intensity.

2.3 Effect of flame strain rate on H₂ enhanced lean premixed LPG/air flame

Lean burning affects the combustion stability because of weak burning, and the processes of flame stretching and heat loss can locally cause extinction. Therefore, the effect of flame strain rate on lean burning is very important. The effects of flame stretching were studied experimentally and numerically by Egolfopoulos *et al.*^[18]; the experimental determination of the flame structures and the extinction strain rates used both the opposed-jet and the single-jet wall configuration. Ren *et al.*^[19] simulated opposed-jet, symmetric, twin-flame configuration, and studied the strain rate effects on H₂ enhanced lean premixed CH₄/air combustion. Ref.[19] focused on $\phi=0.45$ –0.6, at very lean burn combustion condition. In the present study, opposed-jet, symmetric,

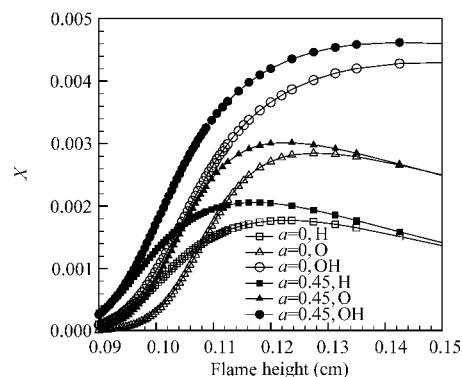


Fig. 7 H, O, OH mole fractions for premixed LPG/air flame at $a=0$ or $a=0.45$, $\phi=0.7$, and dilution factor $D=0.21$

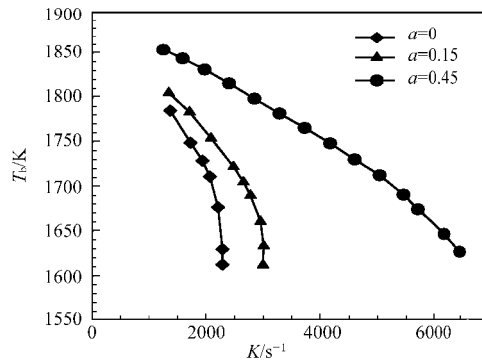


Fig.8 Flame temperature variation with strain rate at $\phi=0.7$ with different levels of hydrogen addition

twin-flame configuration is also used to determine the effects of strain rate on H_2 enhanced lean premixed LPG/air flame. The reactant stream consists of a premixed LPG+ H_2 +air mixture at various hydrogen fractions in fuel. The temperature at the inlet is set to 300 K and the pressure is 1.01×10^5 Pa for all the test cases. Fig.8 shows the variation of the maximum flame temperature at the stagnation plane (T_b) as a function of the imposed strain rate for an LPG/air flame ($a=0$) and with two levels of hydrogen addition ($a=0.15, 0.45$). Herein, the strain rate K is defined as the radial velocity gradient at the location of maximum heat release.

$$K = \left[\frac{1}{r} \frac{\partial}{\partial r} (vr) \right]_{\max \text{ heat release}}$$

In all the cases, the reaction layer is attached to the stagnation plane with the increase of the strain rate, and T_b decreases monotonically, until the extinction point is reached. We do not calculate the situation of $a=0.25$ and $a=0.35$, but we can predict that the same tendency will be obtained, and the curve lines will be between $a=0.15$ and $a=0.45$. The extinction strain rates at $a=0, 0.15$, and 0.45 are $K_{\text{ext}}=2284, 2993$, and 6485 s^{-1} , respectively. Especially, at $a=0.45$, the flame is more stable near the extinction point.

H_2 addition reduces the sensitivity of the flame, particularly in the early flame zone to this reaction. The early part of the flame zone is controlled predominantly by hydrogen oxidation reactions as mentioned in section 2.2. This can be easily seen in Fig. 7. Thus, H_2 oxidation for the LPG/ H_2 mixtures provides the

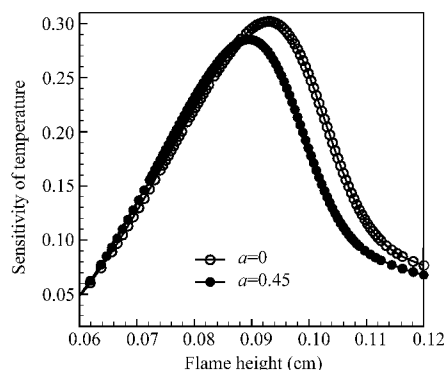


Fig.9 Local temperature sensitivity coefficients for reaction rate of $\text{CO}+\text{OH}=\text{CO}_2+\text{H}$ for premixed LPG/air flame at $a=0$ or $a=0.45, D=0.21$, and $\phi=0.7$

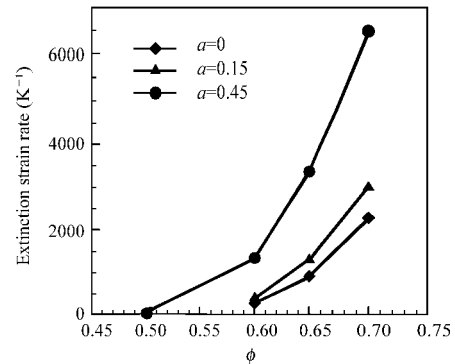


Fig.10 Extinction strain rate as a function of ϕ with various hydrogen fractions in fuel at dilution factor $D=0.21$

exothermic reactions and the radical pool in the early part of the flame and thereby helps to sustain the reactions of the C-containing species under conditions at which it may otherwise not be possible to sustain stable LPG flames. In Fig.9, we can see the local temperature sensitivity coefficients for the reaction rate of important critical reaction $\text{CO}+\text{OH}=\text{CO}_2+\text{H}$ at $a=0$ and $a=0.45$ in lean combustion. The sensitivity decreased clearly with increase of H_2 fraction in Fig.9. It is also presented in the CH_4/air premixed flame in literature^[20] from the temperature sensitivity coefficients analysis of the reactions: $\text{CO}+\text{OH}=\text{CO}_2+\text{H}$, $\text{HCO}+\text{O}_2=\text{CO}_2+\text{OH}$, and $\text{H}+\text{O}_2=\text{OH}+\text{O}$. Thus, opposed flame can live far from the stagnation plane with addition of H_2 , and extinction occurs more lately.

Fig.10 shows the extinction strain rate as a function of ϕ ($\phi=0.5-0.7$) at different levels of hydrogen addition. The results demonstrate the efficient extinction strain rate increases very clearly with the increase of hydrogen addition; and very importantly, the extinction flame temperature decreases with the decrease of ϕ at lean burning, especially at high hydrogen addition. At $a=0.45$, the extinction flame temperatures at $\phi=0.7, 0.65, 0.6$, and 0.5 are 1609, 1584, 1516, and 1449 K, respectively. The effect on extending flammability limit is also present in these figures.

Hydrogen addition increased immunity to stretch driven extinction efficiently. This can result in a significant improvement in the operation of lean burning. Unsteady response to composition fluctuations was also studied by Sankaran *et al.*^[21], but there was no discussion on it.

3 Conclusions

As one of clean fuel, LPG/air flame structure prediction is presented. A good mechanism for LPG flame is applied. The effects of hydrogen addition on the adiabatic burning velocity are studied. Furthermore, the effects of flame strain rate are also included. The results demonstrated an increased adiabatic burning velocity owing to the addition of hydrogen. Additionally, the effects of hydrogen addition increased the extinction strain rate, and enhanced the combustion intensity at the lean burning. Furthermore, at the very lean mixture condition, the lower flame extinction temperature sustained with the addition of hydrogen,

and certainly, the lower flame temperature was close to the flammability limit at the same flame stretched.

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