Study on laminar flame speed and flame structure of syngas with varied compositions using OH-PLIF and spectrograph

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Abstract

Various Bunsen flame information of premixed syngas/air mixtures was systematically collected. A CCD camera was used to capture the flame images. The OH-PLIF technique was applied to obtain the flame OH distribution and overall flame radiation spectra were measured with a spectrograph. Experiments were conducted on a temperature uncontrolled burner and syngas over a wide range of H2/CO ratios (from 0.25 to 4) and equivalence ratios (from 0.5 to 1.2). Results show that increasing hydrogen fraction (X\textsubscript{H2}) extends the blow-off limit significantly. The measured laminar flame speed using cone-angle method based on CCD flame imaging and OH-PLIF images increases remarkably with the increase of X\textsubscript{H2}, and these measurements agree well with kinetic modeling predictions through Li’s mechanism when the temperature for computation is corrected. Kinetic study shows that as X\textsubscript{H2} increases, the production of H and OH radicals is accelerated. Additionally, the main H radical production reaction (or OH radical consumption reactions) changes from R29 (CO + OH = CO\textsubscript{2} + H) to R3 (H\textsubscript{2} + OH = H\textsubscript{2}O + H) as X\textsubscript{H2} increases. Sensitivity analysis was conducted to access the dominant reactions when X\textsubscript{H2} increases. The difference on flame color for different X\textsubscript{H2} mixtures is due to their difference in radiation spectrum of the intermediate radicals produced in combustion.

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1. Introduction

With increasing concerns over the energy saving and environmental protection, “syngas” has recently attracted significant interest from the combustion community. Syngas, which is mainly composed of hydrogen and carbon monoxide, along with variable amounts of CH\textsubscript{4} and diluents like N\textsubscript{2} and CO\textsubscript{2}, can be produced through a variety of industrial processes, such as steam reforming of natural gases, gasification of coal and biomass, or even organic waste [1]. Syngas has been seen as an environmentally clean fuel and is potentially facilitating clean gas house reduction. One prominent application of syngas is in the Integrated Gasification Combined Cycle (IGCC) power plant. With the IGCC technique, minerals, mercury and sulfur compounds in coal are captured and syngas is extracted and subsequently used as a primary fuel for combustion in gas turbine [2]. However, because of the difference in coal quality and gasification methodology, composition of syngas can be vastly different, which complicates the gas turbine design and operation. Additionally, there is a growing interest in the utilization of High Hydrogen Content (HHC) fuels [3] in order to reduce greenhouse gas emissions, but high-temperature associated with hydrogen combustion leads to the increased thermal...
load and higher NOX emissions. Furthermore, lean premixed combustion (LPC) has been shown to offer considerably higher energy efficiency and lower pollutant emissions, but gas turbines operating under LPC are more susceptible to combustion instability and flashback [4]. Thus, understanding the combustion characteristics of premixed syngas/air mixtures over wide range of mixture compositions is essential for the IGCC based power plant.

One aspect of previous studies on syngas combustion is the measurement of laminar flame speed by using different methods. Mclean et al. [5] measured the laminar flame speed of stoichiometric H2/CO/air mixtures for H2/CO ratio of 5%/95% and 50%/50% at atmospheric pressure using the spherically propagating flame method. Subsequently, Brown et al. [6] extended the above study to a full equivalence ratio range, but they focused more on the effect of equivalence ratio and hydrogen fraction on the Markstein length. Vagelopoulos and Egolfopoulos studied the effect of hydrogen addition on premixed counterflow CO/air flames. Laminar flame speeds and extinction strain rate were determined by Laser Doppler Velocimetry (LDV) technique [7], and the result showed that small amount of hydrogen addition would increase the laminar flame speed and extend the extinction limit. Other studies also reported the combustion characteristics of syngas mixture, like laminar burning velocity and flame stability, using the methods of constant-pressure spherical flames and counter-flow flames [8–10]. Bunsen flame configuration and flame cone angle was also widely used to measure the flame speed of syngas/air mixtures. Bouvet et al. [11] recently measured the laminar flame speed of H2/CO/air mixtures using Bunsen flame configuration and OH* chemiluminescence technique. They proposed a flame speed correlation for lean mixtures with hydrogen fraction in the fuel blends ranging from 10% to 70%. Dong et al. [12] measured the laminar flame speed over wide range of equivalence ratios and hydrogen fractions using the Bunsen burner and a CCD camera. However, it is very hard to precisely detect the edge of the reaction zone from CCD camera image due to the weak spontaneous emission signal and the systematic errors caused by the rounded flame tip. Planar laser-induced fluorescence (PLIF) can detect the reaction zone through OH radical emissions with high spatial resolution and high signal-to-noise ratio [13–17]. He et al. [18] measured the laminar flame speeds of lean premixed syngas/air mixtures using Bunsen flame configuration and PLIF technique. However, they only focus on the fuel-lean mixtures with low hydrogen content. Yasuhisa et al. [19] studied the turbulent flame structure premixed syngas/air mixtures with a Bunsen burner by using OH-PLIF. They also investigated the flame radiation characteristics using the spectrograph in which the consistency of flame images and radiation spectrum were obtained.

It is noted that in the above literatures, the Bunsen burner was either wrapped with heating tapes or surrounded by cooling liquid, thus the effect of heat exchange between burner and flame was masked. From the engineering point of view, burner temperature is seldom monitored, thus the real flame speed is inherently influenced by the heat exchange between flame and burner.

In this study, a systematic investigation on the combustion characteristics of premixed syngas/air mixtures is conducted using the Bunsen burner over a wide range of mixture compositions. Laminar flame speed, flame structure and radiation spectrum are obtained using OH-PLIF and spectroscopic techniques.

2. Experiments

2.1. Burner and PLIF system

Fig. 1 shows the schematic of experimental apparatus. The system consists of an OH-PLIF system, a gas delivery system and a premixed Bunsen burner. A straight cylindrical stainless steel tube with an inner diameter (D) of 3 mm is employed as the Bunsen flame burner. The length of the tube is 20 mm (>50 D) to ensure the fully developed flow. The flow rate of each gas is precisely monitored by the delicate MKS mass flow controllers (Models 1179A and 1559A) with an accuracy of 1/1000 SLM to provide the desired mixture composition.

One-way valve is equipped on each gas supplying line to prevent the reverse flow. The gases from different lines are mixed in a mixing chamber (400 ml stainless steel) prior to entering the burner. Total flow rate is controlled to ensure the fully developed flow. The flow rate of each gas is precisely monitored by the delicate MKS mass flow controllers (Models 1179A and 1559A) with an accuracy of 1/1000 SLM to provide the desired mixture composition.

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Fig. 1 – Schematic layout of the OH-PLIF measurement and inlet manifold.
The dye used is Coumarin 153 in ethanol which produces a dye beam with a tunable wavelength (from 350 nm to 610 nm), and is then converted by a Frequency Conversion Unit (FCU) that comprises of a BBO doubling crystal, a compensator and a wavelength separator to produce a UV beam at a wavelength from 175 nm to 305 nm with an average power of 10 mJ per pulse. The laser goes through the energy monitor and sheet optics to produce a laser sheet of 30 mm in height, and aligns to pass through the centerline of the burner nozzle.

LIF signals of OH emission at wavelength around 313 nm are detected with intensified Relay Optics (LaVision VC08-0094) and a CCD camera (LaVision Image Prox) equipped with a UV lens (Nikon Rayfact PF 10545MF-UV) and an OH filter. The gate of the intensifier is decreased to about 200 ns to minimize effects of ambient light.

Ensemble average of 150 images is used for each specified case with all OH-PLIF images corrected by subtracting the background image from the experimental image data along with intensity correction through the energy monitor.

### Flame speed measurement

Measurement of laminar flame speed was performed using the flame cone angle method based on the OH-PLIF images. This approach is particularly convenient for the conic-shaped premixed flames with straight-sides. The velocity of unburned mixture $U_0$ at the nozzle exit can be considered to be uniform, and laminar flame speed $S_u$ can be expressed as the normal flow velocity component with respect to the flame surface, that is, $S_u = U_0\sin\alpha$ [20], here $U_0$ is the flow velocity at burner exit and $\alpha$ is half of cone angle of flame, as shown in Fig. 2 [11]. The intensity of the captured OH radial concentration of flame is analyzed by a built-in Davis software.

The equivalence ratio is given by,

$$\varphi = \frac{(F/A)}{(F/A)_{stoch}}$$

(1)

The definition of $X_{H_2}$ is,

$$X_{H_2} = \frac{V_{H_2}}{(V_{H_2} + V_{CO})}$$

(2)

where $(F/A)$ is the fuel/air ratio, and $V_{H_2}$ and $V_{CO}$ is the volume fraction of $H_2$ and CO in the fuel mixture respectively.

Fig. 3 gives the typical intensity of OH radical distribution for flame at $X_{H_2} = 20\%$, $\varphi = 0.8$. The curve gives the intensity on the vertical axis along the center of flame. It can be seen that the peak of OH radical concentration (2500 counts) appears at approximately 55 mm which is the location of the tip of flame cone. From the vertical axis intensity plots, the height of flame cone ($h$) can be calculated and half of cone angle of flame reaction front is derived from the OH-PLIF images by $\alpha = \arctan(d/2h)$. Here $d$ is nozzle diameter.

The OH-PLIF approach is used to directly detect flame geometric parameter. Previous reports with OH-PLIF used some other softwares such as MATLAB and/or FORTRAN programs for the edge detection [18]. Those previous works based on the measurement of emission signal along the flame cones. As it shows in Fig. 3, the peak intensity of flame cone tip captured by the Davis plot is outside the cone reaction layer. Since the height of flame cone is calculated from the OH-PLIF image pixel information, thus it gives more precise information compared to the determining the boundary of flame cone through flame emission signal.

The uncertainty of the measured laminar flame speed which is primarily caused by the resolution of the camera was found to be within 2%.

### Spectrograph measurements

Flame radiation spectrum was measured using a spectrograph (Andor, Shamrock SR-500i) with a cooled multi-channel ICCD photo-detector (Andor, iStar 340T) through a lens which focuses the entire flame onto the slit of the spectrograph. Solis software is used to measure the radiation spectra from 400 nm to 700 nm (visible spectrum).

### Results and discussion

#### Effects of mixture composition on local flame structure and laminar flame speed

Fig. 4 shows the CCD camera captured flame images for stoichiometric syngas/air mixtures at different $X_{H_2}$. Significant reduction in flame front area is observed with increasing $X_{H_2}$. Furthermore, the perceptual bluish flame luminosity decreases and becomes increasingly pinkish with the increase
in $X_{H_2}$. Flame height decreases with increasing $X_{H_2}$ due to the
increase in flame speeds since the Reynolds number of
upstream flows keeps constantly and flow rates of different
mixtures are almost the same. As $X_{H_2}$ increases, the burned
gas flame propagation speed is increased, pushing the flame
front toward the upstream direction for flow balance. Due to
distinction of different flame emissions, flame radiation color
can be distinguished from each other. For example, for low $X_{H_2}$
flame, major emission is CO$_2$; while for high $X_{H_2}$ flame, water
vapor becomes the major emission. Those emissions influence
the visible light of flames, and result in the differences of
flame shape and color.

Fig. 5 shows the calculated overall height of the conic flame
front as a function of $X_{H_2}$ for different equivalence ratios. For
given equivalence ratio, the flame height decreases with the
increase of $X_{H_2}$. At the same $X_{H_2}$, the reduction rate of flame
height decreases as equivalence ratio increases. Additionally,
the variation of flame height with equivalence ratio becomes
less significant as equivalence ratio increases. This is
reasonable since flame speed shows a quasi-parabolic
dependency on equivalence ratio with the peak point at
fuel-rich side, thus around equivalence ratio of peak point, the
variation of flame speed with equivalence ratio is minimized.
As shown in Fig. 2, flame height $h$ is a function of flame speed.
Flame height is less sensitive to the variation of equivalence
ratio as equivalence ratio increases to around stoichiometry.

Fig. 6 shows the OH-PLIF captured flame structure with
four $X_{H_2}$ at equivalence ratio of 1.0. The pinkish region in Fig. 6
represents high OH radical concentration, and its distribution
shifts toward the outer flame boundary with the increase in
$X_{H_2}$ in the syngas mixture. This difference is not obvious in the
normal camera images in Fig. 4 but quite clear in Fig. 6 in the
OH-PLIF images. It is observed that the region of high OH
radical concentration locates along the conic primary reaction
boundary in low $X_{H_2}$ flames. In the case of high $X_{H_2}$ flame, the
region of high OH radical concentration shifts away from
flame cone region. This result in the concurrent occurrence of
two distinctive combustion layers; a premixed layer and a
diffusional one with OH radical concentration focusing on
the outer diffusion layer and flame cone tip. When $X_{H_2}$
increases up to 40%, flame structure appears to resemble the
standard “W” shape, which indicates that burning is intensified
along the conic flame boundary and on flame surface.

Increasing $X_{H_2}$ will make most of intensified burning region to
the diffusion layer. This phenomenon may be caused by the
highly-diffusive nature of hydrogen. Since hydrogen diffuses
faster than oxygen, when mixture flows out of tube, hydrogen
spreads faster than oxygen and is burned away from the
premixed layer. That is why flames with high $X_{H_2}$ are usually
wider and burning is intensified in the diffusional layer. The
OH-PLIF images in Fig. 6 reveals that even under the same
equivalence ratio, flame structure with different syngas
compositions varies significantly.

Laminar flame speeds were measured by OH-PLIF and
Bunsen flame cone angle method. The experimentally deter-
mined laminar flame speeds of syngas flames at different
equivalence ratio and $X_{H_2}$ are given in Fig. 7. The leanest
condition tested for each $X_{H_2}$ composition is determined by

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3.2. Effects of temperature on the burner rim and simulation results

Simulation on laminar flame speed was made to provide the quantitative comparison with the experimentally determined laminar flame speed. Fig. 9 shows the comparison between the experimental results and the simulated results using one-dimensional freely propagating laminar flame model with PREMIX code [21] of CHEMKIN-II [22] at 298 K. In general, the experimentally determined ones show higher values than those of simulation. 

With increase of $X_{H_2}$, the difference between them when taking the simulation temperature as the room temperature becomes larger. Unlike most of the previous works which controlled the upstream burner temperature constantly to exclude the effect of heat exchange between burner and flame on the determination of laminar flame speed. In this work, the burner temperature was not controlled, thus the temperature profile along the center axis of the burner was measured. The effect of upstream temperature on laminar flame speed and temperature change due to the variation of mixture composition can be analyzed.

Temperature is measured along the nozzle tube at different points ($X$ is 2–10 mm) from burner exit using thermocouples. As shown in Fig. 10, significant changes of temperatures along the burner axis are presented, and the corrected temperatures at different mixture compositions are given. In this figure, different lines represent temperature measurements at different $X_{H_2}$. The temperature for correcting flame speed is increased as hydrogen increases. This trend indicates that the difference in flame speed of mixtures is mostly due to the different temperature enhancement of flame.

Using the corrected temperatures in Fig. 10, the flame speed is re-calculated and comparisons of experimental and the simulated values using the corrected temperatures are given in Fig. 11. Simulated laminar flame speed shows good agreement with those of the measured ones. It can be seen that temperature is the key influential factor in the determination of laminar flame speed.

3.3. Chemical kinetics analysis

Kinetics analysis was made with Li’s mechanism [23] and PREMIX code of CHEMKIN-II. Because of the importance of...
small radicals in the chain propagating combustion chemistry, the effect of increasing $X_{\text{H}_2}$ on the rate of production of typical small radicals such as H and OH is examined. Fig. 12 gives the comparison of reactions for the highest rate of production of H radicals and OH radicals at $\phi = 1.2$ and hydrogen fractions of 20% and 80%. As shown in Fig. 12(a), the main contribution of producing H radical is reaction (R29) $\text{CO} + \text{OH} = \text{CO}_2 + \text{H}$ in low $X_{\text{H}_2}$ mixture combustion. For high $X_{\text{H}_2}$ mixture, reaction (R3) $\text{H}_2 + \text{OH} = \text{H}_2\text{O} + \text{H}$ is the dominant reaction and peak production rate increases from 1.322E-3 mol/(cm$^3$ s) to 8.091E-3 mol/(cm$^3$ s) when hydrogen fraction increases from 20% to 80%. For the consumption of H radicals, the dominant reactions for the two $X_{\text{H}_2}$ are the same, Reaction (R1) $\text{H} + \text{O}_2 = \text{O} + \text{OH}$, but peak consumption rate increases significantly when hydrogen fraction increases from 20% to 80%. For the two mixtures, the production of OH radical, as shown in Fig. 12(b), is mainly attributed to reaction R1 ($\text{H} + \text{O}_2 = \text{O} + \text{OH}$), and peak ROP of R1 significantly increases when hydrogen fraction increases from 20% to 80%. However, the reaction with the highest consumption rate of OH radicals changes from reaction R29 ($\text{CO} + \text{OH} = \text{CO}_2 + \text{H}$) to reaction R3 ($\text{H}_2 + \text{OH} = \text{H}_2\text{O} + \text{H}$). They are the two major reactions that contribute to the production of H radicals, as shown in Fig. 12(a). Furthermore, with increase of $X_{\text{H}_2}$, peak ROP of H slightly shifts to the upstream. The increased production rate with increasing $X_{\text{H}_2}$ facilitates the back-diffusion of H radicals and shifts reaction zone to the upstream.

Sensitivity analysis is made to find the dominant reactions on laminar burning speed. Fig. 13 gives the sensitivity of reactions at different $X_{\text{H}_2}$ and two equivalence ratios. As shown in Fig. 13(a), the most important reactions at $X_{\text{H}_2} = 20\%$ is reaction R29. While at $X_{\text{H}_2} = 80\%$, reactions R3 and R1 are two equally important reactions. R3 has positive effect on flame speed at $X_{\text{H}_2} = 40\%$ and 80%, while it has negative effect on flame speed at $X_{\text{H}_2} = 20\%$. This is consistent with the observation in Fig. 12. The dominant reaction for the low $X_{\text{H}_2}$ flames is R29, and the dominant reaction for the high $X_{\text{H}_2}$ flames is R3, thus there is a competition between reaction R29 and R3 in getting the OH radicals. The negative effect depends strongly on $X_{\text{H}_2}$. Fig. 13(b) shows flame speed sensitivity at $\phi = 1.2$, similar to the case at $\phi = 0.8$, with the increase of $X_{\text{H}_2}$, the dominant enhancing reactions to flame speed shifts from R29 to R3.

3.4. Differences in radiation characteristics of syngas laminar premixed flame

Fig. 14 shows the visible flame radiation spectrum (400 – 700 nm) acquired from the spectrograph of four flames as
Three distinctive peaks around 450 nm, 615 nm and 622 nm are subtracted from the flame radiation spectrum for each flame. The difference in radiation spectrum can explain the perceptual coloration becomes more pinkish with the increase of hydrogen because the main products change from CO2 to CO and H2O (through R3), which is evidenced by the ROP of H and OH in Fig. 12. Background light spectrum is shown in Fig. 4. The perceptual flame coloration from bluish to perceptual coloration variation with changing the syngas composition. For the low $X_{H_2}$ (high CO content) flames, the broad-band radiation from the CO–O radiation (i.e., $CO + O \rightarrow CO_2 + h\nu$) contributes to the increase in blue flame appearance. While for the high $X_{H_2}$ flames, the wide peak at 450 nm is not obvious but the later two peaks assumed to be $CH_2^+$ and CHO$^+$ [24] are still obvious but seem to be weaker than those in the low $X_{H_2}$ (high XCO flames).

4. Conclusions

Effects of hydrogen fraction and equivalence ratio on laminar flame speed of syngas were studied using the cone angle method based on OH-PLIF images at atmospheric pressure. Hydrogen fractions from 20% to 80% and equivalence ratios from 0.5 to 1.2 are covered. Kinetic simulations were made using CHEMKIN-II with Li’s mechanism. Flame structures and radiation spectrum were analyzed using PLIF and spectrograph. Main conclusions are summarized as follows:

1. Laminar flame speeds in a temperature-uncontrolled burner are measured using the OH-PLIF Bunsen flame method. Hydrogen fraction in syngas affects the laminar flame speed and flame shape. Good agreement is achieved between the experimentally measured laminar flame speeds and simulated ones when the temperature is corrected.

2. Kinetic analysis on intermediate radicals and sensitivity analysis show that hydrogen plays very important role in accelerating combustion. The main H radical production (or OH radical consumption reactions) changes from reaction R29 (CO + OH → CO2 + H) to reaction R3 (H2 + OH → H2O + H) with the increase of hydrogen fraction in the syngas. Sensitivity analysis also shows that the dominate reaction on the effect of laminar flame speed changes as hydrogen fraction changes in syngas.

3. Difference in flame color of varied H2 mixture is due to their difference in radiation spectrum of the intermediate radicals produced in combustion.

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