Study on the laminar burning velocity of Medium-Btu syngas flame with N\textsubscript{2} dilution based on OH-PLIF technology

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Abstract: The laminar burning velocity of syngas/air premixed flames was studied by Bunsen burner and OH-PLIF system. The mole fractions of nitrogen in syngas change from 0 to 50\% and the equivalence ratios vary from 0.6 to 1.2 were both considered to investigate those effects on the flame speed. A new method called slope averaging method was proposed to calculate the burning velocity and two classical methods were employed to verify its accuracy. A premixed code with GRI 3.0 mechanism was adopted to predict the burning velocity as well as to expose the dominant reactions on the formation of OH and H radicals. Results show that the slope averaging method represents the equal accuracy as the surface area method and is superior to the cone angle method. In addition, the increase of N\textsubscript{2} fractions decreases the flame speed, and the effects of N\textsubscript{2} dilution turn weak at fuel-lean conditions. Furthermore, the dominant reactions on the formation and consumption of OH and H radicals are inhibited as X\textsubscript{N2} increases.

1. Introduction
Accompanied with the rapid development of the world economy, people's living standard has improved steadily. As a result, the demand for energy is increasing gradually. This demand not only embodies on the "quantity", but also requires the "quality", which means that the kind of energy corresponding to nowadays energy consumption must be vast and clean. In recent, syngas which consists of variable components such as H\textsubscript{2}, CO, CH\textsubscript{4}, N\textsubscript{2} and CO\textsubscript{2}, attracts tremendous...
attentions due to its widely source and low contaminant emission. It can be produced from a variety of industrial process, such as steam reforming of natural gases, gasification of coal and biomass, or even organic wastes (Kim, Lee, Kim, & Sohn, 2011). The utilization of syngas in Integrated Gasification Combined Cycle (IGCC) plants for business has a history of more than decades (FU J et al., 2013), whose superiority is its high efficiency and cleanliness, in comparison with traditional coal-fired power plants (Ratna et al., 2011). Thus, it promises to be one of the most competitive technologies in power generation (Chobthiangtham et al., 2015). However, a critical process in IGCC is the combustion of the syngas in the gas turbine whose operation security is closely related to the properties of the fuel (Bhattacharya et al., 2011). As one of the significant way of combustion, lean premixed combustion (LPC) seems to be preferred in consequence of the higher energy efficiency and lower pollutant emissions, while the combustion instability and flashback seem to restrict its application (Huang et al., 2009). Therefore, conducting a research into fundamental characteristics of syngas premixed flames will benefit the design of the combustion chamber of gas turbine.

The complex compositions of syngas lead to its intricate combustion properties. Besides, the content of each component in the syngas is influenced by a series of factors in the producing process, such as raw material, gasification medium, temperature, gasifier and so on (Ahmed et al., 2012; Naine et al., 2016). Undoubtedly, it is essential to have a better understanding of the combustion before its application to engineering. One concern about the combustion is the laminar flame speed, which has been reported for several experimental approaches such as heat flux method (Dirrenberger, Le Gall, Bounaceur, Glaude, & Battin-Leclerc, 2015; Goswami, Bastiaans, Konnov, & de Goey, 2014), spherical flame method (Chen, 2011; Jiang, Shy, & Li et al., 2016), counterflow method (Contino, Jeanmart, & Lucchini et al., 2011; Voss, Hartl, & Hasse, 2014) and Bunsen flame configuration (Dong et al., 2009; Natarajan, Lieuwen, & Seitzman, 2007). Bunsen flame configuration is widely used in syngas/air premixed flame speed measurement, on account of the advantages that it provides clear flame structures, obvious flame front as well as its reliability (Fu, Tang, Jin, & Huang, 2014). Because the rising development of PLIF technology, the contour of the flame front surface is able to be abstracted according to the OH images (Fan, Dong, & Tang, 2011; Han, Cai, Xu, Bruno, & Abdelkrim, 2014), which make the surface area methodology possible. Bouvet, Chauveau, Gökalp, Lee, and Santoro (2011) measured the laminar flame speed of syngas mixtures (H₂/CO/Air) and the equivalence ratios ranges from 0.3 to 1.2. Both the flame surface area and cone angle methodologies are adopted, indicating that the former yields an overall good accuracy when compared to the available experiment data for H₂/Air mixtures. Lee, Jeong, and Lee (2015) studied the burning velocity of syngas-air premixed flames with a wide equivalence ratios(φ) varies from 0.5 to 5.0, and the experimental results showed that both the cone angle and the surface area methods were in good agreement with the numerical calculations at φ < 2.0, but overestimating the numerical calculations at 2.0 < φ < 5.0. To simplify the research, current investigations focus more on the main combustible components CO/H₂. However, the properties of the practical syngas that encountered in industry act much more complicated. Other components in the mixtures also perform significant effects on the combustion flame.

In the current study, laminar burning velocities of practical syngas with equivalence ratios varying from 0.6 to 1.2 were studied both experimentally and simulatively. To study the effect of the dilution gas, N₂ was adding to the syngas in varying proportions (15%, 35%, 50%), in consideration of the medium which made gasification products contained a variable N₂. And the low-heat value (LHV) of the syngas decreased with the increase of the X_N₂. To calculate the flame speed, a new method called slope averaging method were applied, and two classical methods (cone angle and surface area methods) were employed as comparison. All of these three methodologies were based on images that obtained by OH-PLIF technology with a Bunsen burner. Numerical simulation was conducted with GRI 3.0 mechanism through using a premix code in CHEMKIN-PRO, to predict the burning velocity and analyze the dominate reactions in the syngas/air premixed flame.
2. Experimental approaches

2.1. OH-PLIF measuring system

The schematic of experimental system is shown in Figure 1. A Bunsen burner with the outlet diameter (d) of 5mm and the length of 260 mm (>50d) is designed and fabricated. The Nd: YAG laser (quanta-ray) is used to generate the pulsed source laser with the baseline wavelength of 1064 nm and the frequency of 10 Hz. At the stern of the Nd: YAG laser, there is a THG crystal arm, which performs as a harmonic generator in purpose of tuning the laser into pump beam at 355 nm. The Dye laser (Sirah) working with the chemical solutions (solvend: coumarin 153; solvent: ethanol) is able to obtain tuneable beam which ranges from 517 to 574 nm. As a result, when setting a frequency doubling crystal at the outlet of the Dye laser, the wavelength from 258.5 to 287 nm of the UV beam is available according to the demand. The energy monitor and sheet optics are used to transform the pointolite into a focused laser sheet of 50mm in height and about 0.8 mm in thickness, which then goes through the center axial of the Bunsen burner.

LIF signals of OH radical are recorded by an ICCD camera which comprises an OH-LIF filter to filter interference signals. The length of the object that the ICCD camera records is calibrated by a graduated plate with the system software Davis 8. The gate and delay of the ICCD camera are set to 100ns and 60ns, for purpose of minimizing the effects of ambient lights. The gain of the intensifier is increased to 60% to optimize the captured fluorescence signals.

The required gases (H₂, CO, CH₄, N₂, CO₂ and Air) are stored separately in each high pressure gas cylinder, the purity of which is 99.99%. Each gas is controlled by corrected rotor flowmeter and is filled into the chamber to prepare the required mixture. Flame arrester is applied for preventing pipeline explosion caused by tempering. Flow controlling valves can effectively avoid gas backflow. The combination of Flame arrester and Flow controlling valves ensures the safety of the experiment process.
In the current experiment, the power of the UV beam is 13mJ per pulse, and the wavelength is 283.23 nm, which can best excite the OH radical. For each specified case, 200 OH-PLIF images are taken for average processing and corrected by subtracting the background image.

### 2.2. Medium-Btu syngas samples and combustion system

In order to investigate the properties of the practical Medium-Btu syngas, a kind of syngas produced by co-gasification of coal and biomass is studied in the current study. And the initial components data of the syngas (Table 1) is derived from other reference (Liqun, Xu, & Haosheng, 2008). It is necessary to note that a slight adjustment is made on the initial components to investigate the effects of the nitrogen dilution on the combustion process.

Each gas of the mixtures is separately stored in gas cylinder before the experiment. The flow rate of each gas is well controlled by calibrated rotor flow meter, and a mixing chamber is set to ensure the gases’ homogeneous mixing. The stoichiometric ratio in the current experiment varies from 0.6 to 1.2, and the Reynolds number is set to be 1000. One-way valve in the gas delivery system can prevent the gas circumfluence, when used in conjunction with a flame arrester near the Bunsen burner, which can effectively promise the security of the premix combustion research. The Bunsen burner used in the experiment is a straight stainless cylinder tube, in consideration of the sickness of the laser sheet (0.8 mm) and the quantity of the flow rate, and the inner diameter of the tube is 5 mm. To ensure the fully developed flow, the whole length of the burner is designed to be 260 mm.

### 2.3. Burning velocity measurement

In the current research, three kinds of calculation methods based on Bunsen flame and OH-PLIF images are minutely illuminated. Here, the contour of the flame front is extracted from the OH-PLIF images by marking the positions of the OH radicals that have the maximum concentration on the flame front curve, and then which are collected for polynomial fitting with Origin 9.0 program. Therefore, the equation of the flame front curve can be expressed as:

\[ y = f(x) = \sum_{i=0}^{n} a_i x^i \]  

(1)

#### 2.3.1. Cone angle method

This method is widely used in Bunsen flames. According to the flame inner angle \( \theta \) and the velocity of the unburned gas flow \( U_0 \), the burning velocity can be calculated as follows (Wang, Wei, & Meng et al., 2014)

\[ S_u = U_0 \sin \frac{\theta}{2} = U_0 \times \frac{d}{2 \sqrt{h^2 + \frac{d^2}{4}}} \]  

(2)

In the current study, the position of the peak concentration of OH radicals is found, based on the OH-PLIF images as shown in Figure 2, which is used to precisely determine the height of the inner flame \( h \). It might be properly used in the situations where the flame front section is a triangle shape.

| Case | Components | CH₄ | H₂ | CO | CO₂ | N₂ | LHV |
|------|------------|-----|----|----|-----|----|-----|
| \( X_{N2} = 0\% \) |              | 9.8 | 47.2 | 27 | 16 | 0 | 12.03 |
| \( X_{N2} = 15\% \) |              | 8.33 | 40.12 | 22.95 | 13.6 | 15 | 10.22 |
| \( X_{N2} = 35\% \) |              | 6.37 | 30.68 | 17.55 | 10.4 | 35 | 7.82 |
| \( X_{N2} = 50\% \) |              | 4.9 | 23.6 | 13.5 | 8 | 50 | 6.01 |
2.3.2. Flame surface area method
This method is also a classical approach, which is used in rim stabilized conical flames. One significant issue on this method is to precisely obtain the surface area (S) of the flame front. Actually, OH-PLIF technology provides a possibility to extract the flame front curve, which is useful for calculating the surface area. Thus, the flame speed can be expressed as (Bouvet et al., 2011):

$$S_u = \frac{Q_0}{S}$$  \hspace{1cm} (3)

where $Q_0$ is the volume rate of the flow. It is worth mentioning that when the function of the curve is a straight line, Equation (3) can be simplified as Equation (2), which means that the cone angle method could be regarded as a special circumstance of the flame surface area method.

2.3.3. Slope averaging method
As shown in Figure 3, this method also depends on the flame front curve, and it can be detected from the cone angle method. Only half of the flame front curve is extracted for computation, in consideration of the axisymmetric characteristics of the Bunsen flame. With the function of the front curve, it is possible to calculate the slope on each point of the curve. As a result, the burning velocity on each corresponding location can be well expressed.

$$S_u(x_i) = U_0 \sin \alpha = U_0 \times \frac{1}{\sqrt{f'(x)^2 + 1}}$$  \hspace{1cm} (4)

Here, $U_0$ is the initial speed of the unburned gas flow at the nozzle, $\sin \alpha$ can be expressed as the function of the coordinate $x$. The mean speed of the flame could be obtained by averaging $S_u(x_i)$:

$$S_u = \frac{1}{n} \sum_{i=1}^{n} S_u(x_i)$$  \hspace{1cm} (5)

When $n$ tends to be infinite, the average flame speed should be:

$$S_u = \lim_{n \to \infty} \frac{1}{n} \sum_{i=1}^{n} S_u(x_i) = \frac{1}{b-a} \int_{a}^{b} S_u(x_i) dx$$  \hspace{1cm} (6)

The constants “a” and “b” are the starting and ending position on the curve. Interestingly, when the function of the curve is a straight line, this method performs the same property as the second method, and is equal to the cone angle method.

Figure 2. OH concentration distribution and inner height of syngas/air premixed Bunsen flame.

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Typically, the correlation coefficient is set as the criteria to determine the accuracy level and $R^2$ is the proportionally value between the predicted and actual results (Ganesan, Rajakarunakaran, Thirugnanasambandam, & Devaraj, 2015). The $R^2$ can be calculated using the following expression:

$$R = \frac{1}{N} \sum_{i=1}^{n} \frac{(X_i - \bar{X})(Y_i - \bar{Y})}{\sigma_X \sigma_Y}$$  \hspace{1cm} (7)

Where $N$ is the number of sample. $\frac{X_i - \bar{X}}{\sigma_X}$ represents the standard score of the sample $X_i$, $\bar{X}$ is the sample average and $\sigma_X$ means the sample standard deviation.

### 3. Results and discussions

#### 3.1. Effects of mixture composition on OH radical distribution

Figure 4 shows the syngas/air flame images obtained via CCD camera for varying $N_2$ fractions at $\phi = 1.05$. One distinguishing feature that can be observed with the images is the change on the height of the inner flame, which increases along with the increasing of the $X_{N2}$. The main reason that leads to this phenomenon is that the addition of the $N_2$ decelerates the burning velocities of the flame by influencing the temperature. Because of the increasing of the flame front surface area, more adequate reaction conditions are available to the fuel. Thus, another effect on the flame structure is the constriction of the reaction zone of the flame when $X_{N2}$ increases, where the height of outer flame decreases slightly.

Figure 5 shows the distribution of OH radicals of the flame for different $X_{N2}$ at the equivalence ratio of 1.05. Different from the images captured by CCD camera in Figure 4, OH-PLIF images are more suitable for clearly reflecting the reaction region. The different colours represent the different concentration of OH radicals as shown in Figure 2. The density of the OH radicals decreases as the increasing of $X_{N2}$, especially at the root of the flame, due to the poor producing rate of the OH radicals at high $N_2$ dilution. What’s more, the flame front is observed moving to the outer flame with the addition of the $N_2$, for the shrinking of the reaction area.
3.2. Comparison of three methods on flame speed calculation

Three kinds of calculation results based on experiment and the simulation results predicted by numerical computation with different N\textsubscript{2} fractions are exhibited in Figure 6 separately. A consistent Reynolds number of 1000 for each case is adopted to minimize the effects of flow rate. All the
Experimental results slightly exceed the numerical computed results due to the effects of the temperature on the burner rim, which contribute to the raising on the initial temperatures of the mixture upstream as well as the activation energy of the unburned fuel. As a result, the practical combustion rates are higher than simulations at the same conditions. Especially for fuel-lean cases, the reaction areas are more closely to the nozzle of the burner, which leads to a more obvious heating phenomenon on the rim. And the initial speed of the upstream $U_0$ is no longer a constant, while the jet distance increases. Therefore, the difference between predicted flame speed and experimental results is more obvious.

As shown in Figure 6, the laminar burning velocities of the syngas/air mixtures calculated by three methods are in good agreement. The maximum relative error between cone angle method and surface area method is less than 9%, while the results of the slope averaging method keep more consistent with the later where the maximum difference is within 5% deviation. Besides, the co-efficient of determination value ($R^2$) between slope averaging method and surface area method of Figure 6 a,b,c and d is obtained as 0.99922, 0.99925, 0.9981 and 0.98867 respectively, which is very close to 1. It is a strong indication that the slope averaging method performs the same accuracy with the surface area method, and is superior to the cone angle method. Although both the former two precise methods are depend on the contour of the flame front, in contrast, the surface area method is more strict with the extraction accuracy, where the physical dimension of the flame front dominates the computational accuracy. Actually, the errors caused by image definition on extraction and the process of polynomial fitting are inevitable, which directly affect the accuracy of the surface area method. However, the slope averaging method is not depend on the actual size of the front contour and only is determined by the slope of the curve, which effectively avoids the errors mentioned above. That is to say, the slope averaging method might be more practical and high rate of fault tolerance when the high accuracy is also guaranteed. As for high hydrogen fraction syngas, the tip
opening phenomenon probably occurs, which increases the difficulty of the extraction of the integrate flame front. At the moment, slope averaging method has its advantage to calculate the flame speed with segmental front curve.

3.3. Burning velocity analysis based on slope averaging method
Actually, both the cone angle method and the surface area method are based on the assumption that the speed of the unburned upstream is a constant. However, it’s necessary to take the speed attenuation into account while the jet distance is more than four times of the diameter. It’s possible for the slope averaging method to take this factor into consideration during calculation by correcting the $U_0$ with the empirical equation (Kuo, 2015) of the free propagation model of the jet flow:

$$U_0(x, y) = \begin{cases} U_0 & y \leq 4.1875d \\ 0.5U_m(1 + \cos \frac{\pi x}{2y \tan \alpha_u}) & y \leq 4.1875d \end{cases}$$

(8)

Here, the $U_0(x, y)$ is the speed of the upstream on each point of the flame front curve, and the $\alpha_u$ is the half angle of the jet flow which approaches to 4.85°. $U_m$ is the central speed of the upstream, which is given by:

$$U_m = \frac{0.48U_0}{\frac{y}{\lambda} + 0.145}$$

(9)

The $\lambda$ is a constant which equals to 0.08.

Figure 7 shows the overall results of the laminar burning velocities of the syngas/air premixed flame measured by slope averaging method with and without $U_0$ correction. With $U_0$ correction, the burning velocity at fuel-lean condition decreases a lot, and this phenomenon becomes more obvious with the increase of the $X_{N_2}$, which shows a better agreement to the simulation results. The reason for this trend is mainly due to the fact that part of the flame front is located in the speed attenuation area, while the flame has low burning velocity. Besides, the decrease in heat release, the increase in heat capacity of the mixture with dilution and significant reductions of flame temperature and thermal diffusivity of the mixture, all of which signals a decrease in laminar burning velocity values (Prathap, Ray, & Ravi, 2008).

3.4. The effects of $N_2$ dilution on dominant reactions
The H and OH radicals play important roles in the combustion flame of the syngas mixtures. To study the parameters in the process of the formation and consumption of these radicals, a simulation analysis based on GRI 3.0 mechanism is conducted in CHEMKIN-PRO with a premixed code. To be synchronous with the experimental results, simulation conditions are made in consistent with the experimental conditions.
Figure 8 shows the total rate of production (ROP) on OH radicals at varying N$_2$ dilution and equivalence ratio. It is obvious to note that the ROP of OH radicals increases along with the equivalence ratio within experimental parameters, mainly due to the more adequate fuel in the premixed mixtures. However, it decreases as X$_{N2}$ increases at the same equivalence ratio. It is worth mentioning that as the X$_{N2}$ increases not only the proportion of the combustible components but also the temperature of the flame is reducing. Comparing $\phi$ = 1.2 and $\phi$ = 0.6, the effects caused by N$_2$ dilution on the composition changes turn to weakness. The cardinal parameters contributing to the decrease of the flame speed is the poor ROP of the intermediate radicals such as OH radicals at high nitrogen fraction and low equivalence ratio.

Figure 9 exhibits the production rates and the dominant reactions for the formation and consumption of the H and OH radicals for X$_{N2}$ = 15% and 50% at $\phi$ = 1.2. Table 2 gives the specification of the dominant reactions in the GRI 3.0 mechanism, which is presented in the pictures.

As shown in the Figure 9, the main contribution for the producing of the OH radicals is R38, while R84 is the primary consumption step. These two reactions are also the highest ROP of H radicals, but influence the formation of the H radicals on the other way around. As shown in Figure 8 (a, c), the magnitude of the ROP of the OH radicals presents a decrease along with the increase of the X$_{N2}$. One significant change on the dominant reactions is the R3 which is the most sensitive to the N$_2$ dilution when X$_{N2}$ varies from 15 to 50%. This is mainly because the addition of the N$_2$ contributes directly to the reduction of the concentration of H$_2$ in the syngas/air premixed mixture. By contrast, the reaction R46 acts nearly impregnability with the varying X$_{N2}$ at $\phi$ = 1.2. Thus the ROP of OH radicals on R46 surpasses that of the R3, which makes the R46 becoming the reaction that produces OH radicals in second highest level when X$_{N2}$ = 50%. The producing rate of H radicals decreases as the increasing of the X$_{N2}$, whereas each reaction on the formation of H radicals which is influenced by N$_2$ shows the almost same trend.

Sensitivity analysis is made to reveal the effects on dominant reactions when the syngas is diluted with N$_2$. It is worth figuring out that partial N$_2$ derives from the oxidizer (Air), thus the sensitivity analysis is also made at the case of X$_{N2}$ = 0%. Figure 10 shows the sensitivity of reactions with varying N$_2$ dilutions at equivalence ratio of 1.2 and 0.6. With the increasing of the X$_{N2}$, the dominant reaction of OH radicals’ production (R38) and consumption (R84) is heavily inhibited. Especially at $\phi$ = 0.6, both the reaction of R38 and R84 are strengthened by reverse, which makes a further explanation to the lower ROP of OH radicals at low
equivalence ratios. As shown in Figure 10(I), the reaction of R99 is inhibited at low $X_{N_2}$, while it is promoted at $X_{N_2} = 50\%$. This phenomenon is quite different at $\phi = 0.6$, where the reaction is promoted most at $X_{N_2} = 15\%$, which indicates that the $N_2$ has a positive effect on R99 at low proportions in the premixed mixture, however it plays a negative role in high proportions on the contrary. However, this effect on R99 can't determine the total ROP of H and OH radicals, due to its poor influence on the consumption of OH radicals as well as the producing of H radicals.

4. Conclusions
Effects of $N_2$ dilution and the equivalence ratio on the laminar burning velocities of syngas/air premixed flame are studied both experimentally and simulatively. A new computation method
(slope averaging method) is proposed on the purpose of improving the calculation accuracy yet pragmatic for the flame speed investigation fields. It is free from the flame front shape in calculating flame speed and thus is more broadly applicable than the other two methods. The main results can be formulated as following:

(1) Three methods which are adopted to calculate the experimental flame speed are in good agreement. But the experimental results are slightly greater than the simulations, due to the heated burner rim. While in the fuel-lean cases, the speed attenuation of the unburned upstream can’t be ignored.

(2) Comparing the calculation results, slope averaging method performs the same accuracy as the flame surface area method. However, the former has its unique advantages to consider the speed attenuation by correcting $U_0$. Thus, the results in speed attenuation area are more precise.

(3) Both the experimental and numerical results indicate that the effects of N\textsubscript{2} dilution turn to weakness as the equivalence ratio decreasing, due to slightly composition changes at fuel-lean premixed conditions.

**Nomenclature Table**

| Symbol   | Description                      |
|----------|----------------------------------|
| $X_{N_2}$ | Mole fraction of species N\textsubscript{2} |
| $\phi$   | Equivalence ratio                |
| LHV      | Low heat value                   |
| IGCC     | Integrated Gasification Combined Cycle |
| LPC      | Lean premixed combustion         |
| PLIF     | Planar Laser Induced Fluorescence |
| $d$      | Outlet diameter of Bunsen burner |
| $\theta$ | Flame inner angle                |
| $U_0$    | Unburned gas flow velocity       |
| $h$      | Height of the inner flame        |
| $S$      | Surface area of the flame front  |
| $S_a$    | Burning velocity                 |
| $Q_0$    | Volume rate of the flow          |
| $R^2$    | Co-efficient of multiple determination |
| $N$      | Number of sample                 |
| $X_i$, $Y_i$ | Sample value, Equationm(7) |
\( \bar{X}, \bar{Y} \)  
Sample average, Equation (7)

\( \sigma_{X}, \sigma_{Y} \)  
Sample standard deviation, Equation (7)

\( U_{0}(x, y) \)  
Speed of the upstream on each point of the flame front curve

\( \alpha_{u} \)  
Half angle of the jet flow

\( U_{m} \)  
Central speed of the upstream

\( \lambda \)  
A constant which equals to 0.08

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