

Development of Chemiluminescence Sensor for Equivalence Ratio and Temperature Measurements in Turbulent Hydrocarbon Flames

T. S. Cheng¹, Y.-Y. Cheng², Y.-C. Chao², Y.-H. Li², and C.-Y. Wu³

¹Department of Mechanical Engineering
Chung Hua University, Hsinchu, 300, Taiwan, ROC

²Department of Aeronautics and Astronautics
National Cheng Kung University, Tainan, 701, Taiwan, ROC

³Advanced Engine Research Center
Kao Yuan University, Kaohsiung County, 821 Taiwan, ROC

1 Introduction

It has been shown that the chemiluminescence emissions of OH*, CH*, and C₂*, resulted from electronically excited state, in hydrocarbon flames can be related to chemical reaction rate and heat release rate [1]. The ratios of chemiluminescence of CH*/OH* and C₂*/OH* were used to determine local equivalence ratio in laminar and turbulent flames [2, 3]. Most of these chemiluminescence measurements used traditional lenses to collect the global emissions, and there is insufficient spatial resolution to measure the local equivalence ratio at the flame front. To overcome this deficiency, a Cassegrain optics with high spatial resolution must be used [3-5]. In the present study, a Cassegrain optics coupled with an optical fiber and a monochromator is used to simultaneously detect OH*, CH*, and C₂* emissions in premixed CH₄-air flames. The correlation between the intensity ratio (CH*/OH*, C₂*/CH*, and C₂*/OH*) and the equivalence ratio and the ratio of C₂* emissions at two different vibrational bands are obtained to elucidate the capability of simultaneous measurements of local equivalence ratio and temperature in turbulent premixed hydrocarbon flames.

2 Experiments

A Bunsen burner (i.d. = 10 mm) is used to produce laminar premixed CH₄-air flames operated at several different equivalence ratios ranging from fuel-lean to fuel-rich conditions ($\phi = 0.85-1.5$) for system calibration. To generate the laminar premixed flames, the exit velocity of the flow is kept at 1 m/s so that the flowrates of fuel and air are varied. Chemiluminescence signals emanating from the sample volume are collected and focused by the Cassegrain optics and relayed to the entrance slit of a monochromator (Spectral Products, DSK 242, 1/4 m, $f/\# = 3.9$) through a 2-m long optical fiber (core diameter = 100 μm). The Cassegrain optics consists of a primary and a secondary mirror, which avoids the generation of chromatic aberrations for different wavelengths. The monochromator is coupled with a liquid nitrogen cooled charge-coupled device (LN-CCD). For turbulent flame measurements, a larger diameter jet burner (i.d. = 20 mm) is employed to produce turbulent premixed

flames operated at $\phi = 1.0$ and 1.3 . The exit velocity and Reynolds number are 2.5 m/s and 3200 for $\phi = 1.0$ and 1.3 turbulent flames.

3 Vibrational Temperature Measurement

The flame temperature can be measured using the intensity ratio of C_2^* emission intensities at two vibrational bands as reported by Ishiguro et al. [6]. The total emission intensity from a band can be expressed as [7]:

$$I = \sum_{\nu''} \frac{I_{em}^{\nu'\nu''}}{\nu''^4} = \frac{64}{3} \pi^4 c \sum_{\nu'} N_{\nu'} = \frac{64}{3} \pi^4 c \sum_{\nu'} \frac{N}{Q_{\nu}} e^{-G_0(\nu')hc/kT} = \frac{C}{Q_{\nu}} \sum_{\nu'} e^{-G_0(\nu')hc/kT} \quad (2)$$

where Q_{ν} is the vibrational partition function and $G_0(\nu')$ is the energy term value above the zero-point energy and C is the grouped constant.

Because the emission of $C_2^*(1, 0)$ band starts from $\nu' = 1$ and that of $C_2^*(0, 0)$ band starts from $\nu' = 0$, therefore the intensity ratio of the two vibrational bands can be expressed as:

$$\frac{I_{\Delta\nu=1}}{I_{\Delta\nu=0}} = \frac{C_1 \sum_{\nu'=1}^{\infty} e^{-G_0(\nu')hc/kT}}{C_2 \sum_{\nu'=0}^{\infty} e^{-G_0(\nu')hc/kT}} = \frac{C_1 \frac{e^{-T_v/T}}{1 - e^{-T_v/T}}}{C_2 \frac{1}{1 - e^{-T_v/T}}} = C_3 e^{-T_v/T} \quad (3)$$

$$T = \frac{T_v}{\ln(C_3) - \ln(I_{ratio})} \quad (4)$$

where the characteristic vibrational temperature for C_2 molecule is $T_v = 2572.89$ K and C_3 is the instrument constant which can be determined from the calibration.

4 Results and Discussion

Typical chemiluminescence emission spectra from the laminar premixed CH_4 /air Bunsen flames operated at $\phi = 0.85$, 1.0 , and 1.3 are shown in Fig. 1. It can be seen from Fig. 1 that for $\phi = 0.85$, the peak emissions of $OH^*(0, 0)$ at 307 nm, $CH^*(0, 0)$ at 430 nm, $C_2^*(1, 0)$ at 470 nm, and $C_2^*(0, 0)$ at 516 nm are visible in the spectrum. In addition to the peak emissions, there is also a broad underlying CO_2^* continuum that extends from 280 nm throughout the sampled spectral region. This broadband emission varies with equivalence ratio and is especially significant when the spectral bandwidth of the detection system is broad. Therefore, to obtain a more accurate measurement of chemiluminescence intensities, background correction based on the side bands must be made. The polynomial fits of the side bands used for background corrections are also shown in the figure (red dashed curves). The curve fit value at ~ 310 nm is subtracted from the intensity measured, to obtain a corrected OH^* intensity, and likewise at ~ 430 nm for CH^* and from ~ 450 to ~ 590 nm for C_2^* . When the equivalence ratio is increased to 1.0 , the peak intensities of $OH^*(0, 0)$, $CH^*(0, 0)$, and C_2^* Swan band all increase. The intensity of C_2^* Swan band reaches to a maximum value at $\phi = 1.3$. Since the chemiluminescence intensity of OH^* , CH^* , and C_2^* varies with flame equivalence ratio, the ratio of CH^*/OH^* , C_2^*/OH^* , and C_2^*/CH^* can be used to determine flame local equivalence ratio.

In order to obtain a relationship between the intensity ratio and equivalence ratio for turbulent flame measurements, the intensity ratio of C_2^*/CH^* , C_2^*/OH^* , and CH^*/OH^* against the equivalence ratio at the local flame front of the laminar premixed methane-air jet flames at $h = 9$ mm is shown in Fig. 2. It can be seen that the C_2^*/CH^* , C_2^*/OH^* , and CH^*/OH^* ratios increase linearly with increasing equivalence ratio over the range from $\phi = 0.85$ to 1.3 . The high degree of correlation suggests that the local flame stoichiometry at the flame front of premixed CH_4 -air flames can be determined by the spatially resolved chemiluminescence measurements. However, it seems difficult to measure the equivalence ratio in CH_4 -air flames for $\phi > 1.35$ by using this system because the nonlinear

relationship is observed within this range. These findings are in excellent agreement with those obtained by Kojima et al. [3]. The log curve fit equations of the dependence of C_2^*/CH^* , C_2^*/OH^* , and CH^*/OH^* on the equivalence ratio are also shown in Fig.2. These curve fit equations are used for equivalence ratio measurements in turbulent flames. For all calibration conditions and for a given value of equivalence ratio, the C_2^*/CH^* , C_2^*/OH^* , and CH^*/OH^* ratios are within $\pm 10\%$ of the value given by each curve fit equation, which leads to an uncertainty of approximately ± 0.1 for determination of equivalence ratio for $0.85 \leq \phi \leq 1.3$. For temperature measurement, the instrument constant C_3 appeared in Eq. (4) must be determined from calibration. Thermocouple temperature measurements are performed in laminar premixed flames at $h = 9$ mm and at the radial location where the maximum emission intensity occurs. The measured flame temperatures and the intensity ratios of $C_2^*(1, 0)/C_2^*(0, 0)$ are used to obtain the instrument constant C_3 from Eq. (4).

Simultaneous measurements of OH^* , CH^* , $C_2^*(1, 0)$, and $C_2^*(0, 0)$ chemiluminescence emissions are made in turbulent premixed methane-air jet flames ($\phi = 1.0$ and 1.3) to examine the applicability of the developed sensor system for simultaneous measurements of equivalence ratio and temperature. By applying the equations obtained from the laminar premixed flames, the histograms of measured equivalence ratio in turbulent premixed stoichiometric CH_4 -air flame at $h = 10$ mm and $r = 0$ mm are shown in Fig. 3. The measured average equivalence ratios from the emission intensity ratio of C_2^*/CH^* , C_2^*/OH^* , and CH^*/OH^* are $\phi = 1.02$, 0.99 , and 0.95 , respectively. Similar measurements of the local equivalence ratio are made in turbulent premixed rich CH_4 -air flame at $h = 60$ mm and $r = 0$ mm (not shown here). The measured average equivalence ratios from the emission intensity ratio of C_2^*/CH^* , C_2^*/OH^* , and CH^*/OH^* are $\phi = 1.35$, 1.30 , and 1.22 , respectively. The histograms of temperatures measured in the center of turbulent premixed stoichiometric and rich CH_4 -air flames at $h = 20$ mm and $h = 60$ mm, respectively, are shown in Fig. 4. The measured mean temperatures using the emission intensity ratio of $C_2^*(1, 0)/C_2^*(0, 0)$ for $\phi = 1.0$ and 1.3 are 1834 and 1692 K, respectively. The flame temperatures measured by thermocouple are 1751 and 1636 K for $\phi = 1.0$ and 1.3 flames, respectively. The flame temperatures measured by chemiluminescence sensor are in good agreement with those measured by thermocouple.

4 Conclusions

A low cost, non-laser based optical sensor that consists of a Cassegrain optics coupled with an optical fiber and a monochromator is developed for simultaneous measurements of local equivalence ratio and temperature in turbulent premixed hydrocarbon flames. Experimental results show that the high correlations between the integrated intensity ratios of C_2^*/CH^* , C_2^*/OH^* , and CH^*/OH^* and the equivalence ratio can be used to determine the local flame stoichiometry. Of these three intensity ratios, the C_2^*/CH^* curve is the most sensitive to the equivalence ratio when $1.0 \leq \phi \leq 1.3$. The CH^*/OH^* can be used to determine the local flame stoichiometry in the reaction zone of premixed hydrocarbon flames for a wider range of equivalence ratios. Simultaneous measurements of OH^* , CH^* , $C_2^*(1, 0)$, and $C_2^*(0, 0)$ chemiluminescence emissions are made in turbulent premixed CH_4 -air jet flames ($\phi = 1.0$ and 1.3) to demonstrate the applicability of the developed sensor system for simultaneous measurements of local equivalence ratio and temperature with reasonable accuracy.

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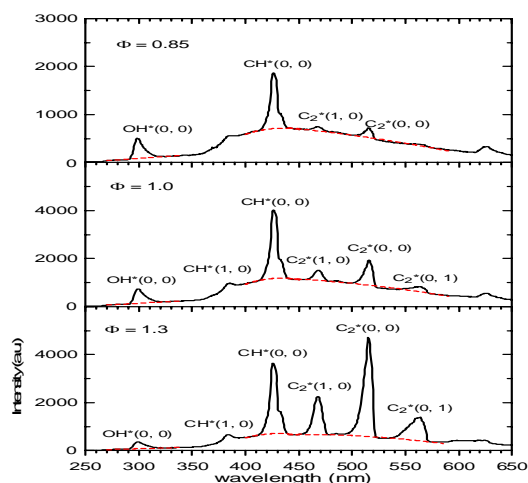


Figure 1. Flame emission spectra measured from laminar premixed methane-air flames at $\phi = 0.85, 1.0,$ and 1.3 .

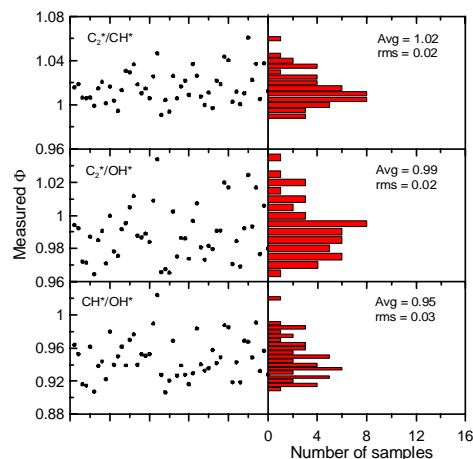


Figure 3. Histograms of measured equivalence ratio in turbulent premixed stoichiometric CH₄-air flame at $h = 20$ mm and $r = 0$ mm.

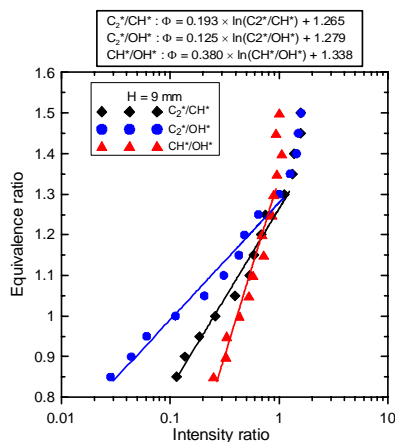


Figure 2. Correlation of the intensity ratios of C₂*/CH*, C₂*/OH* and CH*/OH* to the equivalence ratios at $h = 9$ mm.

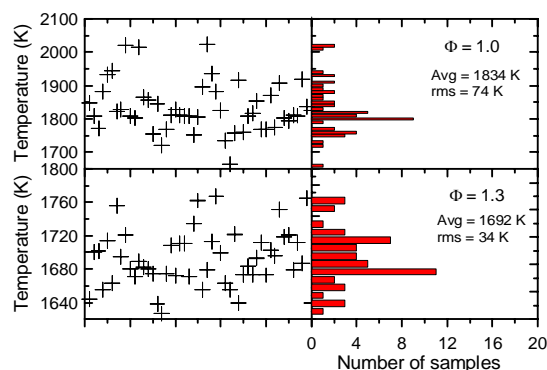


Figure 4. Histograms of measured temperatures measured in the center of turbulent premixed stoichiometric and rich CH₄-air flames at $h = 20$ mm and $h = 60$ mm, respectively.