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Chemiluminescence-based multivariate sensing of local equivalence ratios in premixed atmospheric methane–air flames

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ABSTRACT

Chemiluminescence emissions from OH*, CH*, C_2^*, and CO_2^* formed within the reaction zone of premixed flames depend upon the fuel–air equivalence ratio in the burning mixture. In the present paper, a new partial least square regression (PLS-R) based multivariate sensing methodology is investigated and compared with an OH*/CH* intensity ratio-based calibration model for sensing equivalence ratio in atmospheric methane–air premixed flames. Five replications of spectral data at nine different equivalence ratios ranging from 0.73 to 1.48 were used in the calibration of both models. During model development, the PLS-R model was initially validated with the calibration data set using the leave-one-out cross validation technique. Since the PLS-R model used the entire raw spectral intensities, it did not need the nonlinear background subtraction of CO_2^* emission that is required for typical OH*/CH* intensity ratio calibrations. An unbiased spectral data set (not used in the PLS-R model development), for 28 different equivalence ratio conditions ranging from 0.71 to 1.67, was used to predict equivalence ratios using the PLS-R and the intensity ratio calibration models. It was found that the equivalence ratios predicted with the PLS-R based multivariate calibration model matched the experimentally measured equivalence ratios within 7%; whereas, the OH*/CH* intensity ratio calibration grossly underpredicted equivalence ratios in comparison to measured equivalence ratios, especially under rich conditions ($\varphi > 1.2$). The practical implications of the chemiluminescence-based multivariate equivalence ratio sensing methodology are also discussed.

1. Introduction

Real time measurement of local and global fuel–air equivalence ratios (φ) is essential for monitoring and closed-loop control of premixed combustion systems. Pollutant emissions such as oxides of nitrogen (NO_x), carbon monoxide (CO), unburned hydrocarbons (HC), and particulate matter (PM) can be reduced by controlling fuel–air equivalence ratios [1–3]. Precise control of equivalence ratio can also help in preventing serious accidents in gas turbines such as blow-off, flashback due to pressure waves, or damage to the combustor due to combustion oscillations as these turbines operate close to the lean limit [3,4]. In internal combustion (IC) engines, advanced combustion strategies such as homogeneous charge compression ignition (HCCI) [5], low temperature combustion [6], and direct injection spark ignition [7] utilize premixed or partially premixed combustion modes to simultaneously improve engine efficiencies and reduce pollutant emissions. To achieve controlled premixed or partially premixed combustion in IC engines, the real-time measurement and closed-loop control of in-cylinder local equivalence ratios are desirable. The present work is an attempt to utilize natural chemiluminescence emissions to diagnose equivalence ratios in premixed atmospheric methane–air flames with two different sensing methodologies: (1) a whole-spectrum multivariate calibration model and (2) an OH*/CH* peak intensity ratio calibration model.

Chemiluminescence emissions from OH*, CH*, and C_2^* formed within the reaction zone of premixed flames depend upon the air–fuel ratio in the burning mixture [8–10]. Easy, nonintrusive experimental detection of chemiluminescence coupled with fast response times provides a convenient approach for equivalence ratio determination. Many researchers have investigated the variations in OH*, CH*, and C_2^* intensity ratios with equivalence ratio [11–13]. For example, Docquier et al. [11] studied the effect of pressure on OH*, CH*, and C_2^* chemiluminescence intensity in methane–air premixed flames. They found a strong correlation between the chemiluminescence signal and pressure and

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Abbreviations: LOOCV, leave-one-out cross validation; PCs, principal components; PLS-R, partial least-square regression; RMS, root mean square; RMSEC, root mean square error in calibration; RMSEP, root mean square error in prediction.

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suggested the need for a multi-wavelength sensor for direct monitoring of equivalence ratio. Hardalupas and Orain [2] showed that in natural-gas-fueled, premixed, counter-flow flames OH*/CH* is independent of strain rate and that C_2^*/CH* and C_2^*/OH* show a strong dependence on strain rate. In a recent study [14], it was shown that in premixed counter-flow methanol and ethanol flames, OH*/CH* intensity ratio also shows dependence on strain rate. The aforementioned studies clearly indicate the need for the development of a robust sensing methodology that is applicable over a range of combustion conditions.

Many approaches have being employed for sensing equivalence ratios from chemiluminescence spectra [15]. Muruganandam et al. [3] and Hardalupas et al. [16] used the peak intensity ratio of OH*/CH* for sensing equivalence ratio in combustors fueled with natural gas. Artificial intelligence-based data analysis techniques have also been explored for monitoring as well as controlling combustion processes [17]. Recently, Ballester et al. [18] applied the artificial neural network technique on chemiluminescence spectra obtained from combustion of natural gas blended with hydrogen in a swirl combustor for monitoring equivalence ratios.

All of the previous studies show that the chemiluminescence emissions of OH*, CH*, C_2^*, and CO_2^* (the nonlinear continuous background) from the reaction zone depends on the equivalence ratio. In this work, a new multi-wavelength (multivariate) sensing methodology is proposed for the determination of equivalence ratios in premixed methane–air flames. Most of the previously reported work did not consider the whole spectrum approach. A model based on multivariate statistics is relevant for a two-dimensional (*X*, *Y* variables) data set, where the response *Y*-variable (equivalence ratio) depends on several explanatory *X*-variables (spectral wavelengths). A wide chemiluminescence spectrum (250–650 nm) was used to develop the multivariate calibration model. This approach ensures that the spectral intensity variations from all the excited species within the measured wavelength range are used in the development of the multivariate calibration model. The experimental results presented in this paper demonstrate that the multivariate calibration model can differentiate fuel-rich, fuel-lean, and stoichiometric premixed flames. A partial least square regression (PLS-R) based multivariate calibration model is developed and it is shown that the PLS-R model successfully predicts the equivalence ratios within the developed calibration range.

2. Specific objectives

The specific objectives of the present work are

1. To develop a multi-wavelength sensing methodology for equivalence ratio estimation and monitoring in premixed atmospheric methane–air flames.
2. To evaluate the predictive capability of the developed multivariate calibration model and compare its performance with an OH*/CH* intensity ratio calibration model over a range of equivalence ratios.

3. Experimental setup

A rectangular slot burner (length: 11.2 cm, width: 0.55 cm, and height: 26 cm) was used to generate the atmospheric, premixed methane–air flames used in this study. Fuel (99.97%-pure methane) and oxidizer (breathing grade air) were mixed at room temperature, prior to entering the slot burner from the bottom end. Correlated rotameters (±2% accuracy full scale) were used to measure the flow rates of methane and air. A schematic diagram of the experimental setup is shown in Fig. 1. To cut-off fuel supply to the burner in case of an emergency; a safety shutdown switch was used in the fuel line as shown in Fig. 1. A flash arrester was also attached to the fuel cylinder as a safety precaution.

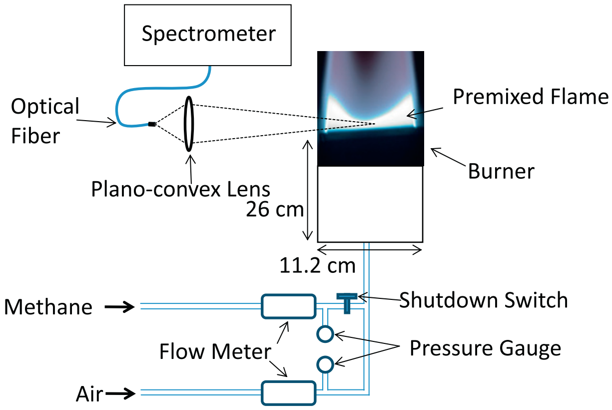

Fig. 1. Schematic diagram of the experimental setup.

Flame equivalence ratios were calculated from the volume flow rates of fuel and air measured with the rotameters and the measured air and fuel pressures (±1% accuracy of full scale) at the outlet of the rotameters. Both fuel and air at the rotameter outlets were assumed to be at ambient temperature (298 K) and the ideal gas equation of state was used to obtain the respective mass flow rates. The equivalence ratio was defined as follows:

$$Equivalence\ ratio\ (\varphi) = \frac{AFR_s}{AFR_a}. \tag{1}$$

where AFR_s is the stoichiometric mass-based air–fuel ratio (17.16 for methane–air combustion), and AFR_a is the actual mass-based air–fuel ratio computed from measured fuel and air mass flow rates.

The uncertainty of the measured equivalence ratios in this study was between 3 and 5%. The uncertainty was estimated using the following equation [19]:

$$\frac{U_\varphi^2}{\varphi^2} = \left(\frac{V_f}{\varphi}\frac{\partial\varphi}{\partial V_f}\right)^2\left(\frac{U_{V_f}}{V_f}\right)^2 + \left(\frac{V_a}{\varphi}\frac{\partial\varphi}{\partial V_a}\right)^2\left(\frac{U_{V_a}}{V_a}\right)^2 + \left(\frac{P_f}{\varphi}\frac{\partial\varphi}{\partial P_f}\right)^2\left(\frac{U_{P_f}}{P_f}\right)^2 + \left(\frac{P_a}{\varphi}\frac{\partial\varphi}{\partial P_a}\right)^2\left(\frac{U_{P_a}}{P_a}\right)^2 \tag{2}$$

where φ is equivalence ratio, V_f and V_a are fuel and air volume flow rates respectively, P_f and P_a are the pressures of fuel and air at the outlets of the respective flow meters, U_φ, U_{vf}, U_{va}, U_{Pf}, and U_{Pa} are the uncertainties associated with φ, V_f, V_a, P_f and P_a, respectively.

All optical measurements were performed approximately 2 mm above the burner edge. A fused silica, plano-convex, spherical lens with 10 cm focal length and 1.27 cm diameter focused at the center of the burner was used to collect the chemiluminescence spectra. The collected optical signal was coupled to the single end of a bifurcated optical fiber (Ocean Optics, QBIF400-UV–VIS). One of the bifurcated ends of the optical fiber was connected to an Ocean Optics USB 2000 spectrometer (600 grooves/mm grating) with a spectral resolution of 3.8 nm (full-width half-maximum) to record the chemiluminescence emission spectra. The other end was used to locate spatial position in the flame by sending a separate laser beam through it before the experimental measurements were performed. This end was closed while recording the chemiluminescence spectra. Each spectrum was acquired with a 25 ms exposure time and each recorded spectrum was an average of 20 accumulated spectra. A total of 10 such averaged spectra were recorded for each equivalence ratio condition. It is well known that the chemiluminescence emissions are superimposed on the dark current background spectrum. Hence, the dark current background was also collected for each experimental condition by blocking the