



Full Length Article

## Effect of producer gas addition and air excess ratio on natural gas flame luminescence



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### ABSTRACT

This paper presents the emission spectroscopy method for registering chemiluminescent radical species of  $\text{OH}^*$ ,  $\text{CH}^*$ ,  $\text{C}_2^*$  by means of optical flame imaging using optical bandpass filters, corresponding to emission wavelengths of the radicals of interest, and by scanning the flame using optical fibers coupled to a spectrometer for recording the emission spectra. The aim of this research is to analyse the distribution of chemiluminescence intensity along the flame axis from various angles simultaneously in order to determine the effect of the air equivalence ratio (ER) and mixing of producer gas (PG) from biomass gasification into the natural gas flow to spectral characteristics of flame at specific wavelengths representing formation reactions of  $\text{OH}^*$  radical species. The experiments were carried out with pure natural gas (NG) and air mixture and a premixed air/gas mixtures (80%NG and 20% PG, 65%NG and 35%PG). Flows of air, NG and PG were premixed before entering the combustion chamber. For flame emission spectroscopy, two different methods were used: 1) imaging by an ICCD camera through optical bandpass filters suitable for  $\text{OH}^*$ ,  $\text{CH}^*$ ,  $\text{C}_2^*$  chemiluminescence intensity registration; 2) by a combination of a spectrometer and five optical fibers to collect the flame spectra from 5 different angles and in various heights from the burner outlet. The distribution of chemiluminescent species intensity along the burner vertical axis was analyzed and zones with most intense  $\text{OH}^*$ ,  $\text{CH}^*$ ,  $\text{C}_2^*$  generation in flame were identified, and the effect of addition of PG to NG and ER to the  $\text{OH}^*$  chemiluminescence was analysed.

### 1. Introduction

As the estimated worldwide use of natural gas (NG) is projected to increase by nearly 12% in 2020, one of the strategic objective is the reduction of emissions from stationary combustion plants using gases [1]. Due to these reasons alternative fuels are being sought to partially or fully replace natural gas. One of such fuel type is producer gas (PG) created from biomass via the gasification process. During the gasification process, low quality solid fuel is converted into more valuable combustible gases. These gases are mixtures of methane, hydrogen ( $\text{H}_2$ ), carbon monoxide (CO) and inert gas such as carbon dioxide ( $\text{CO}_2$ ), nitrogen ( $\text{N}_2$ ) [2]. Due to its composition, producer gas could be used to substitute natural gas in various combustion devices such as water boilers, drying units, chemical heating, incineration, ceramic kilns etc. through combustion of the gas in a burner. Despite the great potential, the combustion characteristics of PG are different from those of NG. According to several studies [3,4], PG is more prone to flashback, has different behavior of flame stability and different dynamic responses. Also in mixtures of PG and NG the heat release rate profiles are wider

for PG, compared to NG, indicating slower flame propagation in the combustion chamber. This affects the combustion stability and results in power loss, cyclic variation, maximum pressure and temperature decrease in thermal applications when trying to mix or completely substitute NG with PG [5]. On the other hand, some works have demonstrated [6,7], that the hydrogen rich PG advances the gas mixture ignition and extends the flammability limits, thus ensuring the flame stability. In this way, burning the fuel with higher air excess ratio the flame temperature decreases, leading to the decrease in  $\text{NO}_x$  generation. Due to these reasons, it is necessary to understand the specifics of the combustion process in order to design novel burners for producer gas or adapt the existing ones. One of the most promising non-intrusive combustion process optimization technique is flame chemiluminescence monitoring [8–10]. In a chemiluminescent reaction, a part of the released energy is used to excite an electronic state which is short-lived and relaxes through a number of mechanisms, including collisional non-radiative deactivation and spontaneous photon emission. In the case of photon emission, species emit at a characteristic wavelength, signaling the presence of the species [11]. Some studies of emission

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spectroscopy with an ICCD camera [12,13] show that strongest chemiluminescent intensities can be registered at the beginning of the flame, near the burner outlet. According to [14–17], spectral emission of hydroxyl radical ( $\text{OH}^*$ ) correlates with the equivalence ratio (ER, denoted as  $\alpha$ ), therefore, it can be used for combustion process measurements and control as it is one of the most important parameters for effective and clean combustion. A few authors point out that ER in some cases can be monitored from the  $\text{OH}^*/\text{CH}^*$  or  $\text{OH}$  intensity relations [12,18]. Other authors [19] working on chemiluminescence emission of premixed syngas flames determined that  $\text{OH}^*/\text{CO}_2^*$  ratio is suitable to monitor stoichiometry of combustion but only in a certain range of ER. Hernandez et al. [6] performed research on  $\text{OH}^*$  and  $\text{CH}^*$  radical emissions from the combustion of NG and PG and have found that the presence of  $\text{OH}^*$  radical could indicate formation of  $\text{NO}_x$ . The spatial distribution of radical chemiluminescence intensity values measured in flame by emission spectroscopy method was also modeled with CHEMKIN resulting in acceptable similarities with experimental data [18]. Modeling with GRIMEch also confirmed the relation between ER and  $\text{OH}^*$ ,  $\text{CH}$  with multiple methane oxidation mechanisms and an additional sub-mechanism that accounts for the formation and destruction of the chemiluminescent species  $\text{OH}^*$  and  $\text{CH}^*$  [20]. For more in-depth analysis, the combination of a spectrograph, an ICCD camera and optical fibers are often used [21]. By measuring a narrow point in the flame, light from an optical fiber is dispersed into spectra and then can be analyzed in depth [22]. Parameswaran et al. [23] used a fiber probe connected to a spectrometer to acquire the radiation from flames generated with various gas mixtures. The results showed that the  $\text{OH}^*$  peak intensity has a strong correlation with ER and with heating value of the gas mixture as well. However, to obtain more precise results, the use of optical fibers from multiple angles enables to measure such properties as the flame radial front (defined as regions with strongest chemiluminescence emissions) and the precise flame curvature [24,25]. Besides, in most cases flame is turbulent and non symmetrical in industrial boilers, therefore, using a single sensor to scan a limited flame region cannot provide adequate diagnostics for, e.g., automated process control. Therefore, it is necessary to scan the whole combustion area and to identify the most informative flame regions for positioning the sensor in order to maintain the optimal combustion regime, including ER. For these reasons, this work aims to apply a spatial flame chemiluminescence scanning technique to compare  $\text{OH}^*$ ,  $\text{CH}^*$  and  $\text{C}_2^*$  intensity values in the line-of-sight chemiluminescence intensity contour along the flame axis when using both pure natural gas and its mixture with producer gas in various proportions. Also, this article presents comparison of chemiluminescence along the flame axis from five different angles registered at same time with optical fibers and flame imaging through optical bandpass filters using an ICCD camera.

## 2. Materials and methods

### 2.1. Experimental setup and procedure

The experiments were performed using the experimental setup consisting of an air/gas supply system, a gasification reactor system for producer gas supply, a combustion chamber, and a flame optical analysis system. Schematic diagram of the experimental setup is presented in Fig. 1. The images of flame in the burner are shown in Fig. 2. In order to estimate the flame characteristics of PG addition to NG, the air equivalence ratio  $\alpha$  was set in range from 1.0 to 1.3 to recreate optimal combustion conditions used in industrial boiler burners. The constant 0.6 kW thermal output of the burner for the mixture was maintained in all the experiments for result comparability (Table 1). The combustion process was organized in a combustion chamber made of a 56 cm high and 6 cm diameter quartz glass pipe. A Bunsen type burner was modified to work in a closed chamber with premixed gas flow by adding a premixing chamber and a mesh type bluff body for flame stabilisation. The burner was fired by mixtures of natural gas (NG) and producer gas

(PG) at different proportions (Table 1). NG, PG and air flows were connected to the premixing chamber via steel pipes. For each measurement of flame chemiluminescence, an initial volumetric gas flow was set with flow meters (operating range 0.5 ÷ 20 l/min) (Table 1). At the exit of the combustion chamber, the flue gas analyzer probe was inserted at a 50 cm distance from the burner nozzle. The samplings with a gas analyzer were made relatively far from the combustion zone to recreate conditions of an industrial flue gas measurement system. These result were later compared to the results obtained by using the ICCD camera in with varying ER. The sampling and composition measurement of flue gas was used to analyse whether the spectral data of flame emission can indicate the combustion parameters similarly to gas sampling with a faster response time.

### 2.2. Preparation of air/gas supply

Producer gas was generated by using a lab scale gasification reactor [26]. The reactor was filled with pine and spruce wood pellets and gasified at the temperature of 800 °C. The producer gas generated in the process was transported from the reactor to the burner by  $\text{N}_2$  used as the carrier gas. In all the experiments, the same batch of commercially available wood pellets was used and producer gas was extracted at stable operation regime of the reactor. By measuring the gas composition multiple times, no significant variation in gas composition was detected, therefore, it was assumed to be constant. Natural gas was supplied from the district gas distribution grid. The compositions of producer gas (measured) and natural gas (from supplier data sheet) are presented in Table 2. Air was supplied from the compressed air system located in the research facility. Both flows were controlled with flow meters (operating range 0.5 ÷ 20 l/min). The inlet parameters for fuel and air flows are given in Table 1.

### 2.3. Optical system

The whole combustion process was observed using an optical system for analysis of spatial distribution of the excited species  $\text{OH}^*$ ,  $\text{CH}^*$  and  $\text{C}_2^*$  in the flame at atmospheric pressure. The flame images were captured using an ICCD (Intensified Charge Coupled Device) camera Andor iStar DH734. The diameter of the photocathode (intensifier) was 18 mm; a pixel size was 13  $\mu\text{m}$ . The matrix contains 1024 × 1024 active pixels sensitive to 200–800 nm wavelength emissions. The Equivalent Background Illuminance (EBI) is  $<0.2 \text{ e}^-/\text{pix}/\text{sec}$ . The readout noise is as low as 2.9  $\text{e}^-$ . The peak quantum efficiency at room temperature is 25%. For spectral analysis, the spectrometer Andor Shamrock SR-303i coupled to the camera was used. The spectrometer focal length is 303 mm, the aperture  $f/4$ . The grating resolution was 300 lines/mm.

In order to compare the spatial distribution of selected radicals, two types of experiments were carried out:

- Flame imaging: the camera is focused to capture the entire image of the flame; an optical filter is placed in front of the camera objective, thereby, the flame images were recorded at different wavelengths corresponding to emissions of the different radicals (Fig. 1 and Table 4);
- Flame scanning: 5 optical fibers were placed around the flame in a ring-shaped holder, with its plane oriented horizontally, to provide the spatially-integrated spectra obtained from different angles. The plane was scanned along the vertical axis of the flame and the spatial distribution of the radicals in the flame was thereby obtained (Fig. 3).

Both experiments are described in more detail below.

#### 2.3.1. Flame imaging

To obtain a spatially resolved distribution of radical species in the

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