Experimental and Numerical Studies of Paraguayan Chaco Natural Gas in a Counterflow Diffusion Flame

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Abstract: Combustion is a chemical phenomenon in which a multitude of elementary chemical reactions take place, resulting in the overall process of fuel oxidation. Natural gas fuel has been explored for a few decades and extracted for a few years in the region of Paraguayan Chaco, near Bolivia border. Currently, natural gas is not very important in Paraguay's energy matrix, however it could be in the near future if higher volumes are extracted and transported to the most populated cities, specially to the capital. In order to improve Paraguayan natural gas combustion performance, an understanding of its fundamental properties and the combustion pathways is required. This study presents new data for Paraguayan Chaco natural gas combustion in a laminar counterflow diffusion flame configuration at atmospheric pressure. Visible chemiluminescence of excited radicals CH^* and C_2^* is employed experimentally. 1D numerical simulation was carried out using Paraguayan Chaco natural gas chemical composition and a standard kinetic mechanism, to which we added CH^* and C_2^* reactions. Typical flame structures resulting from simulation are presented and a validation of the model is realized comparing experimental and numerical CH^* and C_2^* radicals profiles.

Key words: Natural gas, counterflow, diffusion flames, numerical simulation, kinetic modeling, chemiluminescence

1. Introduction

Mastering energy is a key factor for human to preserve its economic growth and maintain his life standard. According to the International Energy Agency (IEA) report and Shahid et al. [1], the world will increase by 40 % its energy requirements in 2035.

Taking into account all primary energy produced around the world, combustion of liquid, gas and solid fuels corresponds to about 85% of the total. These resources are non-renewable, therefore combustion efficiency is one of the way to overcome this limitation. In the same time, due to climate changes, lower emissions to the atmosphere are one of the main requirements nowadays in most combustion systems and a challenge for research.

This work aims to study the combustion of

Paraguayan Chaco natural gas, from experimental and numerical point of view, for which the counterflow diffusion (non-premixed) flame configuration at atmospheric pressure and constant injection temperature was chosen.

Combustion is a chemical phenomenon in which a multitude of elementary chemical reactions take place, resulting in the overall process of fuel oxidation and reduction. During these elementary chemical reactions, the chemical species are transformed into other species. These reaction intermediates may absorb part of the reaction energy placing them in an excited state. A possible way of de-excitation is the spontaneous emission of photons from the populated state of the specific species, called chemiluminescence. Chemiluminescence is the emission of light by molecules returning to their ground state after being excited chemically, rather than thermally. The emission shows a weak dependence on temperature and a greater dependence on chemical composition. One way to understand these complex chemical

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processes is to follow the evolution of chemiluminescence of specific species produced in the reaction processes.

In order to acquire a better understanding of combustion fundamental phenomena, it is preferable to study flames configurations with flows relatively simple that are easy to control.

The counterflow laminar flames meet these criteria: the flow is well known, it can be modeled from a theoretical point of view and experimentally it can be controlled in a relatively easy manner.

The interest of these stretched flames also lies in the fact that they act as the main element to describe some turbulent combustion models.

The primary combustion zone of hydrocarbon flames is distinct from the other parts of the flame by its emission of mostly visible bands, namely CH bands at 431 nm ($A^2\Delta \rightarrow X^2\Pi$) and C_2 band at 516 nm ($d^3\Pi_g \rightarrow a^3\Pi_u$) (Swan band).

A convenient way to experimentally study the flame behavior is to analyze the space and time resolved emission of excited species CH^* and C_2^* , which are not at equilibrium under flame conditions. Indeed, these radicals are naturally present in the combustion zone and permit to determine important macroscopic properties such as flame location, flame speed, equivalence ratio and heat release rate fluctuations.

We intend to study two flames operating conditions (equivalence ratio and strain rate), for which the existing experimental and computational equipments of the Mechanical and Energy Laboratory were used. The flame structure of natural gas diffusion flame will be analyzed. CH^* and C_2^* experimental and numerical profiles will be compared to validate the chosen chemical mechanism as well as added CH^* and C_2^* reactions.

2. Experimental Configuration

2.1 Natural Gas Chemical Composition

The natural gas chemical composition was analyzed

using Gas Chromatography and Mass Spectrometry (GC-MS) techniques. This information is needed to estimate the equivalence ratio of natural gas flames, and also is used as input values to carry out numerical simulations.

The estimation of natural gas composition is presented in Table 1. In comparison to literature values, it corresponds to the classical composition of this fuel, where the major component is methane, with some minor components, as shown in Table 1. 'Others' (with a mole fraction $\times 100$ of 0.987) refers to species not present in the chosen kinetic scheme. This point will be discussed in Section 3.2.

2.2 Counterflow Burner Device

Flames experiments were carried out in a counterflow burner. This configuration is briefly described in the next paragraph. More information of the burner can be found in [2, 3].

The burner consists of two opposed axisymmetric convergent nozzles of 20 mm inner diameter (see Figure 1A). A flow of air is injected at ambient temperature in the upper side of the burner, while a mixture of natural gas and nitrogen is transported from the lower side, also at ambient temperature. Nitrogen component in this mixture is added in order to compensate the momentum of the air flowing from the upper side of the burner. After ignition, a laminar diffusion flame is formed between the nozzles. The distance between the two nozzle exits was kept constant to 20 mm . Each nozzle contains two layers of honeycomb to reduce turbulent structures in the plenum chamber of the nozzle. Each nozzle is

Table 1 Composition of Paraguayan Chaco natural gas.

Paraguayan Chaco Gas Composition	
Components	Mole fraction $\times 100$
Nitrogen	3.905
Carbon Dioxide	0.030
Methane	94.277
Ethane	1.611
Others	0.987
Total	100

surrounded by a coaxial nozzle fed by nitrogen, which isolates the reaction zone from ambient perturbations that could disturb the measurements.

In order to limit the thermal effect of the burnt gases circulating in the upper zone of the system, the upper burner has a thermal screen, as well as an isolator (glass wool) with plaster to optimize the temperature uniformity, as one can see in Figure 1A.

Two natural gas flames operating conditions have been chosen, considering the equivalence ratio and the strain rate: Flame I, with an equivalence ratio $\phi = 4$ and strain rate $\varepsilon = 134 \text{ s}^{-1}$, and Flame II, with an equivalence ratio $\phi = 3.5$ and strain rate $\varepsilon = 136 \text{ s}^{-1}$. The injection temperature was kept constant throughout the study in both nozzle exits (Injection Temperature = 300 K).

Strain rates (dV/dz) are estimated using the following equation:

$$\varepsilon = \frac{2V_u}{H} \left(1 + \frac{V_l}{V_u} \sqrt{\frac{\rho_l}{\rho_u}} \right) \quad (1)$$

Where V_u , V_l , ρ_u , ρ_l are the upper and lower nozzle exits velocities and densities, respectively. H is the distance between the two nozzle exits.

All gaseous mass flow rates are controlled by the mass flow meters with an accuracy of $\pm 1.5\%$ and a repeatability of $\pm 0.5\%$ of the full scale. The overall uncertainty of strain rates and natural gas flames equivalence ratios can therefore be estimated at $\pm 0.78\%$ and $\pm 1.52\%$, respectively.

2.3 Visualization

Images of CH^* and C_2^* emission have been recorded using a 576×384 pixels ICCD camera (Princeton Instruments) equipped with a lens (Nikkor, 105 mm focal length) (see Figure 1B).

Narrow-band interference filters were interposed along the optical path for capturing CH^* and C_2^* emission. The filter used for CH^* has 60% transmission and a 10nm wide bandpass centered around 430 nm. The C_2^* filter is centered at 515 nm, 30 nm bandpass and 70% transmission in

Fig. 1 (A) Counterflow burner device, (B) Visualization setup.

the maximum. The exposition time was kept constant at 30 ms.

Figure 2A presents a typical natural gas flame front obtained from CH^* visualization. This figure shows that the flame front is slightly convex due to gravity effects, but near the normal axial direction to the flame surface it remains plane and similarity hypothesis may be applied.

The line of sight creates an integrated signal along its trajectory and increases the emitted signals. As the flame front is axis-symmetrical, Abel integral inversion is used to eliminate the effects of the integration and get the tomography trace of the flame front in the symmetry plane of the burner as done in [4, 5] (Fig. 2B, Fig. 3).

Axial profiles of both CH^* and C_2^* are normalized to corresponding maximum peaks. Due to experimental difficulties, emission intensity calibration was not performed. Therefore, the comparison with numerical profiles will be done considering relative intensities of the position of both CH^* and C_2^* radicals, and not the absolute concentration.

Fig. 2 (A) Averaged image of Flame I CH^* emission, (B) Abel Inversion of the averaged image of Flame I CH^* emission.

Fig. 3 Averaged (Integrated in the line of sight) and Abel inverted axial profiles of Flame I CH^* emission.

The resolution of the CCD camera along the flame axis is $77 \mu m$. Then, due to the slight flames oscillation, the flame position was estimated with an accuracy of $\pm 115 \mu m$.

3. Numerical Simulation

3.1 Governing Equations

We consider an axisymmetric counterflow configuration shown in Figure 4. Air is injected from the left side whereas a mixture of natural gas and nitrogen is injected from the right side. We model our system using similarity approach by searching for similar solutions of gaseous flow equations in the vicinity of the central axis [7]. These similar solutions have the form: gas density $\rho = \rho(z)$, gas radial

velocity $u = rU(z)$, gas axial velocity $v = v(z)$, gas temperature $T = T(z)$ and species mass fractions $Y_k = Y_k(z)$, $k = 1, \dots, N_{sp}$ (N_{sp} is the number of species).

By assuming a pressure gradient in the radial direction so that $-\frac{1}{r} \frac{\partial p}{\partial z} = J$ is constant along the z axis, the gaseous phase are described by the following balance equations [7]:

$$2\rho U + \frac{\partial \rho v}{\partial z} = 0 \quad (2)$$

$$\rho U^2 + \rho v \frac{\partial U}{\partial z} = \frac{\partial}{\partial z} \left(\mu \frac{\partial U}{\partial z} \right) + J \quad (3)$$

$$\rho v c_p \frac{\partial T}{\partial z} = \frac{\partial}{\partial z} \left(\lambda \frac{\partial T}{\partial z} \right) -$$

$$\sum_{k=1}^K h_k W_k \Omega_k - \left(\sum_{k=1}^K Y_k V_{kz} c_{pk} \right) \frac{\partial T}{\partial z} \quad (4)$$

$$\rho v \frac{\partial Y_k}{\partial z} = - \frac{\partial}{\partial z} (\rho Y_k V_{kz}) + W_k \Omega_k, \quad k = 1, \dots, N_{sp} \quad (5)$$

In these equations μ is the gaseous mixture viscosity. c_{pk} and c_p are the heat capacity at local constant pressure of species k and of the mixture respectively. h_k , W_k , Ω_k are the specific enthalpy, the molar weight and the molar chemical production rate of the k^{th} species respectively, and V_{kz} is the diffusion velocity of the k^{th} species in the axial direction.

The above system of equations is completed by the ideal gas equation and by specifying that $\frac{\partial J}{\partial z} = 0$.

Fig. 4 Counterflow diffusion flame configuration.

3.2 Numerical Conditions

The boundary conditions of the system are given in Table 2.

The set of equations is then replaced by a fully coupled set of discrete relations. The solution of this system is based on a global adaptive nonlinear method using Newton iterations (see [8, 9]). The grid is adapted to first and second order derivatives of all variables and the smallest grid size $5 \mu m$.

The kinetic modeling for natural gas oxidation in the counterflow flame was performed using the REGATH-1D-COUNTERFLOW code of the REGATH package with detailed thermochemical and transport properties [7]. The inputs to each simulation include a chemical kinetic reaction mechanism, a dataset of thermochemical properties and a dataset of transport properties.

A methane mechanism (Lindstedt [10] including 29 species and 141 reactions) was chosen to carry the simulations. We have chosen this mechanism for the natural gas/air combustion due to the composition of this fuel (see Table 1), where it can be seen that methane is the main component of this fuel. In order to take into account natural gas composition, species N_2 , CO_2 , CH_4 and C_2H_6 were introduced as input. Furthermore, the mole fractions of these species were normalized in order to have the total mole fraction equals 1 ('Others' species are not present in Lindstedt chemical scheme).

3.3 CH^* and C_2^* Reactions

CH^* and C_2^* species are not present in Lindstedt [10] mechanism, therefore chemical reactions describing the production, quenching and chemiluminescence of CH^* and C_2^* were added to this mechanism. Also species C_3 was introduced since it appears in C_2^* production reactions.

As expected, due to the low concentration of the excited species, no significant difference in the results was found with the addition of these elementary reactions. The thermochemical data for CH^* and C_2^*

Table 2 Boundary conditions.

	Left ($z = -10$)	Right ($z = 10$)
Gas Temperature	$T_g = T_g^{left}$	$T_g = T_g^{right}$
Species mass fractions	$Y_k = Y_k^{left}$	$Y_k = Y_k^{right}$
Gas axial velocity	$v_g = v_g^{left}$	$v_g = v_g^{right}$
Gas radial velocity	$u_g = 0$	$u_g = 0$

were added as well. The transport coefficients for the excited species were the same as those of the ground state species.

For species CH^* and specially for C_2^* literature data is very scarce. However, Kathrotia et al. [11] recently presented production and destruction reactions for these species. Therefore, [11] was chosen for estimating CH^* and C_2^* species concentration.

4. Results and Discussion

4.1 Natural Gas Flame Structure

Figure 5 presents a typical natural gas flame structure. The boundary conditions correspond to the Flame I (see Section 2.2). In this figure $z = -10$ corresponds to the upper burner edge and $z = 10$ to the lower burner edge. The diffusion natural gas/air flame front is located at about $z = 0.1 mm$. At this point, the gas temperature passes through a maximum (1820 K).

In Figure 5 (top), methane (CH_4) species profile remains constant until $z = 3 mm$, then decreases rapidly before reaching the natural gas/air flame front. On the left side of the figure, Oxygen (O_2) species profile remains constant until $z = -2 mm$ and then also decreases rapidly before reaching the natural gas/air flame front. In this type of flame, molecular diffusion between species is the most important phenomenon.

In Figure 5 (bottom), water vapor (H_2O) species profile is presented, and as it can be seen the profile is very similar to that of temperature. H_2O is one of the main products of natural gas combustion. In hydrocarbons flames reactions usually begin with hydrogen (H) abstraction. In natural gas, methane (CH_4) is the main component; therefore hydrogen (H)

Fig. 5 Species and temperature profiles of natural gas, Flame I ($\phi = 4$ and $\varepsilon = 134 \text{ s}^{-1}$).

abstraction produces methyl radical (CH_3). Consequently, as CH_4 starts to decrease at about $z = 3 \text{ mm}$ (Figure 5 (bottom)), CH_3 starts to increase. Methyl radical in turn decomposes into other radicals, explaining its presence in a thin region.

4.2 Experimental and Numerical CH^* and C_2^* Axial Profiles

Figure 6 gives a comparison between experimental and numerical profiles of CH^* (top) and C_2^* (bottom) radicals for the natural gas/air Flame I.

As the measurements do not give absolute values, in order to make comparisons with the numerical results, the experimental profiles of CH^* and C_2^* were normalized by their maximum values. In Figure 7 is presented the same comparison but with the profiles of Flame II.

Fig. 6 Experimental and numerical CH^* (top) and C_2^* (bottom) species profiles, Flame I ($\phi = 4$ and $\varepsilon = 134 \text{ s}^{-1}$).

As we can see in Figures 6 and 7 (top), the numerical predictions of CH^* are very similar to the experimental profiles along the axis. The positions of the peaks match almost perfectly. The experimental profiles are slightly thicker than the simulated ones, however this can be explained by the poorer spatial resolution in the experiments ($77 \mu\text{m}$).

However, as we can see in Figures 6 and 7 (bottom), the numerical predictions of C_2^* are slightly displaced compared to those of the experimental profiles along the axis. For Flame I, the difference between the numerical and experimental C_2^* is less than 1 mm .

For Flame II, the difference is slightly higher than 1 mm . Considering the experimental errors presented in Sections 2.2 and 2.3 and taking into account the distance between the two burners (20 mm), these

Fig. 7 Experimental and numerical CH^* (top) and C_2^* (bottom) species profiles, Flame II ($\phi = 3.5$ and $\varepsilon = 136 \text{ s}^{-1}$)

differences are not very important, therefore this good agreement between experimental and numerical CH^* and C_2^* radicals validates the chosen kinetic mechanism, and consequently the reactions of these radicals added to it.

5. Conclusions

Paraguayan Chaco natural gas combustion in a laminar counterflow diffusion flame configuration at atmospheric pressure was studied.

The visible chemiluminescence of the excited radicals CH^* and C_2^* is employed experimentally.

Numerically, a 1D simulation was carried out using a standard kinetic mechanism, to which we add CH^* and C_2^* reactions.

Typical natural gas flame structure was presented.

A comparison between experimental and numerical

profiles of CH^* and C_2^* radicals for the natural gas/air was done for two operating conditions, validating the model chosen in this study, and the reactions of these excited radicals added to it.

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