



Full Length Article

Experimental and numerical investigation of low-pressure laminar premixed synthetic natural gas flames in rich conditions



B. Lefort ^{a,*}, A. El Bakali ^b, L. Gasnot ^b, J.F. Pauwels ^b

^a DRIVE EA1859, Univ. Bourgogne Franche-Comté, F58000 Nevers, France

^b PhysicoChimie des Processus de Combustion et de l'Atmosphère (PC2A), UMR CNRS 8522, Université des Sciences et Technologies de Lille, 59655 Villeneuve d'Ascq Cedex, France

HIGHLIGHTS

- Low-pressure laminar premixed flames of natural gas were realized in rich conditions.
- A detailed mechanism was developed and validated against different experimental conditions.
- One focus of the study is to explain the formation of single ring in our conditions.

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ABSTRACT

The present work concerns the study of rich laminar premixed CH₄/C₂H₆/C₃H₈/O₂/N₂ flames operating at two equivalence ratios ($\phi = 1.66$ and 2.05) and low pressure (80 Torr). In the case of the flame $\phi = 2.05$, the influence of pressure was studied between 80 and 300 Torr. Mole fraction of reactants, stable intermediate and combustion products have been obtained using a quartz microprobe coupled to gas chromatography, mass spectrometry and infrared spectroscopy technique analysis. Temperature profiles measured with a coated thermocouple were corrected from radiative heat losses by electrical compensation method. A detailed chemical kinetic mechanism has been developed. Globally the main experimental observations are well captured by the present mechanism, especially the effect of the equivalence ratio and pressure on the chemical species distribution. The proposed mechanism has been also tested to predict detailed and global data in other experimental conditions (ignition delay times, burning velocities, and jet stirred reactor). Reactions paths analyses have been carried out to identify the main reactions routes governing natural gas combustion in rich conditions. Both effect of pressure and initial composition on C₂ (ethylene, acetylene), C₃ (allene, propyne) and C₄ (butadiene, butyne) hydrocarbon species known as first ring precursors, have been examined. It was observed that self propargyl radicals is the main reaction route determining benzene formation in natural gas combustion in rich conditions.

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1. Introduction

The combustion chambers are the main sources of greenhouse gases and harmful pollutants for health. In the context of the fight against the climatic warming, several ways of “clean combustion” are implemented right now or at the study. For example, the track being based on the use of not-carbonaceous fuels such as hydrogen (H₂) is the object of various research projects. However, the direct and massive use of H₂ to produce energy still raises many difficulties, related to the high cost of its ecological production, storage on a large scale and transport. The use of H₂ in the presence of natural gas can constitute a transition solution which will allow a

progressive decarbonizing of the combustion chambers. Moreover, natural gas will continue to play a major role to meet the future growth of global energy demand in the context of the transition to sustainable energy systems. Basics, such as Environment, Technique, or Economy, converge towards a sustained growth of natural gas consumption in the future. The recent growth in gas consumption is linked in large part to its development in the electricity sector. Fukushima disaster that has shaken the nuclear industry has strengthened the development of natural gas potential that appears as an alternative choice to meet electricity needs.

Methane is the main component of natural gas (70–95% depending on the source). In natural gas, methane is associated with other hydrocarbons, mainly ethane, propane and butane. Therefore, as has been shown in many studies [1–4] natural gas combustion cannot be fully assigned to methane oxidation.

* Corresponding author.

E-mail address: benoite.lefort@u-bourgogne.fr (B. Lefort).

Table 1
Initial conditions of the flames studied at low pressure.

	$\phi = 1.66$		$\phi = 2.05$		
	Flow (L/h)	%	Flow (L/h)	%	
C ₁ C ₂ C ₃	81	27	100	33	
C ₁	72.1		89.0		
C ₂	7.3		9.0		
C ₃	1.6		2.0		
O ₂	107	36	107	36	
N ₂	112	37	93	31	
Total flow	300	–	300	–	
Pressure (Torr)	80		80	150	300

Nevertheless, it is needed to simplify natural gas mixture, so some fundamental studies over a large range of experimental conditions shown that methane, ethane and propane blend is the best mixture for capturing the natural gas combustion chemistry. Natural gas oxidation was the subject of numerous studies in our laboratory but all have been limited to stoichiometric and lean conditions. These investigations have been showed that 89% methane/9% ethane/2% propane is a good representative mixture of natural gas oxidation [3–6]. Recently we also studied the effect of hydrogen additive to natural gas in laminar premixed low and atmospheric pressure flames [7,8] at lean and stoichiometric conditions. To our knowledge, no investigation concerning laminar premixed natural gas flames in sooting conditions at low pressure could be found in the literature. However, some investigations on the combustion of alkanes as constituents of natural gas have been studied in rich fuel premixed laminar flames at atmospheric and low pressures [9–16]. These data bases allowed identifying a significant effect of the pressure on the first aromatic rings. Indeed benzene and toluene formation at atmospheric pressure [9,12–14] was observed whereas both species could not be detected at low pressure under similar equivalence ratio [10,11,14]. The objective of this work is to pursue our study on the combustion of the natural gas by completing the experimental data base previously obtained in stoichiometric and lean premixed laminar natural gas flames. For that purpose, we studied methane–ethane–propane–O₂–N₂ flames representative of natural gas combustion in conditions which allow examining the effect of the equivalence ratio and pressure. Two equivalence ratios are chosen $\phi = 1.66$ and 2.05. The second equivalence ratio $\phi = 2.05$ corresponds to the conditions producing soot particles. The effect of the pressure was examined in the case of the sooting flame in 80–300 Torr. These experimental results have been used to develop and validate a new detailed reaction kinetic mechanism for natural gas combustion.

2. Experimental set up

The chemical structure of laminar premixed CH₄/C₂H₆/C₃H₈/O₂/N₂ flames has been investigated. The oxidation of the synthetic natural gas methane–ethane–propane (NG) was found to be representative of natural gas combustion. The initial conditions of all flames studied in the present work are shown in Table 1. The flow rates of the natural gas blend ($2.00 \pm 0.04\%$ C₃H₈ and $9.00 \pm 0.18\%$ C₂H₆ in methane), nitrogen and oxygen (99.996% pure) were measured and regulated by thermal mass-flow controllers (Tylan FC260). The flames have been stabilized on a water-cooled plug flat flame burner (6 cm in diameter) at low pressure. Species analyses were achieved by quartz probe sampling along the symmetry axis of the flame. Mole fraction profiles of species have been described by moving the burner vertically. Low-pressure samples of the reacting mixtures were taken by probe sampling and collected in an enclosure at 0.03 atm for gas chromatography (GC) analyses (Agilent 6890N). To improve the GC detection, these samples were

pressurized up to 0.7 atm before injection into the GC column, using a homemade piston. Hydrocarbon species were analyzed with an HP-Plot Al₂O₃ capillary column and a flame ionization detector (FID). A molecular sieve 5A column allowed separation of hydrogen, oxygen, nitrogen, and carbon monoxide, and these species are detected with a thermal conductivity detector (TCD). Finally, carbon dioxide is analyzed with a Poraplot Q column and TCD detector. The internal diameter of each column is 0.53 mm. Helium was used as a carrier gas. The species are directly calibrated before each measure range. Known concentration standards (MicroMAT, $\pm 2\%$ accuracy) allow the determination of sensitivity coefficients for each species. Fourier transform infrared spectroscopy (FTIR) analyses of H₂O, CO, and CO₂ were also performed. The spectrometer used is a FTIR (NEXUS THERMO-OPTEK). Gas effluents are taken within the flame by the same microprobe as for GC analyses. A rotary vane pump (flow 25 m³/h) allows the sampling (0.05 atm) and conveys it to a 2-l heated gas cell (100 °C) combined with FTIR. The chemical species present in the gas sample are analyzed at the time of their passage in the cell according to an optical path of 10 m. Fifteen scans are realized for a 1 cm⁻¹ resolution. The FTIR was piloted by OMNIC/QUANT-PAD software. A spectral line instead of a spectral zone is chosen to analyze for each species in order to improve sensitivity. The spectral zone for CO₂ is 2394–2276 cm⁻¹, the one for CO is 2142–2092 cm⁻¹, and the spectral zone for H₂O is 1719–1698 cm⁻¹. A background is realized before each analysis. The calibration is done by a homemade standard of different known composition of H₂O, CO, and CO₂. Temperature profiles are obtained using a coated Pt/Rh 6%–Pt/Rh 30% thermocouple with a 100 μ m of wire diameter. Conduction heat losses are avoided by setting the thermocouple in a plane perpendicular to the laminar flow. Radiative heat losses are corrected using the electric compensation method [17]. Errors in the peak temperatures were estimated to be ± 100 K. Uncertainties are around 10% for major species (CO, CO₂, CH₄, C₂H₆), around 10–15% for minor species, and about 20% for H₂O.

3. Experimental results

3.1. Temperature

The experimental temperature profiles measured by a coated Pt/Rh 6%–Pt/Rh 30% thermocouple are shown in Fig. 1. The temperature profiles are corrected from radiative heat losses by electrical

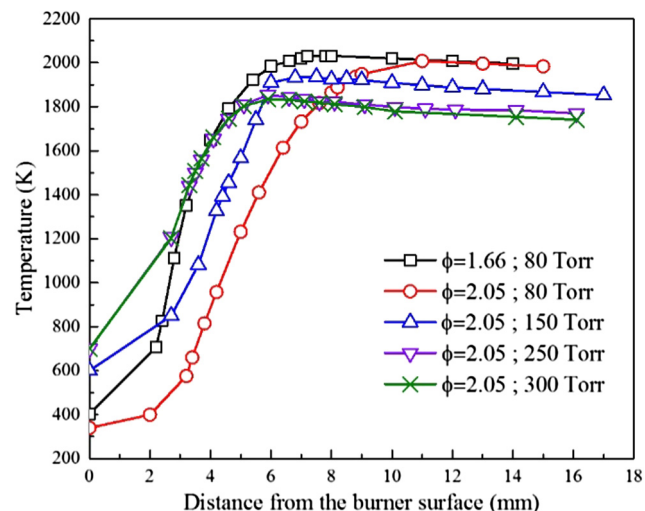


Fig. 1. The experimental temperature profiles measured in laminar premixed CH₄/C₂H₆/C₃H₈ flames at two equivalence ratios and different pressures.

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