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An experimental study of the structure of laminar premixed flames of ethanol/methane/oxygen/argon

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Abstract

The structures of three laminar premixed stoichiometric flames at low pressure (6.7 kPa): a pure methane flame, a pure ethanol flame and a methane flame doped by 30% of ethanol, have been investigated and compared. The results consist of concentration profiles of methane, ethanol, O₂, Ar, CO, CO₂, H₂O, H₂, C₂H₆, C₂H₄, C₂H₂, C₃H₈, C₃H₆, *p*-C₃H₄, *a*-C₃H₄, CH₂O, CH₃HCO, measured as a function of the height above the burner by probe sampling followed by on-line gas chromatography analyses. Flame temperature profiles have been also obtained using a PtRh (6%)-PtRh (30%) type B thermocouple. The similarities and differences between the three flames were analyzed. The results show that, in these three flames, the concentration of the C₂ intermediates is much larger than that of the C₃ species. In general, mole fraction of all intermediate species in the pure ethanol flame is the largest, followed by the doped flame, and finally the pure methane flame.

Keywords

Premixed laminar flame; Methane; Ethanol

1. INTRODUCTION

In the recent years, the announced decrease of petroleum reserves and the tightened regulations for emissions of environmental concern from internal combustion engines have stimulated research to find alternative fuels.

Ethanol is regarded as an attractive renewable alternative fuel with a high octane number [1]. This oxygenated fuel can be obtained through the fermentation of sugars or starches which can be produced from very common crops, such as sugar cane or corn [2]. Recently ways to produce ethanol from cellulose [3] or algae [4] have also been proposed. Therefore the use of ethanol allows a reduction of the dependence on fossil fuels. Moreover burning this renewable fuel should not lead to an increase of the total amount of greenhouse gases in the atmosphere. In addition this oxygenated fuel can decrease the formation of soot, CO and unburned fuel or partly oxidized hydrocarbons in practical applications, but tendencies to form aldehydes have been observed depending on fuel structure and combustion conditions [2], [5]. Ethanol is now one of the most common and abundant bio-fuels [6], [7], [8]. Figure 1 shows the worldwide fuel-ethanol production in years 2007, 2008 and 2009 [6]. A noticeable increase of this production can be observed.

Several previous studies have already been conducted for better understanding each step of ethanol combustion and for developing an accurate oxidation mechanism. Ethanol

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combustion has been studied in diffusion flames [9], laminar premixed flame [10], [11], static reactors [12], shock tubes [13], [14]. The first kinetic mechanisms proposed, e.g. by Natarajan and Bhaskaran (in 1981) [15] and by Dunphy et al. (in 1991) [16], were based on experimental data obtained in shock tubes. Norton and Dryer (in 1992) [17] published an oxidation mechanism which has been validated using experimental data obtained in a flow reactor. Later, Marinov et al. (in 1999) [18], Saxena and Williams (in 2007) [19], Li et al. (in 2007) [20], Cancino et al. (in 2010) [21], and Leplat et al. (in 2011) [10] have published some detailed kinetic models for the combustion of ethanol; these mechanisms have been validated by experiments in several devices, including laminar premixed flame [10].

Continuous developments and improvements of kinetic mechanisms require new experimental data. The validation of kinetic mechanisms over a large range of conditions is very important. Currently, ethanol/methane mixture flame structure data are not available. In addition, only very few data are available for C_3 intermediates formations in ethanol flame. In order to fill this gap and to further improve the understanding of ethanol combustion, the objective of this study is to experimentally investigate the structure of a laminar premixed stoichiometric methane flame containing 30% of ethanol at low pressure. A comparison of this flame with a flame of pure ethanol and a flame of pure methane is also presented. The use of a methane flame allows us to have a reactive mixture rich in methyl radicals and representative of combustion mixtures containing larger hydrocarbons.

2. EXPERIMENTAL METHODOLOGY

The experimental setup used to determine the structure of one-dimensional laminar premixed flames has been described previously [22], [23]. This apparatus has been developed in our laboratory to study temperature and stable species profiles in a laminar premixed flat flame at low pressure and has been used recently in the case of rich methane flames doped by light unsaturated soot precursors [24], [25], [26].

All flames were stabilized on the Mac Kenna burner (diameter 60 mm, water-cooled) housed in a vacuum chamber which is maintained at 50 Torr (6.7 kPa) and equipped with a quartz probe with a hole of about 180 μm diameter at the tip for sampling (see Figure 2 for parameters of the end of this sampling probe). The cooling water has a constant temperature of 60°C.

Methane (99.95 % pure) was supplied by Alphagaz - Air Liquide. Oxygen (99.5% pure) and argon (99.995% pure) were provided by Messer. Liquid ethanol was supplied by Sigma-Aldrich (purity > 99.5%). Liquid ethanol was contained in a metallic vessel pressurized with argon. After each load of the vessel, argon bubbling and vacuum pumping were performed in order to remove oxygen traces dissolved in the ethanol. The ethanol flow rate was controlled by using a liquid mass flow controller, mixed with the argon and then evaporated by passing it through a CEM (Controlled Evaporator and Mixer). The temperature of this CEM was set at 100°C.

Analysis was made by gas chromatography (GC) with a heated on-line connection to the probe and using three types of columns (Carbosphere, HP-Plot U and HP-Molsieve) and two types of detectors (flame ionization (FID) coupled with methanisor and thermal conductivity (TCD)). Stable species were also identified by mass spectroscopy (GC/MS). Calculated uncertainties of the measurement of mole fraction on the species quantifications were about $\pm 5\%$ for the major compounds, and $\pm 10\%$ for minor products (<100 ppm). The FID detection threshold was about 1 ppm, while the TCD detection limit was 50 about ppm for H_2 and O_2 .

Flame temperature profiles were obtained using a PtRh (6%)-PtRh (30%) type B thermocouple (diameter 100 μm and 105 μm without and with anticatalytic protective layer respectively). The thermocouple wire was supported by an arm and crossed the flame horizontally to avoid conduction heat losses. The junction was located at the center of the burner. The thermocouple was coated with an inert layer of $\text{BeO}-\text{Y}_2\text{O}_3$ to prevent catalytic effects when it is placed in flame [27]. The ceramic layer was obtained by dipping the thermocouple in a hot solution of $\text{Y}_2(\text{CO}_3)_3$ (93% mass) and BeO (7% mass) followed by drying in a Meker burner flame. This process was repeated about 10 times until the whole metal was covered. Radiative heat losses are corrected using the electrical compensation method [28]. Uncertainty of the measurement of temperature was about ± 20 K in burned gas. In fresh gas, there is an uncertainty ± 0.05 mm of the position of the temperature profile in the direction perpendicular to the burner surface. A sighting telescope (cathetometer) measured the position of the burner relative to the probe or the thermocouple with an accuracy of 0.01 mm.

Initial operating conditions of the three flames used in the current study are presented in Table 1. The cold gas velocity (at 20°C) is 57 cm/s for the three flames.

3. RESULTS AND DISCUSSIONS

The flame temperature profiles and mole fraction profiles of reactants, major and intermediate species are presented.

3.1. Flame temperature profiles

The experimental temperature profiles constitute an essential parameter for the interpretation of laminar flame data. Figure 3 represents the temperature profiles measured for the three flames without (Figure 3a) and with (Figure 3b) the probe. These figures show that the presence of the probe induces a thermal perturbation causing a lower measured temperature. The maximum temperature in the post flame region is almost similar for the three flames.

3.2. Mole fraction profiles of reactants, major and intermediate species

Figures 4-8 present mole fraction profiles of several stable species as a function of the height above the burner for the three flames. It can be stated that concentrations of most identified intermediate products in the three flames have their maxima at the distance of about 1-3 mm from the burner surface. A displacement of the position of the maximum concentrations is observed for CO , H_2 , C_2H_4 , and C_2H_2 . The major final products are, to a large extent, CO_2 and H_2O .

Figure 4 presents the profiles of reactants ($\text{C}_2\text{H}_5\text{OH}$, CH_4 , Ar , O_2) and shows that ethanol is completely consumed close to the burner, at 2.5 mm height (Figure 4a), while some methane remains up to 3.5 mm (Figure 4b). The reason for this difference is that the laminar flame speed of methane/oxygen mixture is slower than that of ethanol/oxygen mixture [29], implying that the pure methane flame front is further from the burner.

It can be seen from Figure 4c that there is almost no difference in O_2 mole fraction in the three flames, which reveals that the consumption rate of O_2 does not vary depending on the flames in this experiment and there is a significant remaining mole fraction of O_2 in the post flame region.

There is also almost no difference in argon mole fraction for the three flames (Figure 4d).

Mole fraction profiles of major species, including CO_2 , H_2O , H_2 and CO , are shown in Figure 5.

The mole fraction profile of CO_2 is shown in Figure 5a. CO_2 is the main green house effect gas and will be controlled by future emission legislated regulations. The mole fraction of CO_2 formed in pure ethanol flame is the largest and in pure methane flame is the lowest. The increase of CO_2 mole fraction in the pure ethanol flame is mainly due to the increase of C/H ratio (see table 1). There is only slight difference in H_2O mole fraction for the three flames (Figure 5b).

In the pure ethanol flame, the profiles of H_2 display a marked maximum at 2.5 mm height, which is at 3.0 mm in the doped flame and in the pure methane flame. It can be seen from Figure 5c that there is a remaining mole fraction of hydrogen in the post flame region.

In the doped flame and in the pure methane flame, the profiles of CO (Figure 5d) display a marked maximum at 3.5 mm height, which is at 3.0 mm in the pure ethanol flame. The maximum mole fraction of CO formed in the pure ethanol flame is the largest and in the pure methane flame is the lowest. The increase of CO mole fraction in the pure ethanol flame is mainly due to the increase of C/H ratio. The CO formation occurs as an essential intermediate step in the process of oxidation of fuels, leading to the final product CO_2 by the reaction: $\text{CO} + \text{OH} \rightarrow \text{CO}_2 + \text{H}$. It can be seen from Figure 5d that there is a remaining mole fraction of CO in the post flame region.

Mole fraction profiles of intermediates CH_4 , C_2H_6 , C_2H_4 , and C_2H_2 are shown in Figure 6.

In the pure ethanol flame, CH_4 is an abundant intermediate, formed from the CH_3 radicals and consumed to give this same radical. The profile of CH_4 (Figure 6a) have a peak near 2 mm height (peak mole fraction of 2700 ppm).

For C_2 (non-oxygenated) species, the most important is C_2H_6 in the pure methane flame (peak mole fraction of 1000 ppm), and C_2H_4 in the pure ethanol flame (peak mole fraction of 5000 ppm) and in the doped flame (peak mole fraction of 2000 ppm). C_2H_6 is formed first in the pure methane flame (peaks around 1.8 mm height) and in the doped flame (peaks around 1.8 mm height) (Figure 6b), while C_2H_4 is formed first in the pure ethanol flame (peaks around 2 mm height) (Figure 6c). This shows differences in the most important and first formation of C_2 species in the three flames. These differences can be explained as follows. High temperature chemical kinetic models show clearly that there are two well-known pathways for methane oxidation, i.e., C_1 and C_2 pathways; the C_1 pathway being the dominating one [30], [31]. In the C_2 pathway, two CH_3 radicals combine to produce C_2H_6 and C_2H_5 . C_2H_6 forms C_2H_5 by reacting with H and OH radicals, and C_2H_5 finally leads to the formation of C_2H_2 through intermediate species C_2H_4 and C_2H_3 . Therefore, in the pure methane flame, C_2H_6 is the most abundant C_2 species and is produced first. While in ethanol flame, C_2H_4 would be formed essentially from ethanol by the following decomposition reaction: $\text{C}_2\text{H}_5\text{OH} (+\text{M}) \rightarrow \text{C}_2\text{H}_4 + \text{H}_2\text{O} (+\text{M})$ or via the $\text{CH}_2\text{CH}_2\text{OH}$ (hydroxylethyl) radical: $\text{CH}_2\text{CH}_2\text{OH} (+\text{M}) \rightarrow \text{C}_2\text{H}_4 + \text{OH} (+\text{M})$ [10], [18]. $\text{CH}_2\text{CH}_2\text{OH}$ radicals are obtained from ethanol by H-abstractions. Therefore, in the pure ethanol flame, C_2H_4 is the most abundant C_2 (non-oxygenated) species and is produced closer to the burner.

The maximum concentration of C_2H_2 (Figure 6d) and C_2H_4 (Figure 6c) which are significantly increased in the doped flame compared to the pure methane flame, even reaches a higher level in the pure ethanol flame. The highest concentration of C_2H_6 (Figure 6b) is almost similar in both pure ethanol and pure methane flames, and is slightly reinforced in the doped flame. In addition, the similarity in the C_2H_6 concentration in the pure ethanol and pure methane flames can be explained as follows. C_2H_6 is produced by combination of two CH_3 radicals. The C/H ratio of the pure ethanol flame is the highest leading to large mole fraction of C_2H_6 . However, CH_3 radical is the most important intermediate radical in methane combustion.

The maximum concentration of C_2H_4 varies as the C/H, C/O ratios of the fresh initial gases. Indeed, in Figure 6c, we observe that the concentration of C_2H_4 in the pure ethanol flame is the highest (5000 ppm), followed by the doped flame (2000 ppm), and finally the pure methane flame (500 ppm).

C_2H_2 is considered as the most representative soot precursor in a variety of hydrocarbon fuel flames because it yields benzene, which is the first step toward the production of soot [32]. The maximum concentration of C_2H_2 varies also as the C/H, C/O ratios of the fresh initial gases. Figure 6d shows that the concentration of C_2H_2 in the pure ethanol flame is the highest (1300 ppm), followed by the doped flame (550 ppm), and finally the pure methane flame (150 ppm).

Figure 7 presents the profiles of the C_3 species including C_3H_8 (Figure 7a), C_3H_6 (Figure 7b), p - C_3H_4 (Figure 7c), and a - C_3H_4 (Figure 7d). These figures show that a very small amount of C_3 products is also observed in the pure methane flame.

In the three flames, C_3H_6 and C_3H_8 reach the highest concentration first around 1.8 mm above the burner, while the maxima of p - C_3H_4 and a - C_3H_4 are around 2.5 mm. A very small amount of these products is also observed in the pure methane flame and the concentration of p - C_3H_4 and a - C_3H_4 is lower than the limit of detection of the gas chromatograph. The peak mole fraction of C_3H_6 , which is the most important C_3 species in the pure ethanol flame, is 105 ppm, while that in the pure methane flame and doped flame is 26 ppm and 54 ppm, respectively.

The concentration of C_3 species in the pure ethanol flame is the largest, followed by the doped flame, and finally the pure methane flame, i.e. the maximum concentration of C_3 product varies also as the C/H, C/O ratios of the fresh initial gases.

Figure 8 displays the profiles of light oxygenated species: formaldehyde (CH_2O) and acetaldehyde (CH_3CHO).

The aldehydes, which are hazardous air pollutants, are produced promptly in the flames and reach their maximum concentration close to the burner, around 1.5 mm height for CH_2O (Figure 8a) and around 1.0 mm height for CH_3CHO (Figure 8b).

The concentration of aldehydes in the pure ethanol flame is the largest, followed by the doped flame, and finally the pure methane flame. CH_3CHO is not detected in the pure methane flame.

In ethanol flame, the two radicals (CH_3CHOH , CH_3CH_2O) initially formed by H-abstraction from the ethanol generate CH_3HCO via following reactions [15], [18], [33]:

The production of CH_2O can also be rationalized from this scheme through the C-C scission of the CH_3CH_2O radical ($CH_3CH_2O + M \rightarrow CH_3 + CH_2O + M$) [18]. These

reaction paths show how large aldehydes emissions may be expected from ethanol combustion. The results of this study confirm that acetaldehyde formation during ethanol combustion is important. Acetaldehyde is the main intermediate species formed in the flame front (the peak mole fraction is 7500 ppm).

In the methane flame, formaldehyde is essentially formed from CH_3 and C_2H_3 radicals [31] by the following reactions:

CONCLUSION

This paper presents new experimental results about the structure of three low-pressure stoichiometric laminar premixed flames: a pure flame methane, a pure ethanol flame, and a methane flame doped by 30% of ethanol. Profiles of temperature and mole fraction of 17 stable species from C_0 to C_3 have been obtained. Among these 17 species, there are 2 aldehydes. A comparison between the profiles of temperature and of mole fraction is also presented. This comparison shows that, in general, mole fraction of all species (except reactants and C_2H_6) in the pure ethanol flame is the largest, followed by the doped flame, and finally the pure methane flame. The reaction paths consuming the organic reactant (depending on the nature of fuel), the H/C and C/O ratios of the fresh initial gases have allowed us to identify the origin of the differences among these flames. Despite its oxygen content, ethanol produced much more unsaturated hydrocarbons, which are soot precursors, and toxic aldehydes than methane despite the close C/O ratio of the flame studied. The structure of the molecule where two carbon atoms are linked favors indeed the formation of C_2 and C_3 compounds in comparison with methane.

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Figure 1.
World fuel ethanol production by country (adapted from [6])

Figure 2.
Scheme of the end of sampling probe (*Length unit: mm*).

Figure 3.
Temperature profiles: without (a) and with (b) the sampling probe.

Figure 4.
Mole fractions profiles of reactants C_2H_5OH , CH_4 , O_2 , Ar

Figure 5.
Mole fraction profiles of major species CO_2 , H_2O , H_2 , CO

Figure 6.
Mole fractions profiles of CH_4 (intermediate) and C_2 species

Figure 7.
Mole fractions profiles of C_3 species

Figure 8.
Mole fraction profiles of oxygenate intermediates

Table 1
Flame inlet conditions.

Flame Name	a	Mole fraction				Gas velocity at 20°C (cm/s)	Dilution (%)	C/O ^a	C/H ^a
		Ethanol	CH ₄	O ₂	Ar				
Pure ethanol flame	1.0	0.06840	0	0.20437	0.72722	57	78	0.287	0.333
Doped flame ^b	1.0	0.02072	0.06907	0.20076	0.70945	57	78	0.262	0.276
Pure methane flame	1.0	0	0.09910	0.19888	0.70202	57	78	0.249	0.250

^a -equivalence ratio; C/H-Carbon/Hydrogen ratio ; C/O- Carbon/Oxygen ratio

^b =ethanol/methane mixture flame.