



# The influence of ethanol addition on a rich premixed benzene flame at low pressure



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## ABSTRACT

The aim of the study is to analyze the effect of ethanol in rich benzene flame, to observe the influence of this oxygenated species and to understand the kinetics of ethanol in the benzene combustion. Two premixed rich benzene/oxygen/argon (11.5% C<sub>6</sub>H<sub>6</sub>, 43.2% O<sub>2</sub>, 45.3% Ar) and benzene/ethanol/oxygen/argon (10.7% C<sub>6</sub>H<sub>6</sub>, 2.1% C<sub>2</sub>H<sub>5</sub>OH, 43.2% O<sub>2</sub>, 44.0% Ar) flat flames are stabilized at low pressure (45 mbar) on a burner with the same equivalence ratio of 2.0. Identification and monitoring of signal intensity profiles of species within the flames are carried out using molecular beam mass spectrometry (M.B.M.S.). The substitution of some C<sub>6</sub>H<sub>6</sub> by C<sub>2</sub>H<sub>5</sub>OH is responsible for a reduction of the maximum concentrations of main intermediate species such as C<sub>2</sub>H<sub>2</sub>, C<sub>4</sub>H<sub>2</sub>, C<sub>4</sub>H<sub>4</sub> and C<sub>5</sub>H<sub>6</sub>. The UCL mechanism is extended to heavier hydrocarbons, tested against these flames to check its validity and used to underline the effect of ethanol on soot precursors formation. It contains 1028 elementary reactions and 184 chemical species.

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## 1. Introduction

To reduce harmful emissions from internal combustion engines, some oxygenated species and fuel additives, such as dimethyl ether (DME) and ethanol (C<sub>2</sub>H<sub>5</sub>OH), have been investigated these last decades.

Ethanol is a very important species because it is widely used as neat fuel, as well as blended with gasoline; it can be used as an inhibitor of soot precursors formation. These last years, ethanol has been added in premixed flames of ethylene [1–3], propene [4–6], *n*-heptane [7,8], and gasoline [9,10] to underline its effect on the combustion of these hydrocarbons.

Concerning the addition of ethanol in rich ethylene flames, Wu et al. [1] studied at atmospheric pressure the influence of ethanol in rich ethylene flames, on the reduction in polycyclic aromatic hydrocarbons (PAHs) and soot formation. They concluded that the effect of ethanol addition is greater at equivalence ratio  $\phi = 2.34$  than at  $\phi = 2.64$ , due to the lower amount of carbon available to form precursor species. In 2011, Korobeinichev et al. [2] analyzed the effect of ethanol in a low pressure ethylene/oxygen/argon flame, keeping the same equivalence ratio of 2.0. They observed a decrease of propargyl radicals formation in the presence of ethanol, which leads to a reduction of benzene formation. This effect is due to the replacement of ethylene by ethanol in the initial mixture; the

ethanol pathways do not lead to soot precursors formation. The same group, then, studied the addition of ethanol in a rich premixed ethylene flame at atmospheric pressure, keeping the equivalence ratio constant at 1.7 [3]. They concluded that at low or atmospheric pressure, the presence of ethanol in rich ethylene flame has the same effect: a decrease of the formation of small hydrocarbons (C<sub>2</sub>H<sub>2</sub>, C<sub>3</sub>H<sub>4</sub>, C<sub>3</sub>H<sub>3</sub>) but an increase of the production of oxygenated species (formaldehyde and acetaldehyde). These results are due to the kinetics of ethanol which is different from the ethylene one.

These previous works underline that the presence of ethanol reduces the formation of soot precursors, but increases the formation of oxygenated compounds. The same effect was observed in rich propene flames, according to the work of Kohse-Höinghaus et al. [4]. The ethanol was added to a rich flame of propene at low pressure, keeping the Carbon/Oxygen (C/O) ratio equals to 0.77. Experimentally, they observed a reduction of benzene and small aromatic compounds formation but an increase of other regulated air toxics such as aldehydes. Then, they compared different amounts of dimethyl ether (DME) or ethanol, added to a premixed rich propene flame at low pressure keeping the carbon/oxygen ratio at 0.5. This experimental work is presented in [5] and the modeling one in [6]. They concluded that C<sub>3</sub>H<sub>x</sub> and C<sub>6</sub>H<sub>6</sub> production are reduced with the presence of oxygenated additives, whatever their nature. Nevertheless, the presence of DME increases the concentration of formaldehyde (CH<sub>2</sub>O); and ethanol increases the acetaldehyde one (CH<sub>3</sub>CHO). Both these oxygenated intermediates are considered as pollutants.

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The effect of ethanol was also tested in premixed *n*-heptane flames. Inal and Senkan [7] tested three oxygenated fuels (MTBE, methanol and ethanol) in a premixed *n*-heptane flame at atmospheric pressure, by keeping the equivalence ratio constant at 1.97. The experiments showed that the increase of oxygen weight percent in *n*-heptane/oxygenated mixtures reduced mainly the concentration of aromatics and PAH, of CO and small hydrocarbons. Song et al. [8] studied the stoichiometric *n*-heptane flame at low pressure with MTBE and ethanol as additives. They concluded that the presence of ethanol increased the concentration of acetaldehyde, vinyl alcohol and butanol; and the presence of MTBE, the concentration of ketones, ethane methoxy and propane 2-methoxy. Moreover, the presence of these oxygenated additives enhanced the *n*-heptane oxidation and reduced the aromatics formation. The *n*-heptane is considered as one of the primary reference for gasoline octane number and these results improved the understanding of its kinetics.

The last studied flames with ethanol as additive are with gasoline fuel. Yao et al. [9] underlined that the presence of ethanol at 10% by volume lead to an increase concentration of acetaldehyde and retarded the overall fuel oxidation. The MTBE was also tested as additive but no significant effect on the formation of acetaldehyde was observed. In 2012, Maricq [10] examined the effect of ethanol addition in gasoline on soot formation. He studied the different modes of the soot formation (agglomeration, nucleation, coagulation) and the influence of the ethanol presence on the size distribution of these particles. But from the experimental results, he concluded that the efficiency of ethanol on soot particles formation was very low for the small quantities of ethanol added. He suggested that other factors such as fuel volatilization and mixing could be more important than combustion chemistry to affect particles matter emissions of motor vehicle.

In addition to the oxygenated species studies, one of the main goals in the combustion field is the knowledge of the formation of polycyclic aromatic hydrocarbons (PAHs) and soot. These heavy and stable compounds are produced during the incomplete hydrocarbons combustion, as in engine or industrial processes. They should be reduced due to their carcinogenic effect. The main precursor of these PAHs is the first aromatic ring, the benzene (C<sub>6</sub>H<sub>6</sub>). Several studies on rich premixed benzene flames tried to model its kinetics to predict the formation of heavier hydrocarbons and soot precursors [11–16]. Our group has recently performed some experiments of a rich flat premixed benzene flame with acetylene addition to underline the role of this additive on the formation of heavier hydrocarbons and PAHs [17,18]. Following this work on the benzene combustion, one perspective was to study this rich benzene flame in the presence of an oxygenated species.

In 2012, Golea et al. [19] presented a study on the reduction of PAHs and soot precursors in benzene flames by the addition of ethanol. This numerical work studies the decrease of the soot precursors concentration in benzene flame [18] with different amounts of C<sub>2</sub>H<sub>5</sub>OH added. They concluded that the PAH reduction was simply the result of a replacing sooting benzene with nonsooting ethanol without influencing the chemistry of the benzene.

The aim of this work is, thus, to analyze experimentally and numerically the effect of ethanol in a rich premixed benzene flame, to observe the influence of this oxygenated species and to understand the kinetics of ethanol in benzene combustion.

## 2. Experimental

Two premixed flat benzene-oxygen-argon flames (with and without ethanol added), have been stabilized on a Spalding-Botha burner. The setup consists of a combustion chamber where a flat flame is stabilized at low pressure (45 mbar) on a movable burner of 8 cm in diameter. Facing the burner surface, a conical quartz

probe with a small hole of 0.1 mm at the apex allows sampling the flame. Behind this probe, three differentially pumped chambers lead to the formation of a molecular beam that is directed towards the electronic ionization source of a quadrupole mass spectrometer (Balzers QMG 511). The axis of the mass spectrometer is perpendicular to the molecular beam one. Such an arrangement allows the initial composition of the sampling to be “frozen” at the point where the mass spectrometer is reached and therefore reactive species can be detected and monitored. Moreover, the molecular beam is chopped at 30 Hz, allowing a phase detection of the signal coming from the electron multiplier which strongly enhances the signal-to-noise ratio. The experimental setup has been described in detail elsewhere [20].

For the investigated flames, the initial flow velocity is 58.3 cm/s for the neat benzene flame (FB), and 58.8 cm/s for the benzene/ethanol flame (FBE), at 298 K, and the total mass flux is 5.5 l/min. Initial molecular oxygen concentrations were kept constant and mole fractions of fuels (benzene and ethanol) were adjusted to keep the equivalence ratio equal to 2.00 (Table 1).

Identification and monitoring of signal intensity profiles of stable, atomic and radical species within the flames have been carried out using molecular beam mass spectrometry (M.B.M.S.). For every species, interferences from fragmentation during the electron impact or from overlapping of species with similar mass have been kept low or taken into account. The ionization technique used in this work is the electronic impact ionization (EI). To avoid fragmentation, applied ionization potentials are therefore always carefully adjusted just above ionization energies of targeted compounds (molecule or radical). The conversion of signal intensities to mole fractions has been performed by using a calibrated mixture for the stable compounds and by estimating ionization cross sections for the unstable species. The estimation of the ionization cross sections of species has been made by adding the individual ionization cross sections of the constituting atoms:  $Q_C = 1.8$ ,  $Q_O = 1.3$ ,  $Q_H = 0.65$  [21]. For stable species such as CO, CO<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>, Ar, the ratio  $S_i/S_{Ar} = I_i \cdot X_{Ar}/I_{Ar} \cdot X_i$  (with  $S_i$  the sensitivity coefficient of species  $i$ ,  $I_i$  the intensity of species  $i$  and  $X_i$  the mole fraction of species  $i$ ) can be determined using the sensitivity ratios derived from the calibration gas mixture. In the post flame zone, the H<sub>2</sub>/O<sub>2</sub> system is in partial equilibrium. Three equilibrium equations can be written to determine the concentrations of H<sub>2</sub>O, H, O and OH, if the concentrations of CO, CO<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>, Ar and the temperature are known. In this work, the experimental mole fraction profile of H<sub>2</sub> is not measured but the calibration is performed by using parameters from a previous work [17]. The last equations needed to determine the composition of burned gases are derived from the sum of mole fractions equal to 1.0 and the equation of conservation of the total number of atoms. More details are available in a previous article [20]. The quantification by the mass spectrometry technique allows to obtain a calibration without excessive experimental errors. Each chemical species profile was successively measured in similar conditions in every investigated flames, in order to perform reliable calibration and direct comparison. The standard deviation for measures close to the maxima of a profile is smaller than 10% of the total signal for detected species down to 1000 ppm; and of the order of 20% for species lower than 100 ppm. These estimations are based on various sources: the composition of the initial gas mixture (from the mass flow controllers), the measure of intensity for each species by the mass spectrometer

**Table 1**  
Flame inlet compositions.

Flame	X <sub>C<sub>6</sub>H<sub>6</sub></sub>	X <sub>C<sub>2</sub>H<sub>5</sub>OH</sub>	X <sub>O<sub>2</sub></sub>	X <sub>Ar</sub>	φ	C/O
FB	0.115	0	0.432	0.453	2.00	0.80
FBE	0.107	0.021	0.432	0.440	2.00	0.77

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