



Effect of furanic biofuels on particles formation in premixed ethylene–air flames: An experimental study



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HIGHLIGHTS

- Sooting tendency of F, MF and DMF is studied in atmospheric premixed flames.
- LIF and LII are the probing techniques for nanoparticles and soot, respectively.
- Furans are added up to 20% of the total carbon.
- Increasing additive percentage LII decreases while LIF is slightly affected.
- As equivalence ratio increases LII signal reduction increases.

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ABSTRACT

Furanic fuels have been recently investigated because they are considered possible fossil fuel substituents due to their high energy density, their renewable origin and consequently low green-house gas emissions. Few data are available about the behaviors of these fuels in laboratory flame reactors. We have recently studied their sooting tendency in a counter-flow diffusion flame showing a different propensity to form particulate matter depending on the richness of the combustion conditions. In this paper, we explore their tendency in forming particulate matter in atmospheric pressure premixed flames burning mixtures of ethylene and furanic fuels at four different equivalence ratios, namely 2.01, 2.16, 2.31 and 2.46, thus covering the range from slightly sooting to fully sooting conditions. Liquid furan, 2-methylfuran and 2,5-dimethylfuran have been added to ethylene as 10% and 20% of the total carbon fed to the flame.

In-situ spectroscopy, namely laser UV-induced emission, has been used as diagnostic tool to detected different classes of precursor nanoparticles by changing the detection wavelength from the UV to the visible. Laser induced incandescence has instead been used to detect soot particles. Equivalence ratios, the total carbon flow rate and cold gas velocity have been kept constant, allowing the combustion temperature to be almost the same also when furans were added.

LII signal decreases significantly when furans are added showing a strong effect of furanic fuels in decreasing the amount of soot formed in atmospheric pressure premixed flames. Particularly the reduction increases as the equivalence ratio increases. On the other side, LIF UV and LIF VIS slightly decrease when furans are added. The absence of a significant reduction of the LIF UV signal when furans are added suggests that small particle formation is not effectively inhibited by these additives. This behavior is similar to what found for ethanol and dimethyl ether addition to ethylene premixed flames, showing a similarity in the behavior of these oxygenated fuels.

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1. Introduction

Biofuel consumption has increased through years due to their intrinsic capability to reduce the net carbon dioxide release as well

as the possibility to reduce the dependence on fossil fuels. However, the opportunity of using biofuels coming from different sources, such as low valuable biomasses, is essential for their environment friendly usage. Also, biofuels need minor retrofitting to be fueled in the actual combustion devices and they have been found to generally reduce combustion-generated particle emissions.

Fuel-borne oxygen and the higher reactivity of soot particles generated during biofuel combustion have been found to be

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determinant for particulate emission reduction. Other studies have found that the changes in the overall combustion conditions as well as the presence of trace elements in biofuels could be responsible for particle reduction rather than the fuel structure [1–3]. Despite of large efforts, a definitive consensus on the effective role played by biofuels in the reduction of combustion-generated particle emissions is not reached. Before introducing a new biofuel on the market, systematic study of its impact on both total amount and reactivity of combustion-generated particles should be carefully explored.

Several oxygenated biofuels such as alcohols (methanol, ethanol, and butanol), ethers (methyl tertiary butyl ether – MTBE, ethyl tertiary butyl ether – ETBE) and biodiesels (methyl esters) are already available as potential additives for conventional gasoline and diesel. Studies on these potential biofuels have been carried out recently, generally showing for biofuels a decrease of the total amount of produced particulate matter due to different oxidation/pyrolysis pathways and to the changes of the overall fuel/air ratio [1–3]. However, their effectiveness in reducing total number concentration of combustion-generated particles is less evident, especially regarding the small nanoparticles. These latter are negligible in terms of mass but they dominate the number particle size distribution (PSD). Nanoparticle formation seems to be not inhibited or it is even enhanced in biofuel combustion [4–7].

Salamanca et al. [4,5,7] have showed that in premixed and diffusion flames soot particles were largely depleted by adding ethanol and DME to ethylene whereas nanoparticles were practically not affected. PSDs have been also measured by Camacho et al. [6] in rich premixed flames of n-butanol and i-butanol with equivalence ratios ranging from slightly sooting to sooting conditions. The number concentration of particles with sizes smaller than 10 nm was comparable with those found in n-butane and i-butane flames in similar conditions thus showing any effect on nanoparticle reduction when using biofuels.

In last years, furanic compounds have been selected as platform for biofuels, in particular, furan (F), 2-methylfuran (MF) and 2,5-dimethylfuran (DMF). These potential biofuels have been investigated with respect to their combustion chemistry, including informations on the nature and amount of undesired and possible harmful combustion by-products.

Thermal decomposition of furanic fuels has been investigated in a pioneering study by Cullis and Norris [8] within the temperature range of 1173–1323 K at 1 atm [8], tracking the main combustion by-products, such as methane (CH₄), acetylene (C₂H₂), ethylene (C₂H₄) and benzene (C₆H₆). Similar investigation has been conducted by Grela et al. [9] in a flow reactor to determine the decomposition rates of furan, 2-methylfuran and 2,5-dimethylfuran, and to analyse the pyrolysis products at very low pressure (0.0013 mbar) and over the temperature range of 1050–1270 K [9]. Thermal decomposition in shock tube of furan, 2-methylfuran and 2,5-dimethylfuran has been performed by Lifshitz et al. [10] over a temperature range of 1050–1460 K, at pressures of 2 atm. The rate coefficients for thermal decomposition of these furanic fuels have been found to increase as the alkylation of the furan ring increases. Theoretical calculations have been performed by Liu et al. [11] to compute the thermal decomposition of furan although any information has been given about the rate coefficients [11]. Sendt et al. [12] have reported the thermochemistry and rate parameters of several key reactions, concluding that 1,2-hydrogen atom transfer is the most important pathway for furan consumption in pyrolysis.

Recently the sooting propensity of furan, 2-methylfuran and 2,5-dimethylfuran has been studied in shock-tubes [13–15], and engines [16,17]. It has been found, particularly at the engine exhaust, that furanic fuels, in particular DMF, exhibit very similar combustion and emissions characteristics with respect to gasoline but DMF/diesel blend has been found to be more efficient in

particle reduction [18]. These results suggest that DMF could be suitable as fuel for the existing gasoline and diesel engines without major modifications and adjustments, still achieving equivalent engine performances and emission levels.

Different results have been obtained from studies performed in lab-scale reactors, such as jet-stirred reactor at pressure of 10 bar [19], flow-reactor [20] and low-pressure premixed flames [13–15,21–23]. In these conditions, furanic fuels produced a large amount of soot precursors including acetylene, 1,3-butadiene, 1,3-cyclopentadiene and benzene. In particular, DMF among the three furanic fuels seems to be the most effective in increasing soot precursor formation. These results and the kinetic model retrieved from their analysis constitute a benchmark for the study of furanic fuel oxidation and pyrolysis.

The lack of information on the sooting tendency of F, MF and DMF at atmospheric pressure has aimed our research group to perform experimental investigations on this topic. In particular, the effect of F, MF and DMF addition in ethylene flames on nanoparticle and soot formation has been investigated in counter-flow diffusion flames at atmospheric pressure by using optical techniques, i.e., laser induced incandescence and laser induced fluorescence, for the detection of combustion-generated particles. A detailed kinetic gas-phase model coupled to a multi-sectional approach has been used to predict combustion-generated particle formation and evolution and for the interpretation of the sooting tendencies [24]. In diffusion flame condition, furanic fuels, particularly MF and DMF have showed a significant increase in the formation of combustion-generated particles. Modelling analysis has showed, in agreement with experimental results, that the main pathways involved in the 2-methylfuran decomposition lead to formation of large amount of C₄-species (such as 1,3-butadiene and its radicals), which consequently increase benzene and polycyclic aromatic hydrocarbons (PAHs) concentrations, i.e., the concentrations of soot precursors. DMF decomposition to phenol increases the production of cyclopentadiene and thus naphthalene formation, although the overall PAHs reduction and particle nucleation inhibition resulted less strong than in MF.

In order to expand the knowledge on the sooting tendency of F, MF and DMF in other flame conditions, the present work reports an experimental study performed in atmospheric pressure premixed ethylene flames doped with up to 20% of the total carbon as F, MF and DMF. The detection of combustion-generated particles has been performed by means of in-situ laser spectroscopy, following our previous studies. In particular, UV-induced fluorescence emission at different wavelengths is used to detect different types of high-molecular mass aromatic compounds/nanoparticles constituted by clusters of aromatic compounds held together by van der Waals interactions, whereas laser induced incandescence emission is used to follow the formation of soot particles and their aggregates.

2. Experimental methods

Atmospheric pressure, premixed ethylene/air flames with equivalence ratios of 2.01, 2.16, 2.31, and 2.46 were stabilized on a capillary burner. The burner was cooled with recirculation water – 1 l/min – kept at 70 °C with a thermostatic bath and a steel plate was positioned at 3 cm from the burner exit to stabilize the flames. The flames so produced constituted our reference cases.

Furan, 2-methylfuran and 2,5-dimethylfuran were added to the premixed ethylene/air mixture by substituting part of the ethylene present in the reference cases. F, MF, and DMF were added as 10% and 20% of the total carbon fed. These large amounts of additive allowed us to explore combustion conditions similar to those encountered in real combustion devices.

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