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Characterization of the impact of ethanol on the formation of soot particles in gasoline turbulent diffusion flames

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Abstract

The objective of this study was to characterize the impact of ethanol as a fuel additive on soot formation in turbulent diffusion flames for different gasoline mixtures of varying alcohol concentrations. These jet spray flames were stabilized at atmospheric pressure. A TRF mixture (Toluene Reference Fuel) defined as a surrogate representative of the composition of standard gasolines of type SP95 was used as reference flame. To characterize the impact of ethanol on soot formation, 11 flames of TRF mixed with various ethanol mole fraction from 0 to 100% were studied by keeping either the lower heating value (LHV) or the mass flow rate (MFR) constant. Soot volume fraction (f_v) were measured by laser induced incandescence (LII). Absolute f_v values were obtained by an auto-calibrated method requiring a radiation source of known luminance to calibrate the detection system. The experimental results highlight a quasi-linear reduction of the soot formation with the added mole fraction of ethanol, until their complete disappearance for a pure ethanol flame. These results also highlight a stronger effect of ethanol on the soot reduction for ethanol addition with constant MFR characterized by a gradual decrease of the flame height. Experiments with commercial gasoline SP95-E10 and E85 were also carried out. The measured f_v profiles show strong similarities with the f_v profiles determined for their corresponding surrogates. These results illustrate that the TRF is an appropriate gasoline surrogate for studies aimed at characterizing soot formation. A modeling work, currently in progress, will be investigated to explain the inhibiting effect of ethanol on soot formation kinetics.

Keywords: Ethanol, Surrogate gasoline, Soot formation, Laser induced incandescence, Turbulent diffusion flame

Introduction

Ethanol is an attractive alternative fuel with a high octane number (RON/MON = 120/99) for spark-ignited internal combustion engine [1,2]. The addition of ethanol has shown to reduce the soot formation in gasoline sooting flames [3–6]. The soot volume fraction decreases when the quantity of ethanol is added to the laminar diffusion flame of gasoline [3,4]. Maricq *et al.* [5] investigated the impact of ethanol on the particles size distribution function (PSDF) in the laminar diffusion flame of gasoline. They found that substituting a small amount of ethanol in the gasoline had little impact on the PSDF. However, when the ethanol ratio was increased up to 85%, the primary particles constituting soot agglomerates decreased by more than 50% in comparison to that of the reference flame. Lemaire *et al.* [6] also studied the effect of ethanol addition on the formation of soot and polycyclic aromatic hydrocarbons (PAHs) in turbulent spray flames of gasoline and their surrogates. The advantage of spray flames is that it relies on the use of liquid fuel injection which avoids evaporation issues. Lemaire *et al.* [6] pointed out that the addition of 10-30% of ethanol to gasoline flames reduces the total soot quantity by 25-81%.

To better understand the role of ethanol in soot reduction, we investigated the soot formation in several atmospheric pressure spray flames of

gasoline surrogate TRF (Toluene Reference Fuel) with varying alcohol concentrations (0 – 100%) and constant lower heating value (LHV) or mass flow rate (MFR). Soot volume fraction (f_v) were measured by using auto-calibrated laser induced incandescence (LII). The obtained results provide some valuable information regarding the powerful impact of ethanol on soot reduction. Furthermore, we also highlight the very good mimicry of the SP95-E10 and E85 by the TRF-Ethanol mixtures regarding soot formation.

Experimental set-up

Burner and flame conditions

The turbulent spray flames were stabilized on a homemade burner consisting of a nebulizer (DIHEN-170-AA) [7] for liquid fuel's atomization and the oxidation air stabilization system. The atomization equipment plays a major role in the characteristics of the combustion process of two-phase flow. Smaller the droplets, faster is the vaporization of the liquid and more effective is the combustion. The liquid fuels were injected by a capillary tube and cut by a high-speed flow gas circulating in an annulus area to create the spray. In this study, we used nitrogen as the nebulized gas. The air flow was injected under a honeycomb structure which allows a better stabilization of the flame. The nebulizer was placed in the center of the honeycomb structure. For all studied flames, the

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flow rates of N₂ and Air were kept constant at 0.25 L.min⁻¹ and 19.6 L.min⁻¹ respectively. The mass flow rate of the liquid fuel for 11 flame TRF-Ethanol are reported in Table 1 and 2.

The studied mixtures were prepared by mixing 4 pure chemical compounds: iso-octane, toluene, n-heptane and ethanol. The mole fraction of the mixtures was verified by using gas chromatography (GC). The uncertainty associated with the mole fractions of each of the compounds was determined to be less than 3%.

Temperature measurement

	Flame					
	TRF	TRF-E	TRF-E40	TRF-60	TRF-E85	Ethanol
$x_{\text{iso-octane}}$	0.447	0.358	0.268	0.179	0.031	0.000
x_{toluene}	0.423	0.338	0.254	0.169	0.029	0.000
$x_{\text{n-heptane}}$	0.130	0.104	0.078	0.052	0.009	0.000
x_{ethanol}	0.000	0.200	0.400	0.600	0.931	1.000
Mass flow rate of fuel MFR (g.h ⁻¹)	98.2	102.1	107.6	115.8	145.2	158.1
Lower heating value of fuel LHV (kJ.h ⁻¹)	4.24	4.24	4.24	4.24	4.24	4.24
Flowrate of N ₂ (NL.min ⁻¹)	0.25	0.25	0.25	0.25	0.25	0.25
Flowrate of Air (NL.min ⁻¹)	19.6	19.6	19.6	19.6	19.6	19.6
T _{max} (K)	1928.3	1934.6	1943.1	1926.2	1945.2	1941.0

Table 1. Experimental conditions for the 6 flames with same LHV. $x_{\text{iso-octane}}$, x_{toluene} , $x_{\text{n-heptane}}$, x_{ethanol} are the mole fraction of iso-octane, toluene, n-heptane and ethanol in the mixture. T_{max} is the maximum measured temperature by using thermocouple

	Flame					
	TRF	TRF-E*	TRF-E40*	TRF-60*	TRF-E85*	Ethanol*
$x_{\text{iso-octane}}$	0.447	0.358	0.268	0.179	0.031	0.000
x_{toluene}	0.423	0.338	0.254	0.169	0.029	0.000
$x_{\text{n-heptane}}$	0.130	0.104	0.078	0.052	0.009	0.000
x_{ethanol}	0.000	0.200	0.400	0.600	0.931	1.000
Flowrate of fuel (g.h ⁻¹)	98.2	98.2	98.2	98.2	98.2	98.2
Lower heating value of fuel (kJ.h ⁻¹)	4.24	4.08	3.87	3.59	2.86	2.63
Flowrate of N ₂ (NL.min ⁻¹)	0.25	0.25	0.25	0.25	0.25	0.25
Flowrate of Air (NL.min ⁻¹)	19.6	19.6	19.6	19.6	19.6	19.6
T _{max} (K)	1928.3	1953.6	1976.7	1987.2	1949.4	1949.4

Table 2. Experimental conditions for 6 flames with same mass flow rate of fuel MFR. $x_{\text{iso-octane}}$, x_{toluene} , $x_{\text{n-heptane}}$, x_{ethanol} are mole fraction of iso-octane, toluene, n-heptane and ethanol in the mixture T_{max} is the maximum measured temperature by using thermocouple. * symbol indicates the flames with MFR constant

Determination of soot volume fraction profiles

Soot volume fraction profiles were determined using laser induced incandescence (LII) measurements carried out by using a laser excitation wavelength at 532 nm. The laser beam passed through a pinhole to provide a top-hat irradiance profile at the position of the LII measurement in the flame. The LII signal was collected at right angle from the laser axis by a system of two lenses ($f_1 = 400$ mm and $f_2 = 200$ mm) associated to a spectrometer Acton SP300i. A diffraction grating 150 groves/mm, blazed at 800 nm (high transmission

The vertical centerline temperature profiles of the studied flames were measured with a type S thermocouple (TC S.A., Pt-Pt/10%Rh) having 0.35 mm wire diameter and 1.01 mm bead diameter. The thermocouple was cleaned by placing it in the blue part of the flame after every three measurements in the sooting zone. The thermocouple temperature measurements were corrected from the radiation losses by following the recent and simple method described in Elias et al [8]. The uncertainty on the temperature was estimated around ± 100 K.

in near IR) was used. The center wavelength of the diffraction grating was set at 750 nm allowing to collect the emission spectral range from 609 nm to 890 nm. The spectrometer had two outputs providing to collect the signal either by a photomultiplier tube Philips XP2020Q (PMT) or by a Roper PIMAX 4 ICCD camera. A Notch filter centered at 532 ± 9.8 nm was positioned just before the spectrometer to remove part of the intense Rayleigh laser scattering.

The LII signal from soot particles were measured with the detection system characterized by the solid angle of collection Ω and the transmission

coefficient β . Thus, the detected LII signal could be expressed as:

$$S_{LII,detected}(T_p, \lambda) = \frac{12\pi hc^2 E_m(\lambda)}{\lambda^6} \cdot \frac{1}{\exp\left(\frac{hc}{\lambda k_B T_p}\right) - 1} \cdot V_m \cdot \Omega \cdot \beta \cdot \Delta\lambda \cdot f_v \quad eq.1$$

where λ is the emission wavelength, k_b the Boltzmann constant, c the light speed, h the Planck constant. T_p the average temperature of soot particles determined on the LII spectrum. $\Delta\lambda$ is the emission spectral range. f_v is the soot volume fraction.

In order to get absolute f_v values from the LII signal, we calibrated the detection system by using an auto-calibrated method requiring a radiation source of known luminance. This method also relies on the determination of the absorption function of the particle $E_m(\lambda)$ and the measurement volume V_m which can be defined according to the expression :

$V_m = 2 \times l_{pixel} \times \frac{\pi \phi_{laser}^2}{4} \cdot h_{slit}$, where l_{pixel} is the size of 1 pixel, ϕ_{laser} is the diameter of laser beam, h_{slit} is the open of slit. $E_m(\lambda)$ has been determined along the vertical centerline of the flame using the low laser fluence methodology [9]. This method relies on the determination of the soot heated temperature T_p by the laser pulse for different laser energies, connected to the $E_m(\lambda)$ according to the following expression:

$$E_m(\lambda) = \frac{(T_p - T_g) \cdot \lambda_{laser} \cdot \rho_s \cdot c_s}{6\pi \cdot F} \quad eq.2$$

where T_g is the temperature of flame, λ_{laser} is the laser wavelength, F the laser fluence, ρ_s the density of soot particles and c_s is the specific heat of soot particles. The density of soot ρ_s in the sooting flame of this study was extrapolated along the centerline of the flame from 1.3 g.cm⁻³ (50 mm HAB corresponding to nascent soot) to 1.8 g.cm⁻³ (210 mm HAB corresponding to mature soot).

Finally, the term $\Omega \cdot \beta$ in *eq.1* can be obtained from the measurement of calibrated black body emission spectra transmitted from the calibrated integrated sphere. This way, it is possible to calibrated the LII signal issued from the flame into absolute f_v values.

Sprays characterization

The structure of the TRF sprays with and without the flame were measured by using the corresponding Mie scattering signals upon 532 nm excitation wavelength. The Mie scattering signal was collected with the same detection system as for LII measurements without the Notch filter 532 ± 9.8 nm placed in front the spectrometer.

Results and discussions

TRF flame jet characterization

The structure of the TRF sprays with and without the flame are presented in Fig.1. We observe that the liquid fuel droplets in the TRF flame disappear at 20

mm HAB while the liquid fuel droplets evaporate completely at HAB = 70mm in TRF cold jet.

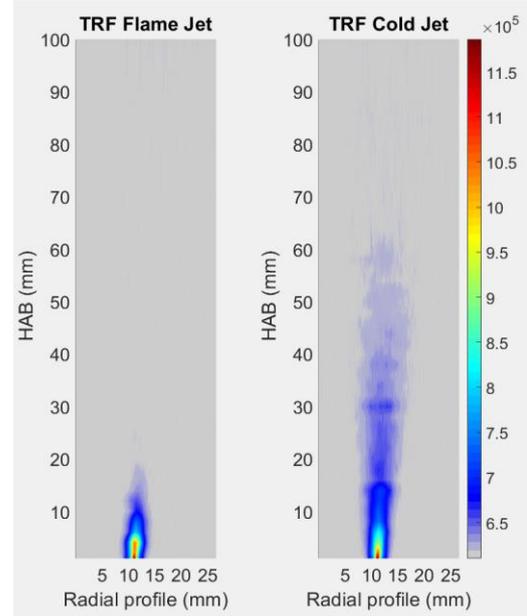


Fig.1. Liquid droplet distribution measured by Mie scattering in TRF flame (left) and TRF cold jet (right)

Impact of ethanol on the temperature profile

Fig.2 shows the impact of ethanol on the temperature profile of TRF flame in the case of constant LHV (Fig.2a) and in the case of constant MFR (Fig.2b). In both cases, we observe that ethanol substitution does not change the maximum temperature reached in both flames as shown in Table 1 and 2. In the case of constant LHV (Fig.2a), the increasing ethanol concentration (from 0 to 100%) shifts the temperature profile to the burnt gas zone. This observation could be explained by the increase of mass flowrate of the fuel.

In the case of constant MFR, the temperature profile shifts towards the nebulizer when the quantity of ethanol increases in the TRF mixture. This observation may be caused by keeping the mass flow rate of mixture leading to decrease of their LHV. The reduction of LHV reduces the flame power meaning decrease in the flame height.

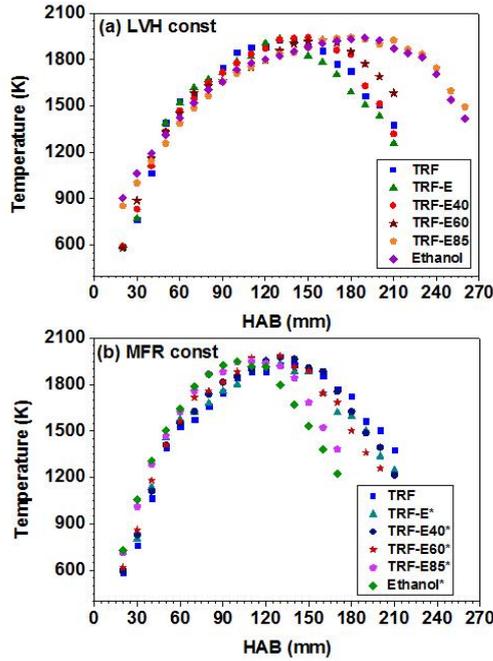


Fig.2. Impact of ethanol on the temperature profile of TRF flame in the case of LVH const (a) and in the case of MFR const (b)

Impact of ethanol on soot formation

LVH constant

Fig.3 shows pictures of the 6 TRF-Ethanol flames in the case of constant LVH. The parameters of the camera to take these pictures have been kept identical for the whole flames. When the quantity of ethanol in the mixture increases, we observe that the flame height increases while the orange color decreases and even disappears completely in the 100% ethanol flame. The 2D imaging of the LII signal rebuilt from the 1D spectral measurements carried out in the 6 flames are reported in Fig.4. From these images, we could determine the evolution of f_v along the vertical centerline for the whole flame. This data is reported in Fig.5 and highlights the impact of ethanol on the soot volume fraction profile in the various TRF flames. The maximum soot volume fraction in the reference TRF flame is about 664 ppb at 130 mm. We observe that the substitution of 20% ethanol in mole (TRF-E) slightly decreases the soot volume fraction (562 ppb) which continues to decrease with increasing ethanol concentration in the mixture. The highest value of f_v in the TRF-E85 flame corresponding to 85% ethanol in volume or 93.1% in mole in the mixture, is only 9 ppb. Finally, we note that the soot particles disappear completely (no measurable LII signal) in the flame with 100% ethanol.



Fig.3. Pictures of the 6 TRF-Ethanol (0-100%) flames with constant LVH

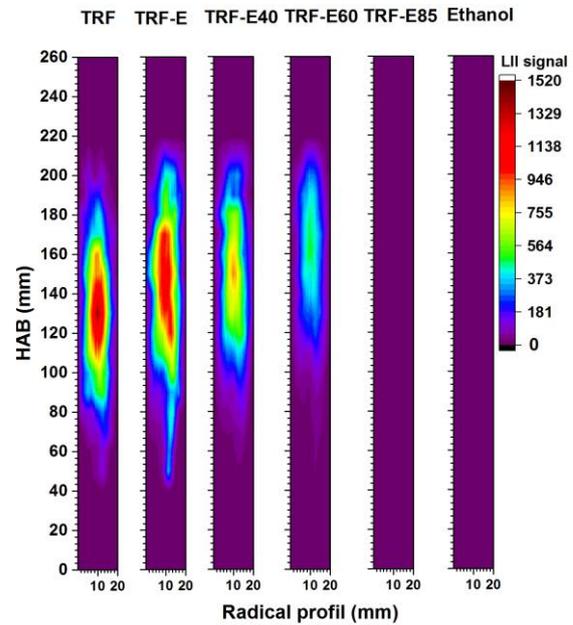


Fig.4. 2D LII imaging in the 6 TRF-Ethanol (0-100%) flames with constant LVH

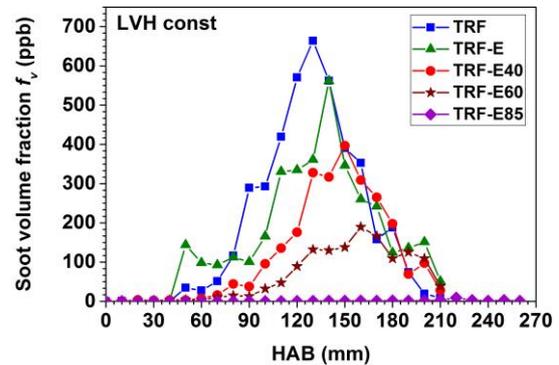


Fig.5. Impact of ethanol on soot volume fraction measured in the center line of the TRF flames with constant LVH

All these experiments were carried out with identical flowrates of N_2 and air for all studied flames but variable ethanol flowrates. It is known that the increase of the mass flowrates of the fuel decreases the efficiency of the nebulizer for the atomization of the injected fuels leading to a reduction of the combustion efficiency [7] and an increase in the soot formation. However, we clearly observe a strong reduction of the soot formation with the increase of the mass flow rate in the case of ethanol indicating the strong chemical effect of ethanol to inhibit the formation of soot in the TRF flame.

MFR constant

Fig.6 shows the pictures of the 6 TRF-Ethanol flames (from 0 to 100% ethanol) stabilized for an identical mass flow rate equal to $98.2 \text{ g}\cdot\text{h}^{-1}$. In this case, we observe that the orange color and the flame height decrease with increase of ethanol. We also reported in Fig.7 the corresponding 2D LII imaging determined for these flames. The decrease of soot signal in the case of constant MFR is faster than in the case of constant LHV. The soot particles disappear completely (no measurable LII signal) in the flame TRF-E85*.

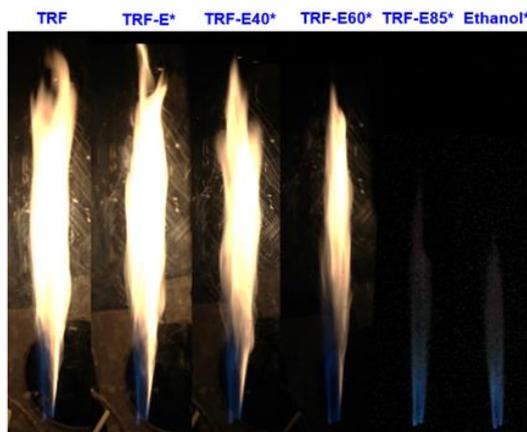


Fig 6. Pictures of the 6 TRF-Ethanol (0-100%) flames with constant MFR

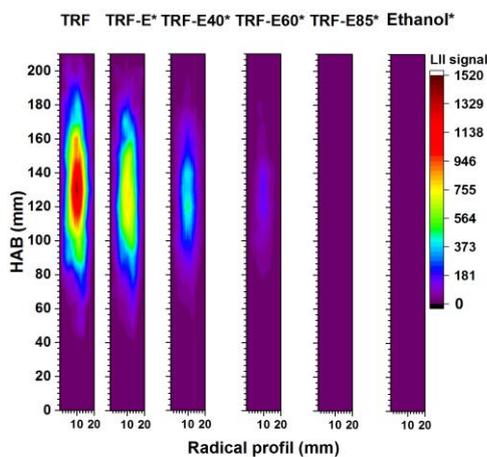


Fig 7. 2D LII imaging in the 6 TRF-Ethanol (0-100%) flames with constant MFR.

The soot volume fraction measured in the flame center line decreases with the ethanol increasing as presented in Fig.8.

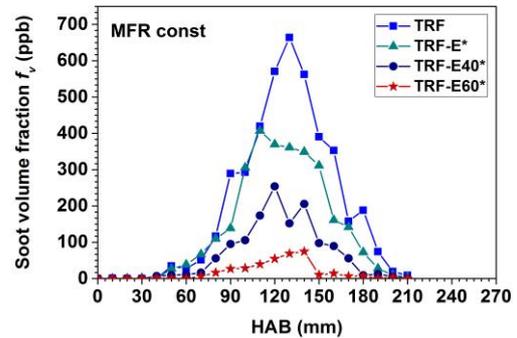


Fig.8. Impact of ethanol on soot volume fraction measured in the center line of the TRF flames with constant MFR.

Comparison between the surrogate and real fuel

The soot volume fraction profiles of the TRF-E, TRF-E* and TRF-E85 flames were compared with that of their real fuels and shown in Fig.9.

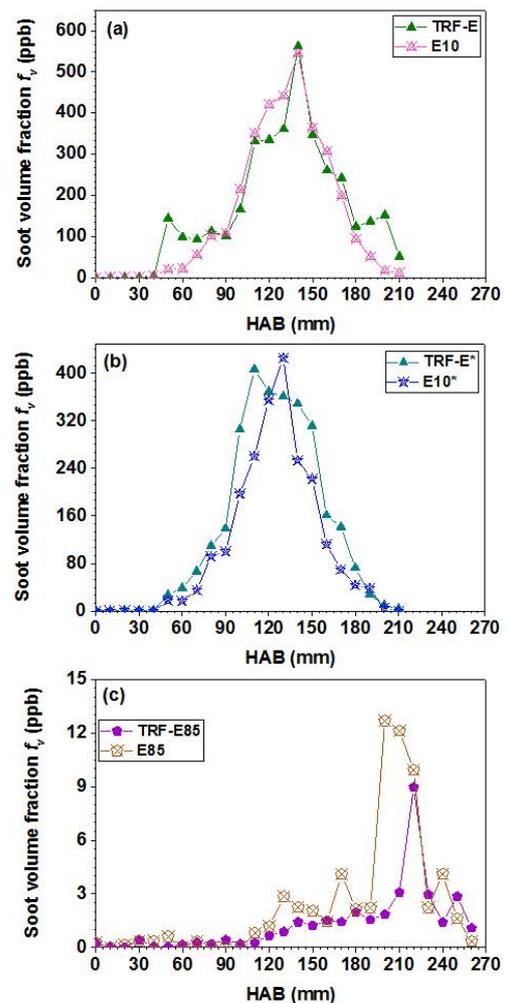


Fig.9. Comparison of the soot volume fraction profiles obtained with the commercial fuel E10, E10* and E85 with their surrogates.

The soot particles were not detected in the flame TRF-E85*, thus, the comparison was not realized for this flame. The soot volume fraction profiles of TRF-E, TRF-E* and TRF-E85 are quite similar with that of their corresponding real fuels. Thus, the surrogates used in the present study are good and well suited to investigate the impact of ethanol on the soot formation in gasoline flame.

Discussion of the kinetic impact of ethanol on soot formation.

Fig.10 shows the maximum soot volume fraction values determined in the whole flame plotted versus the mole fraction of ethanol in the TRF mixtures. In both cases (constant MFR or LHV), we observe a quasi-linear reduction of the soot formation with respect to the mole fraction of ethanol. These results were also observed in the laminar diffusion flame of gasoline published by Khosousi et al [4]. Besides, we also observe in Fig.10 that the reduction of the soot volume fraction is more efficient with substitution of ethanol with constant MFR than that with constant LHV. This difference in efficiency is very probably related to the mass flow rate of mixture in the case of constant MFR lower than that of LHV constant.

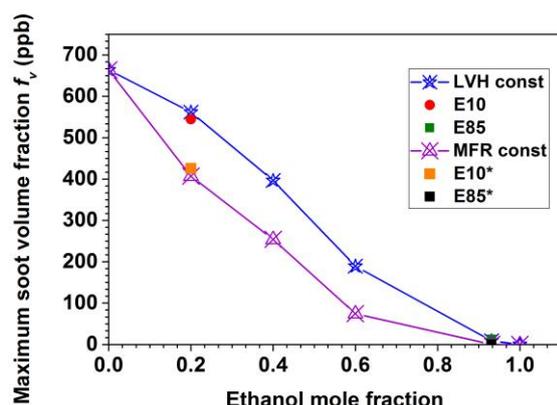


Fig.10. The impact of ethanol on the maximum soot volume fraction of TRF flame

The kinetic impact of ethanol on soot reduction in gasoline and gasoline surrogate flames has been discussed in literature [6,10]. Lemaire et al. [6] pointed out that the soot reduction is due to the combined effects of dilution and oxygen contained in ethanol leading to oxidize the soot particles faster. Xu et al. [10] suggested that adding alcohol can inhibit the formation of PAHs due to massively-produced HO₂ and OH radicals from alcohol.

Conclusion

In this work, we carried out a series of experiments aimed to characterize the impact of ethanol on the soot formation in different turbulent diffusion flames of gasoline. Two approaches have been carried out to investigate the impact of ethanol by keeping the lower heating value of fuel constant or by keeping the mass flow rate of fuels constant. In both cases, we observed a quasi-linear reduction of

soot volume fraction in the TRF flame with increasing of ethanol added but with a stronger efficiency in the case of constant MFR. Moreover, we have also an excellent agreement between the experiment carried out with commercial fuels E10, E10* and E85 and their corresponding surrogates which therefore validate these surrogates for the soot formation studies related to these commercial fuels.

We also showed that the centerline temperature profiles of all these flames highlight similar maximum values. This point therefore indicates the f_v reduction is mainly related to the chemical effect of ethanol, for which a modeling work will be soon performed to lead to a deeper understanding of this process.

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