



Effect of alkylated aromatics on particle formation in diffusion flames: An experimental study



Marielena Conturso*, Mariano Sirignano, Andrea D'Anna

Dipartimento di Ingegneria Chimica, dei Materiali e della Produzione Industriale – DICMAPI, Università degli Studi di Napoli Federico II, Italy

ARTICLE INFO

Article history:

Received 26 August 2015

Accepted 26 August 2015

Available online 1 September 2015

Keywords:

Sooting tendency
Nanoparticles
Alkyl-benzenes
Counter-flow flame
Optical diagnostic

ABSTRACT

The sooting tendency of four C_8H_{10} isomers, specifically ethylbenzene, ortho-, meta- and para-xylene, has been studied in atmospheric pressure, counter-flow diffusion flames of ethylene, whose fuel stream was doped with 10%, 20% and 30% of C_8H_{10} isomers. In-situ spectroscopy, namely laser UV-induced emission, is used as diagnostic tool to detect different types of combustion-formed nanoparticles by changing the detection wavelength from the UV to the visible. Laser induced incandescence has been also measured to detect soot particles. Experimental results are compared to those obtained in an ethylene/toluene flame operated in the same conditions to understand the effect of the number and position of the alkyl groups and of the alkyl chain lengths on particulate formation.

The experimental results show that the effect of the alkyl chain on soot tendency strongly depends on the combustion environment. In low temperature and pyrolytic zones of the flame, the long chain promotes nanoparticle formation. Aromatics containing non-adjacent methyl groups are more stable and hardly react forming small quantities of nanoparticles. In high temperature and oxidative zones, the behavior is the same but nanoparticles are produced in lower amount, especially for higher percentages of aromatic fuel addition. Regarding sooting tendency, in the pyrolytic region of the flame the presence of a longer alkyl chain gives a much stronger soot production, whereas in oxidative region the differences among the additives are smaller. The strongest increase of soot production occurs moving from the methyl to ethyl group addition to the aromatic ring showing that, in all the experimental conditions we have examined, a shorter chain and the presence of methyl groups positioned far from each other on the aromatic ring give the lowest sooting tendency.

© 2016 Published by Elsevier Inc.

1. Introduction

Aromatic hydrocarbons, particularly mono-ring aromatics, are the major constituents of automotive fuels, about 25% by volume in gasoline, 33% in diesel oil, and 16% in jet fuels (JP-8, Jet A/A-1) [1–3]. They are used as anti-knock additives to enhance octane number and suppress auto-ignition or to increase the energy density of transportation fuels. Their resistance to low-temperature oxidation and auto-ignition is due to the inhibitory effect of the aromatic ring on branching radical reactions.

Aromatic ring plays also a crucial role in the formation of particulate matter, enhancing the formation of soot precursors such as Polycyclic Aromatic Hydrocarbons (PAHs) [4,5]. Mono-ring aromatics have greater sooting tendency with respect to aliphatic compounds, presumably because the aromatic ring stays largely

intact during oxidation and thus the kinetically slow process of first ring formation is by-passed. For this reason, the rate-limiting step becomes the formation of the second ring (naphthalene) and rises questions regarding which pathways are significant for the formation of naphthalene from fuels with mono-ring content [6].

Researches about sooting tendency of aromatic hydrocarbons dates back to the seventies but recently a deep and wide collection of experimental data is available, although just few experimental conditions have been investigated. McEnally and Pfefferle [7,8] have studied the sooting tendency of aromatic hydrocarbons by measuring maximum soot volume fractions on the centerline of a coflowing methane/air non-premixed flame doped with 5–400 ppm of the test hydrocarbons. More recently, using a similar experimental system, Witkowski et al. [9] evaluated the soot properties of real fuels and surrogate fuels with higher dopant concentration (2200 ppm), while Xuan and Blanquart [10] investigated through a numerical modeling study the sooting tendencies of some aromatic and aliphatic compounds.

* Corresponding author at: Dipartimento di Ingegneria Chimica, dei Materiali e della Produzione Industriale – DICMAPI, Università degli Studi di Napoli Federico II, Piazzale V. Tecchio, 80, 80125 Napoli, Italy.

E-mail address: marielena.conturso@unina.it (M. Conturso).

Previous studies on sooting tendency have been based on the measurement of the soot volume fraction on the centerline of the flame. This flame zone is not a good indicator of the amount of soot particles formed and particularly of their size and morphological properties for several reasons. Maximum soot formation does not occur on the centerline of a diffusion flame but on the flame wings. Particles measured on the centerline are the result of primary particle formation, agglomeration and maturation that arrive on the flame centerline from other zones of the flame avoiding a clear identification of the role of the fuel chemical structure on the pathways controlling the particle formation process [11]. Furthermore, the concentration of dopants used in the literature is too small related to the percentages of aromatic hydrocarbons in commercial fuels making difficult a direct comparison of research results with real combustion systems.

In order to fill this gap of knowledge, in the present paper the role of larger concentrations (up to 250,000 ppm of additive blended with ethylene) of mono-ring aromatics on particulate matter formation has been studied in an atmospheric pressure counter-flow diffusion flame of ethylene by simultaneous detection of UV-Laser induced Fluorescence (LIF) and Laser Induced Incandescence (LII). The use of the two diagnostics allows to track soot precursor nanoparticles and soot particles contemporarily. Four C_8H_{10} isomers, namely ethylbenzene, ortho-, meta- and para-xylene, have been selected as additives. Results are compared with those obtained by using toluene as additive to ethylene in the same operating conditions.

2. Experimental methods

A counter-flow burner system was used to stabilize flames of ethylene/aromatic fuel blends. The burner consisted of two opposed jet nozzles (ID 2.54 cm) vertically positioned; the oxidizer stream is introduced from the upper nozzle while the fuel stream was introduced from the lower nozzle. Flames were stabilized by feeding 25 vol% ethylene-aromatic mixtures and 75 vol% Ar as fuel stream, and 22 vol% O_2 and the remaining Ar as oxidizer stream. The oxidizer and fuel stream velocities were fixed at 16.1 and 13.2 cm/s at standard conditions, respectively.

The distance between the two opposed jet nozzles was maintained constant at 1.5 cm for all measurements. In the selected operating conditions, the flame front was located in the oxidizer side so that by-products produced in both fuel and oxidizer side were convected toward the stagnation plane without encountering the flame front avoiding particle oxidation.

Four C_8H_{10} liquid isomers, namely ethylbenzene, ortho-, meta- and para-xylene, were selected as additives. Toluene was used as reference mono-ring aromatic. The aromatic component concentration was changed in order to have 10%, 20% and 30% of aromatic carbon of the total carbon fed to the flame, keeping constant the total carbon flow rate. In order to achieve these conditions, the argon flow rate in the fuel stream was adjusted to account for the different molecular weight of the additives. This results in an identical carbon flow rate as well as cold gas velocity, allowing a reliable comparison for the different additives used [14–16]. Fuel stream was preheated to allow aromatic fuels to completely evaporate before exiting the fuel nozzle. The preheating temperature set point and the temperature of recirculating water were fixed to maintain the inlet temperature of fuel stream of 150 °C so that the fuel blend enters the burner as vapor.

Particulate matter formation was detected by spectral laser induced emission measurements in the 200–550 nm range, performed by using as excitation source the fourth harmonic of a Nd:YAG laser at 266 nm. The experimental setup was the same as that used in previous works [12,13]. Some details are reported below.

Laser beam – diameter at focal point of 350 μm – was focused in the center of the flame at different distances from fuel nozzle exit with a spatial resolution of 0.2 mm. The emitted radiation at 90° with respect to the laser beam was focused onto the 280 μm entrance slit of a spectrometer and detected by an intensified CCD camera thermoelectrically cooled down to -10°C to reduce noise. The energy of the laser pulse was kept constant at 0.8 mJ with pulse duration of 8 ns. The chosen laser energy gave the better compromise between laser induced emission signals and species fragmentation interference. The measured spectra were corrected for the spectral response of the detection system.

The emission spectra were detected using a gate time of 100 ns synchronized with the laser pulse and by summing the CCD counts over 150 scans. In this way both fluorescence and incandescence signals were detected contemporarily.

Two broad peaks were distinguished in the emission spectra: one in the UV region around 350 nm and the other in the visible around 400 nm associated with Laser Induce Fluorescence (LIF). For larger wavelength, namely at 550 nm, the signal has been related to Laser Induced Incandescence (LII) due to the almost absence of fluorescence signal.

On the basis of LIF emission wavelengths and LIF lifetimes, fluorescence signals were attributed to nanoparticle of organic carbon having different chemical and morphological structures [12,13]. In particular, LIF in the UV was attributed to single high-molecular weight aromatic molecules with few condensed aromatic rings connected by σ -bonds, whereas LIF in the visible was attributed to highly-packed, sandwich-like clusters of aromatic hydrocarbons. Incandescence signal was attributed to solid soot particles which are able to dissipate the acquired energy by thermal emission more than by fluorescence emission.

In order to evaluate and subtract the contribution of the incandescence signal, a black-body radiation curve at 4000 K has been used. Signal collected between 540 nm and 560 nm is well fitted with a black body curve, suggesting that a minimum contribute would come from C_2 Swan band at 550–560 nm.

Peak temperatures estimated by adiabatic flame temperature show to not significantly vary by changing the additive. This is due to large dilution of fuel (75% of Argon is used) and of oxidant (78% of argon) and to the fact that the total carbon flow rate as well as the cold gas velocity have been kept constant. Although other phenomenon and properties, i.e., diffusivity of added species, could have minor effect on peak temperatures, here the effect on particle formation has not considered being linked to temperature changes.

3. Results and discussion

Emission signals were measured along the distance between the two nozzles, starting from the fuel nozzle. Measurements were performed for five fuel blends, with concentrations of the aromatic additives varying from 0% to 30% of aromatic carbon of total carbon fed to the flame.

Fig. 1 reports typical profiles of the intensities of LIF in the UV at 350 nm and visible at 400 nm, as determined by the measurements of the emission spectra for the ethylene/ethylbenzene blend at 30% of aromatic carbon. The location of the stagnation plane is at 4.2 mm and it is reported as a vertical dashed line to allow an easier identification of the flame zones. The flame front is located in the oxidizer side at around 8 mm. Fluorescence emission is detected along the entire flame, from 2 to 6 mm. The main gas phase by-products such as acetylene and benzene are formed in the oxidizer side of the flame – right side with respect to the stagnation plane in Fig. 1 – and their concentrations maximize across the stagnation plane [14]. Looking at emission signals around the stagnation plane, the incandescence signal starts to overlap

Download English Version:

<https://daneshyari.com/en/article/651062>

Download Persian Version:

<https://daneshyari.com/article/651062>

[Daneshyari.com](https://daneshyari.com)