Sooting tendencies of primary reference fuels in atmospheric laminar diffusion flames burning into vitiated air

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ABSTRACT

In this study, the sooting tendencies of primary reference fuels (PRFs) are measured in term of yield sooting indices (YSIs) in methane diffusion flames doped with the vapors of PRFs. The present paper represents an incremental advance complementing the original methodology prescribed by McEnally and Pfefferle. The influence of both PRF formulation and CO_2 dilution of the coflowing air on the YSIs is also assessed. The diffusion flames burning in a coflowing oxidizer stream are established over the Santoro's burner and vapor of the liquid fuel to be investigated is injected into the fuel stream. Laser extinction measurements are performed to map the two-dimensional field of soot volume fraction in the flame. For the pure liquid hydrocarbons investigated, i.e., *n*-hexane, *n*-heptane, isooctane, and benzene, the YSI reported in the original paper by McEnally and Pfefferle quantitatively predict the sooting propensities, measured here at much higher dopant concentrations. The present study therefore extends the consistency of the YSI methodology on the Santoro's burner. For blends of *n*-heptane and isooctane, the sooting tendency of doped flames exhibits regular and monotonic trends and decreases with increasing *n*-heptane mole fraction or CO_2 dilution. Interestingly, the evolution of YSI with the isooctane mole fraction exhibits a strong similarity for varying CO_2 mole fraction. A quadratic least-squares fit is then derived, providing a phenomenological model of YSI as a function of both isooctane mole fraction in the fuel stream and CO_2 mole fraction in the oxidizer. A non-negligible cross effect of PRF formulation and CO_2 dilution on YSI is revealed. The method elaborated within the framework of the present paper could be extended to surrogate fuels. This would help develop a comprehensive database and empirical correlations that could predict the sooting propensities of different surrogate fuels, therefore their potentially mitigationed soot production through control of fuel composition and/or exhaust gas recirculation. This database would also be useful for the validation of CFD simulations incorporating sophisticated model of soot production.

1. Introduction

The growing concerns about increasing energy demand, energy efficiency policies, energy security issues, hazardous effects of combustion-related emissions on human health, and global warming change have led to more and more stringent regulations on exhaust emissions from combustion devices. The recent legislation and guidelines to mitigate emissions related problems prescribe the integration of renewable fuels into the transportation fuel pool and the use of high-efficiency combustion devices to reduce CO_2 emissions [1,2]. These challenges can be converted into opportunities through a careful selection of renewable/alternative fuel compositions and efficient advanced combustion processes.

Alternative and biofuels exhibit a wide range of combustion characteristics and induce different effects on combustion processes, and therefore ultimately on exhaust emissions. Hence, mixtures composed of alternative fuels could be designed specifically to comply with a set of requirements, for instance, to reduce soot and NO_x at the same time [3].

Soot particles are directly related to the reduction of combustion efficiency [4,5]. Ideally, CO_2 and H_2O are the only products of complete combustion, but in practical devices, intermediate species formed due to the pyrolysis of hydrocarbons in locally fuel-rich zones lead eventually to the production of soot particles through complex physicochemical processes involving gaseous species and solid particles [6]. Increased risks of cardiac arrest, pulmonary diseases, pregnancy complications, and asthma are now associated with the presence of particulate matter in urban air and are also linked to higher mortality rates [4,7–9]. According to some estimate, soot particles may play an even more critical role in global warming than methane, which makes the second highest

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contribution to the greenhouse effect [10]. Hence, reduction of soot emissions into the atmosphere would also be a relevant policy to reduce global warming [10], complementing the regulation of CO_2 emissions.

However, many gaps have been identified in the available knowledge about the production of particulate matter during combustion process. For this reason, the number of studies on soot formation/oxidation has increased for the past two decades [10–13]. Meaningful reviews have namely been published that characterize the main formation pathways for polycyclic aromatic hydrocarbons (PAH) and ultimately soot particles in laboratory reference flames [6,10,14–17].

These new insights into the physicochemical processes governing soot formation and oxidation and the influence of different compounds on these processes will help design combustors and/or fuel compositions for future applications with lower environmental impact. Emission of soot particles can indeed be reduced through either changes in the design of combustor or modification of fuel composition. The basic advantage of fuel modification is the immediate effect on the existing devices. Fuel modification strategies range from doping fuels with additives to reformulating a fuel's bulk composition (e.g., blends of conventional and renewable fuels), which may promote some chemical pathways that avoid soot formation [10]. The number of possible fuel additives and reformulation strategies is unlimited, and they will become more and more attractive with the increasing price of fossil fuels. Thus, there is a need to especially characterize soot production processes induced by these strategies.

Under atmospheric conditions, many practical fuels, such as those derived from fossil oil, are liquid mixtures of hundreds of hydrocarbons that have variable and coupled effects on regulated engine emissions [3,18,19] and especially soot production [10,20]. This makes modeling and simulation of the sooting behavior of these fuels a significant computational task. As a result, many works focus on developing a molecular-level understanding of fuel combustion processes and surrogate fuels. A surrogate fuel is a mixture of selected pure chemical compounds in known concentrations and exhibits physical (density, viscosity, surface tension, vapor pressure, etc.) and chemical (ignition delay, hydrogen/carbon ratio, cetane or octane number, laminar flame burning rate, etc.) properties similar to those of the fuel it represents. The study of such surrogate fuels is required for the validation of multidimensional computational fluid dynamics (CFD) simulations. As an illustration, the simplest gasoline surrogates could be binary mixtures of isooctane and *n*-heptane, called primary reference fuels (PRFs) [21–23].

Among other characteristics, the propensity of a surrogate fuel to produce soot should match the one exhibited by the real fuel it represents [19]. The relevance of a given surrogate fuel's sooting propensity is then assessed when the soot productions by the surrogate and the real fuels in practical devices are componed [11,24]. But the sooting propensity of a surrogate fuel also allows the selection of potential additives that may significantly modify the sooting propensity of the real fuel without any radical change in the other regulated fuel properties. The relatively simple formulation of the surrogate fuel then enables the identification, and therefore possibly the control of the pathways governing the observed trends [10]. Therefore, a thorough characterization of the blending effect on the sooting propensity of surrogate fuels is crucial.

The control of soot production by the addition of gaseous species into fuel and/or oxidizer is also an attractive strategy. The effect of CO_2 addition on the sooting propensity of a given fuel has especially been investigated, because many practical devices implement exhaust gas recirculation (EGR). As discussed by Du

et al. [25], the introduction of an additive such as CO_2 affects soot formation through the following three paths:

1. dilution effect because of reduction in the concentrations of the reactive species;
2. thermal effect because of the change in flame temperature;
3. possible direct chemical effect when the additive contributes to the chemical reactions related to soot formation and oxidation.

CO_2 addition then induces a significant effect on the amount of soot produced. Liu et al. [26] argued that the aforementioned three effects occur simultaneously and are intimately coupled. CO_2 addition not only reduces the concentrations of reactive species (dilution), but also lowers the flame temperature (thermal). In their numerical study of laminar jet diffusion flames [26], Liu et al. highlighted the chemical effects of CO_2 on soot formation. These authors showed that the reaction $CO_2 + H \rightarrow CO + OH$ is primarily responsible for the chemical effects of CO_2 addition, as reported in earlier studies [27]. This effect is especially significant for CO_2 addition to the oxidizer side because of the crucial role of hydroxyl radicals OH into the soot oxidation process [28]. Thus, the effect of CO_2 addition on the sooting propensity of surrogate fuels also needs to be characterized.

When the sooting propensity of a surrogate fuel, is evoluated a major challenge comes from the low volatility of its components, which makes difficult the ignition and the stabilization of the standard experiment conducted. To overcome this challenge, some studies have suggested that combustion of prevaporized surrogate fuels is an effective first step in evaluating their sooting propensities [13,29–32]. To this end, laboratory flames burning the fuel vapor seeded in a carrier gas have been extensively used, setting up different experimental and postprocessing techniques [10,14,29,32–36]. The sooting tendencies of a large number of chemical compounds, surrogate fuels, hydrocarbon- based commercial fuels, and mixture of hydrocarbons and biofuels have then been reported in terms of smoke point height and threshold sooting index (TSI) in laminar diffusion flames [12,33,37,38].

However, the smoke point height h globally tends to decrease with the fuel molecular weight, indicating an increasing TSI that is a linear function of $1/h$. Therefore, at a given spatial resolution of the detection system, the TSI sensitivity decreases with the fuel molecular weight. Further considering the uncertainty associated to any methodology, the discrimination among the TSIs exhibited by heavy fuels then becomes questionable. Moreover, in the TSI methodology, the smoke point height is inferred only from visible flame shape. For this reason, McEnally and Pfefferle [29] suggested a complementing index, called the yield sooting index (YSI). Following this methodology, the maximum soot volume fraction $f_{v,max}$ is actually measured in laminar jet methane/air diffusion flame whose fuel stream is doped with a small amount of vapor of the liquid fuel to be investigated. $f_{v,max}$ is then converted into an apparatus-independent YSI.

In the present study, an experimental procedure that reproduces the methodology recommended by McEnally and Pfefferle is first introduced. The YSI of PRFs are then measured. The influence of the *n*-heptane and isooctane blending ratio is here assessed for the first time. The methodology is finally extended to quantify in terms of YSI the effect of CO_2 addition to the coflowing air, together with the possible cross effect of PRF formulation and CO_2 addition.

2. Experimental setup

The schematic in Fig. 1 depicts the experimental setup that allows the methodology prescribed by McEnally and Pfefferle [29] to be reproduced.