Experimental assessment of the sudden-reversal of the oxygen dilution effect on soot production in coflow ethylene flames

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ABSTRACT

This paper explores the influence of oxygen dilution on soot production when oxygen is added to the fuel stream of a steady laminar ethylene flame established over the Santoro axisymmetric coflow burner. Interestingly, at an oxygen mole fraction $(X_{O_2}^{ax})_{trans}$ located between 30 and 32%, a transition occurs as the influence of $X_{O_2}^{ax}$ is suddenly reversed. While the peak mean soot volume fraction increases with increasing $X_{O_2}^{ax}$ within the range below $(X_{O_2}^{ax})_{trans}$, it is reduced with increasing $X_{O_2}^{ax}$ beyond $(X_{O_2}^{ax})_{trans}$. To help understand this transition, soot temperature and volume fraction fields are measured by the two-dimensional Modulated Absorption/Emission technique. To assess the sensitivity of the transition conditions, carbon dioxide is added to the coflowing oxidizer stream. Increasing CO_2 mole fraction in the coflow, as replacement of N_2 in the air, significantly mitigates soot formation in the flame but does not influence the transitional oxygen concentration $(X_{O_2}^{ax})_{trans}$ within the resolution of the measurements. Due to the persistence of the transitional oxygen concentration $(X_{O_2}^{ax})_{trans}$ over a wide range of CO_2 replacement of N_2 in the coflowing oxidizer stream, it can be considered a distinct characteristic for the assessment of numerical simulations incorporating soot formation and oxidation models. As an original database, the concomitantly measured soot temperature and volume fraction distributions are attached to the present paper as supplemental materials, thus documenting the aforementioned transitions for the whole range of CO_2 content of the coflow investigated.

1. Introduction

Particulate Matter (PM) and especially soot particles released to the atmosphere have been identified as a serious hazard to human health and a significant contributor to global warming [1]. Since soot exhibits a fairly short lifetime in the atmosphere on the order of days to weeks, the mitigation of soot emission has been considered as a near-term climate-change approach to gain time for implementations of long-term strategies [2]. As soot emitted by Internal Combustion (IC) engines contributes significantly to PM pollution in the air, innovative technologies especially designed for the reduction of soot emission have been a hot topic in IC engine related research in the last few decades [3–5].

The Flue Gas Recirculation (FGR) and Exhaust Gas Recirculation (EGR) are considered attractive technologies that are widely applied in industrial furnaces and IC engines to reduce the emission of pollutants. These strategies are also shown to mitigate soot release due to the reduction of both peak temperature and ignition delay for high levels of EGR dilution in Diesel engines [4]. Fundamental processes underlying FGR or EGR strategies consist of recycling part of the combustion products, either on dry or wet basis, to the fuel and/or the oxidizer of the reacting flows. Other soot emission control strategies are to dilute the fuel stream with a certain gaseous species, which is not necessarily a major component of the combustion product. As a result, the effects of such an addition on the sooting tendencies must be carefully assessed to gain a comprehensive understanding of the additive. In

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particular, an additive generally affects soot formation chemically and it is paramount to fully understand its chemical effects for the purpose of mitigating soot production.

Laminar diffusion flames provide relevant conditions to investigation soot related processes, since fuel pyrolysis, soot inception, growth, and oxidation can be readily identified along the flame height. Additionally, laminar diffusion flames represent a configuration that is relevant to the assessment of sooting tendencies, as non-premixed combustion is frequently encountered in various practical combustion devices, e.g., Direct Injection (DI) engines that lead to weakly premixed combustion [6]. Therefore, studies on the sooting tendencies exhibited by laminar diffusion flames are a necessary step towards the understanding and the eventual the control of soot emissions from practical combustion devices.

As discussed by Du et al. [7] and Liu et al. [8], the reduction of soot formation resulting from the introduction of inert gaseous additives, such as N_2 or Ar, can be attributed to two main effects:

(1) a dilution effect because of the reduction in the concentrations of the reactive species;
(2) a thermal effect due to the change in flame temperature as a consequence of the streams' physical properties variation.

When a reactive species, such as CO_2 or O_2, is added, chemical effects also take place affecting soot formation due to the direct participation of the additive in some chemical pathways of soot formation and/or oxidation. It is worth mentioning that the flame temperature changes due to the additive chemical participation are considered as part of the chemical effects.

Though complete isolation of the chemical effect of an additive from the other two effects is impossible, several numerical and experimental studies were conducted to quantify the extent of the chemical effect [8,9]. As an illustration, Liu et al. [8] numerically introduced a fictitious CO_2 that has exactly the same thermochemical and transport properties as real CO_2 but is not allowed to contribute to the chemical reactions. This strategy allows the CO_2 chemical effect to be isolated and assessed. The authors concluded that the chemical effects of CO_2 addition are significantly higher when CO_2 is added to the oxidizer stream where it both inhibits the formation of acetylene during the fuel pyrolysis and reduces globally the flame temperature. Gülder [9] experimentally preheated the reactants to keep the theoretical adiabatic temperature constant within the whole range of O_2 and N_2 dilution levels, that is, to compensate for the thermal effect. These authors could then evaluate the magnitude of the O_2 chemical effects from the discrepancies between the levels of soot produced for O_2 and N_2 dilutions.

O_2 dilution has been largely documented in the literature dealing with laminar diffusion flames. Nevertheless, controversial results are found, since the oxygen influence seems to be altered by the fuel category and the doped side, i.e., the fuel stream or the oxidizer one. Through doping the oxidizer stream with oxygen mole fraction ranging between 9 and 50%, Glassman and Yaccarino [10] found that the sooting tendency reached a minimum in their axisymmetric ethylene flame when oxygen mole fraction is around 24%. The authors argued that increasing the oxygen mole fraction first tends to enhance soot oxidation. Concomitantly, the peak temperature is also increased, which promotes fuel pyrolysis. At some point, the latter effect prevails, leading to the reversal of the oxygen dilution effect. Zelepouga et al. [11] investigated soot formation for three oxygen mole fractions (35, 50, and 100%) of the oxidizer stream in methane laminar diffusion flames, and showed that soot concentration decreased monotonically with increasing O_2 mole fraction. These authors argued that the residence time for soot particle growth is reduced as the flame length becomes smaller at higher O_2 mole fraction. Lee et al. [12] also stud-

ied the effects of O_2 addition to the coflow in laminar methane diffusion flames and found a larger soot reduction for 100% oxygen. On the contrary, in a methane counterflow configuration, increasing the oxygen content of the oxidizer stream can enhance soot formation [13].

For the purpose of studying the effect of adding oxygen to the fuel stream of a laminar coflow diffusion flame, methane, ethylene, and propane have commonly been used as the fuel and different soot formation behaviors were reported [7,9,14–18]. Gülder [9] investigated the effects of adding oxygen and nitrogen to three fuels (methane, propane, and n-butane). These authors isolated the chemical effect from the other two (thermal and concentration dilution) experimentally. For the methane flame, oxygen addition to fuel chemically suppressed soot formation. On the opposite, oxygen addition to fuel enhanced soot formation in the propane and n-butane flames. The authors explained that the unique sooting behavior exhibited by methane is due to the reduction in acetylene concentration among the pyrolysis products as the oxygen mole fraction in the methane stream is increased. Interestingly, addition of oxygen to fuel in ethylene diffusion flames exhibits the features of these opposite trends: soot formation is first significantly promoted when increasing the oxygen mole fraction in the fuel stream, then soot formation is abruptly reduced above a critical oxygen mole fraction of approximately 32% [7,14,15]. Different explanations of this sudden trend reversal were proposed. Hura and Glassman [15] suggested that the addition of a small amount of oxygen accelerates the rates of the radical formation and fuel pyrolysis. These authors speculated that the initial pyrolysis of the fuel is the crucial step in soot formation. Wright [16] and Wey et al. [17] argued that the role of oxygen in promoting soot formation is due to a catalytic process rather than a thermal decomposition of the fuel as a result of the increase in temperature. Recently, in their study of soot formation in partially premixed ethylene flames, McEnally et al. [18] indicated that the increase in soot production can actually be attributed to different soot formation pathways whose magnitudes depend on the ratios of the pyrolysis products produced, but not the overall pyrolysis rate itself. The sharp decrease in soot load can then be due to the formation of double flames, i.e., an inner premixed flame and an outer diffusion flame. The changes in the hydrodynamic and thermal structures of the whole flame lead to an enhancement of soot oxidation [15]. Surprisingly, in the case of oxygen addition to a propane flame, a first decrease followed by an increase of the soot formation at oxygen mole fraction of 50% was reported by these authors [15], which was later confirmed by Liu et al. [14].

Although the aforementioned studies assessed global trends of soot promotion/suppression caused by oxygen addition, the mechanisms of oxygen addition to fuel on soot formation promotion/suppression remain ambiguous. To further address the sudden-reversal behavior in soot production of oxygen addition to fuel stream in ethylene diffusion flames, the two-dimensional soot temperature and volume fraction fields are measured in the present paper using the Modulated Absorption/Emission (MAE) technique [19]. The laminar ethylene diffusion flames investigated in this study are established over the Santoro burner for oxygen mole fraction in the ethylene stream $X_{O_2}^{ax}$ ranging from 0 (pure ethylene) to 32% while keep the ethylene flow rate constant. As an original contribution, the sensitivity of the transitional oxygen concentration $(X_{O_2}^{ax})_{trans}$, at which the sudden-reversal of oxygen addition to soot formation occurs, to the coflow oxidizer stream composition is assessed by replacing different amounts of nitrogen in the air with carbon dioxide. The measured soot temperature and volume fraction distributions obtained in this study are expected to constitute a valuable database for the validation of numerical models for soot formation and oxidation.