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Soot production modeling in a laminar coflow ethylene diffusion flame at different Oxygen Indices using a PAH-based sectional model

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ABSTRACT

A numerical study is carried out to, first of all, investigate the capability of a Polycyclic Aromatic Hydrocarbon (PAH)-based sectional particle dynamics soot model in the prediction of soot production in laminar coflow ethylene diffusion flames. The effects of different oxygen mole fractions in the oxidizer stream, called Oxygen Index (OI), ranging from 17% to oxygen-enriched conditions up to 33% are investigated. Secondly, the relative importance of species responsible for the increase in both soot formation and oxidation rates with increasing the OI is analyzed. The soot production model considers a detailed description of nucleation via collisions among heavy PAHs, particle aggregation, PAH condensation, surface growth and oxidation through the hydrogen abstraction acetylene addition (HACA) mechanism, and fragmentation of soot aggregates. Model predictions are compared with previously-published experimental data and numerical predictions obtained with a semi-empirical acetylene-based soot production model [Comb. Flame 160: 786–795 (2013)]. Results indicate that the flame structure, soot volume fraction and flame cross-section integrated soot volume fraction predicted by the PAH-based sectional soot model are in good agreement with the experimental data over the entire range of OI considered. The temperature and the concentrations of soot precursors increase with the OI, leading to higher soot nucleation, condensation, surface growth, and oxidation rates. Results show that the PAH-based sectional soot model represents a significant improvement over the semi-empirical acetylene based two-equation soot model studied earlier, especially in conditions far from the normal air conditions (21% of O_2). With increasing the OI, the non-dimensional zone of influence does not change for the flame cross-section integrated soot formation rates but increases for the integrated soot oxidation rate. Nucleation occurs just above the burner rim and displays the largest increase in its rate with increasing the OI. Nucleation and condensation are mainly dependent on the concentrations of BGHIF and BAPYR. Condensation is also affected by the increase in the number of aggregates available for collision. Soot formation is mainly dominated by surface growth, through HACA, with a significant influence of soot condensation closer to the burner surface, regardless of the level of OI. The surface growth through HACA is mainly controlled not only by the acetylene concentration and the associated kinetic rate, but also by the specific soot surface area. Oxidation by O_2 is found to dominate at the top of the flame, while oxidation by OH is dominant at the middle height of the flame. The oxidation rate by O_2 increases more rapidly with increasing the OI than that by OH.

1. Introduction

An important side product of the incomplete combustion of hydrocarbon fuels is soot. It presents in the form of solid carbon particles, normally in the range of a few nanometers in the form of up to around one micron forming complex fractal-like aggregates. Even though the emission of soot particles to the atmosphere is highly unwanted, given its large warming influence on global climate and being a serious hazard to human health [1], soot enhances significantly the radiative energy transfer from sooty flames, which has a major impact on industrial applications in terms of their energy recovery, such as in furnaces and boilers. This dependency also affects and even dominates in fire applications [2]. In this case the thermal radiation contributes to the thermal pyrolysis of the surrounding condensed material, determining finally the fire heat release rate and its growth rate. In either case, understanding the soot production process and providing accurate predictions of the soot quantity generated, their evolution and how much is finally emitted is fundamental to design efficient and clean

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combustion systems.

The process of soot formation consists of various physical and chemical processes. Research conducted to date has reached the consensus that these processes include the gas-phase growth of Polycyclic Aromatic Hydrocarbon (PAH), particle nucleation, surface growth via surface reaction and PAH condensation, and surface oxidation. These particles also coagulate, agglomerate into large chains of aggregate, and also fragment during growth and oxidation. All these processes occur essentially simultaneously, typically in a few milliseconds [3], which makes the soot modeling very complex and challenging.

Due to a lack of adequate understanding of these physical and chemical processes involved in soot formation and oxidation, especially the nucleation step from gaseous PAH species to solid particles, it is necessary to simplify these processes to develop tractable yet robust and efficient soot models. A common approximation is to assume that soot nucleation involves only a small number of PAHs that are available in the chosen gas-phase reaction mechanism, though in reality soot nucleation may be caused by a large number of PAHs. A previous study of the effect of Oxygen Index (OI) on soot formation was carried out using a semi-empirical soot formation model [4] based on the assumption that acetylene is the soot nucleation and growth species. In [4] laminar coflow ethylene diffusion flames burning in O_2/N_2 mixtures of different oxygen percentages were studied experimentally and numerically. The variation of oxygen mole fraction in the oxidizer flow gives rise to significant changes in flame height and temperature [5] and strongly affects soot formation and oxidation [6,7], affecting then flame radiation [8].

Based on the results of the previous study, the variation of soot production with OI is affected by two competing factors: the enhanced soot formation rate due to higher temperature on one hand and a decrease in the residence time due to the reduced flame length and increased soot oxidation rate on the other [4,5]. The former factor was found to prevail, hence higher values of the maximum soot volume fraction and the peak integrated soot volume fraction were observed with increasing OI. The increase of the maximum soot volume fraction with increasing OI was found to exhibit two regimes separated by an OI of 25%: a higher increase rate at OI below 25% and a considerably reduced increase rate at higher OI above 25%. Numerical results of the semi-empirical acetylene-based two-equation soot model, which was selected due to its simplicity and its ability to capture the main trends of soot production at different OIs at a reduced computational cost, are in reasonable agreement with the experimental data for OI in the range of 19–35%. However, the rate of increase of the maximum soot volume fraction within increasing OI was substantially overestimated in both the OI regimes [4]. Moreover, the soot production was significantly underestimated by the two-equation soot model at the OI of 17%, further illustrating the limitations of this soot model.

In order to gain better and more comprehensive understanding of how the change in OI affects soot nucleation, surface growth, and oxidation than those obtained in the previous study using the simplified two-equation soot model, the present numerical study simulates the same laminar coflow ethylene diffusion flames at different OIs reported in [4] by employing an advanced PAH-based soot formation model coupled to a sectional model to describe the dynamics of soot particles [9]. The soot model employed in this study has been used with success in simulating soot formation in laminar coflow ethylene diffusion flames in a series of studies [9–16] and represents the state-of-the-art in soot formation modeling. However, to the best knowledge of the authors it has not been applied to simulate soot formation under different OIs. The objective of this work is twofold: (1) to assess the capability of the state-of-the-art chemical mechanism and soot model in predicting the effects of OI on soot production, and (2) to gain comprehensive insights into the effect of OI on the different processes of soot formation and oxidation in laminar coflow ethylene diffusion flames.

2. Numerical model

The overall flame model is composed of the continuity equation, the Navier-Stokes equations in the low Mach number formulation, the conservation equations of gas-phase species, the energy conservation equation, and the transport equations for the number density of primary soot particles and soot aggregates for each of the sections [10,17,18]. The conservation equations are solved in axisymmetric cylindrical coordinates using a standard finite volume method. The correctional diffusion velocities in both the r- and z-directions are used to ensure that the mass fractions of gaseous species and soot sum to unity. The thermophoretic velocities of soot in both the r- and z-directions are taken into account, as well as the interactions between the gas-phase and the soot chemistry. The diffusion terms in the transport equations are discretized by the second-order central difference scheme and the convection terms by the power law scheme [19]. The SIMPLE algorithm [19] is used to treat the pressure and velocity coupling. The transport equations for gas-phase species and number densities of soot particles are solved in a fully coupled fashion at each control volume using a direct solver (Gauss elimination method) to deal with the stiffness of these equations and ensure the convergence process. All other transport equations are solved using the tri-diagonal matrix algorithm (TDMA). The reaction mechanism developed by the DLR group, consisting of 94 species and 719 reactions, is used to model the combustion chemistry [12,20]. This mechanism has been used in several previous studies to model soot formation in flames fueled with C_1-C_2 hydrocarbons [9,12,21]. The mechanism includes the pyrolysis and oxidation of C_1 and C_2 species, linear hydrocarbons up to C_6 species, the formation of benzene and reactions leading to PAHs up to five-rings, as well as the oxidation pathways of the aromatic species. The radiative source term in the energy conservation equation is computed by the discrete ordinate method coupled with a statistical narrow-band correlated-K (SNBCK)-based wide-band model for the radiative properties of CO, CO_2, and H_2O [22]. The spectral absorption coefficient of soot was obtained by Rayleigh's theory for small particles, and the refractive index of soot due to Dalzell and Sarofim [23] as $5.5 f_s/\lambda$, with f_s being the soot volume fraction and λ the center wavelength of each spectral band.

2.1. Soot model

The fixed sectional soot model used in this work has been described extensively in previous studies [9,10,13,24]. Soot aggregates and primary particles in each section are assumed to have identical geometric characteristics: aggregates have a constant fractal dimension of 1.8, which is a typical value for flame generated soot [25], and soot particles have the same size. The soot model solves two transport equations for each section, the first for the soot primary particles number density and the second for the aggregate number density [9,10,13,24]. From these quantities the number of primary particles forming each aggregate can be determined. As in previous works [9,12,24], 35 discrete sections were used in this study to resolve particle mass from nucleation soot to a sufficiently large aggregate, following a geometric progression with a constant spacing factor of 2.35. The number of sections and the spacing factor are large enough to ensure that the mass fraction in the last section is zero. Moreover, Consalvi et al. [26] showed that the numerical results are not sensitive to the number of sections, with differences less than 4% in the results if 12 sections were imposed. The physical and chemical processes considered in the soot model include nucleation, surface condensation, HACA surface growth, oxidation, coagulation, and fragmentation.

The nucleation process, which describes the transformation of gaseous PAH precursor species into nascent solid soot particles, is modeled assuming that soot nucleates through collision and sticking of the assumed precursor PAH species. This conjecture is supported by numerous experimental observations, e.g. [27]. In this study, soot