



Soot modeling of counterflow diffusion flames of ethylene-based binary mixture fuels



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ABSTRACT

A soot model was developed based on the recently proposed PAH growth mechanism for C₁–C₄ gaseous fuels (KAUST PAH Mechanism 2, KM2) that included molecular growth up to coronene (A7) to simulate soot formation in counterflow diffusion flames of ethylene and its binary mixtures with methane, ethane and propane based on the method of moments. The soot model has 36 soot nucleation reactions from 8 PAH molecules including pyrene and larger PAHs. Soot surface growth reactions were based on a modified hydrogen-abstraction-acetylene-addition (HACA) mechanism in which CH₃, C₃H₃ and C₂H radicals were included in the hydrogen abstraction reactions in addition to H atoms. PAH condensation on soot particles was also considered. The experimentally measured profiles of soot volume fraction, number density, and particle size were well captured by the model for the baseline case of ethylene along with the cases involving mixtures of fuels. The simulation results, which were in qualitative agreement with the experimental data in the effects of binary fuel mixing on the sooting structures of the measured flames, showed in particular that 5% addition of propane (ethane) led to an increase in the soot volume fraction of the ethylene flame by 32% (6%), despite the fact that propane and ethane are less sooting fuels than is ethylene, which is in reasonable agreement with experiments of 37% (14%). The model revealed that with 5% addition of methane, there was an increase of 6% in the soot volume fraction. The average soot particle sizes were only minimally influenced while the soot number densities were increased by the fuel mixing. Further analysis of the numerical data indicated that the chemical cross-linking effect between ethylene and the dopant fuels resulted in an increase in PAH formation, which led to higher soot nucleation rates and therefore higher soot number densities. On the other hand, the rates of soot surface growth per unit surface area through the HACA mechanism were similar among the different fuel mixing cases, although slightly lower compared with ethylene base case. The increased soot nucleation rates by fuel mixing and similar soot surface growth rates (per unit surface area) help to explain the increased soot volume fraction/number density and similar average sizes among the tested cases.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) and soot emitted through combustion have adverse health and environmental impacts, such that extensive studies have been directed towards identifying the underlying kinetic mechanisms leading to PAH and soot formation [1–6]. Soot formation processes include nucleation from PAHs, coagulation/agglomeration, and surface reactions [3]. Since nucleation from gas-phase PAHs is the initial step in soot formation, a reliable description of PAH formation is essential in developing soot formation and growth models [7]. In this regard,

the chemistry of PAHs has been studied over the last two decades (see e.g., [2,6] and references therein).

The hydrogen-abstraction-C₂H₂-addition (HACA) mechanism was proposed by Frenklach and coworkers as an important pathway for PAH growth [8,9] with a focus on the role played by acetylene. Reaction channels involving odd carbon species such as propargyl, cyclopentadienyl and indenyl were also identified to contribute to PAH formation [10–18] and a mechanism for C₁–C₂ fuels has been developed [19] and further refined [20] to account for PAH growth up to five-ring aromatics.

Soot modeling studies have been carried out using various approaches [21]. Earlier models were empirical in nature and soot formation was directly linked to the fuel [22], C₂H₂ [23] or the benzene concentration [24]. More recent models considered gaseous

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PAH molecules as soot precursors and treat the inception of soot particles through PAH (typically pyrene) dimerization [7,25,26]. Soot surface growth is mainly through surface HACA mechanism and the particle dynamics has been modeled by the method of moments [27,28]. These models were pioneered by Frenklach and Wang [8] and later used extensively on various types of flames such as laminar premixed flames [7,29–32], laminar counterflow diffusion flames [33–35], axis-symmetric coflow diffusion flames [36], 2D non-premixed turbulent flames [37] and 3D flames in diesel engine combustion [38]. The discrete sectional method was also frequently used [39–46] for particle dynamics, especially focused on soot size distribution. In addition, a simpler two-equation soot model [23,47] (transport equations for soot mass and number density) was also used with a certain level of success in simulating coflow diffusion flames [48,49].

Another group of models treats PAH and soot nuclei formation through chemical growth [50–54] during which high molecular mass species are formed through a radical-molecule sequence of reactions, which eventually reach condensed phase.

Previous modeling studies typically targeted flames of single-component fuels. However, a number of previous experimental studies [49,55–59] on binary fuel mixtures identified synergistic effects on soot formation, which indicates an enhancement in soot production in the flames of mixture fuels as compared to the cases with respective single fuel components. This synergistic effect was attributed to the chemical cross-linking among pyrolysis products of the binary component fuels, e.g., C_2H_2 , CH_2 , and C_3H_3 , in forming incipient rings and subsequent reactions.

To predict such chemical cross-linking effects on soot formation, a reliable gas-phase kinetic mechanism is required, which can predict well the concentrations of soot precursors, i.e. PAH molecules, in the flames of fuel mixtures. In this regard, two PAH mechanisms, namely KAUST PAH Mechanism 1 (KM1) [60] and 2 (KM2) [61] that included molecular growth up to coronene ($C_{24}H_{12}$) were recently proposed for gasoline surrogate fuels and C_1 – C_4 gaseous hydrocarbon fuels, respectively. These mechanisms have been validated in premixed flames for quantitative predictions of PAH concentrations [60,61]. KM1 was tested with counterflow diffusion flames of n-heptane/toluene and iso-octane/toluene mixtures and KM2 was tested with ethylene/propane mixtures. Based on relative intensities of PAHs determined from laser-induced fluorescence (LIF) techniques and computation, the effect of chemical cross-linking of binary fuel mixtures on PAH formation was qualitatively captured [60,61]. As compared with the earlier but widely used ABF PAH mechanism [7], the predictions of pyrene concentrations were improved by orders of magnitude, in agreement with the experimental data [62].

In previous PAH-based soot models, for example in [26,63,64], soot nucleation typically takes place when two pyrene molecules collide. It is expected that only a portion of these collisions could result in successful soot nucleation by forming a dimer while the rest of colliding molecules may simply fall apart due to the high collision energy required or the unfavorable colliding angle. In early pioneering work (e.g. [8]), it was assumed that every collision successfully created a soot nucleus through which reasonable prediction of overall soot loading could be obtained. This agreement, however, could be partially attributed to the fact that the over-estimation of the collision efficiency is counter-balanced by the under-predicted concentration of gas-phase soot incepting PAHs. In addition, the inclusion of only pyrene for nucleation in many existing soot models could also contribute to the under-estimation of the concentrations of PAHs involved in soot nucleation. The exclusion of larger PAHs in most of the existing soot inception models may partly be due to the limitation of the adopted gas-phase kinetic mechanisms, which typically have PAH growth up to 4 or 5 rings. A recent study showed that PAHs with larger sizes

are more likely to stick together [65], leading to higher probability of soot particle inception.

Here, we use a detailed gas-phase mechanism [61] coupled with a new PAH based soot model to predict quantitatively the sooting structures of counterflow diffusion flames of binary fuel mixtures. KM2, with molecular growth up to coronene, was adopted as the gas-phase mechanism. Considering that a quantitative prediction of gas-phase PAH concentrations is essential in soot modeling [7], KM2 was first validated against two counterflow flames [66,67] for which measured PAH concentrations are available. Next, a new soot inception model including soot nucleation from 36 nucleation reactions was coupled with the gas-phase mechanism. A method of moments for soot particle dynamics was applied to several sooting counterflow diffusion flames. A baseline case of pure C_2H_4 fuel and binary mixtures of C_2H_4/C_3H_8 , C_2H_4/C_2H_6 and C_2H_4/CH_4 were simulated. PAHs with sizes between (and including) pyrene and coronene were involved in the soot model for nucleation processes. Soot surface growth reactions included the modified HACA mechanism (described in Section 2) and PAH condensations. The method of moments with interpolative closure was employed for soot modeling [28]. The computed soot volume fraction, average particle size and number density were compared with experimental data for both the pure and binary fuel cases.

2. Numerical simulation

2.1. Gas-phase kinetic mechanism

The PAH growth mechanism of KM2 [61], developed for C_1 – C_4 fuels, has been discussed in detail previously and only a brief summary is provided here. In developing KM2, the reactions for small chemical species up to benzene were adopted from the USC II mechanism [68]. For the growth of aromatic hydrocarbons beyond benzene (and up to coronene), reactions involving odd-carbon species such as propargyl, cyclopentadienyl, benzyl and indenyl radicals along with the HACA mechanism were included. In addition to H atoms, methyl and propargyl radicals were also allowed in the abstraction of H atoms from PAHs to form radicals for their further growth. The mechanism has 202 species and 1351 reactions.

It was shown that [61], as compared to the literature-based PAH mechanisms [7], KM2 provided improved predictions of PAH concentrations in premixed flames [61]. In counterflow diffusion flames of ethylene/propane mixtures, KM2 could also predict the experimentally observed synergistic effect in PAH formation [57]. In the present study, quantitative validation of PAH concentrations in counterflow diffusion flames of ethylene and ethane was first conducted and compared with existing experimental data [66,67], which is detailed in [Supplementary Material 1](#). The simulations were performed using the opposed flow module of the Chemkin Pro package [69]. The criterion for convergence was set at 10^{-10} in terms of absolute tolerance and the grid points were gradually refined until the solution gradient and curvatures were below 0.1 and 0.5, respectively. Mass diffusion caused by a temperature gradient was considered and mixture-averaged diffusion coefficients were used. The capability of KM2 to predict, both qualitatively and quantitatively, the concentrations and behaviors of large PAHs in counterflow diffusion flame environments warrants its use for soot modeling. Note that A1 to A7 were used to denote aromatic species with 1–7 rings, following a previous study [61], where details of the chemical structures can also be found.

2.2. Soot model

The PAH-based soot model used in this study contains four main components: soot nucleation, surface growth through chemical

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