Application of a semi-detailed soot modeling approach for conventional and low temperature diesel combustion – Part I: Model Performance

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Abstract

In the present study a previously developed soot model has been tested extensively for conventional and low temperature diesel combustion engine simulations. The soot model framework, which was implemented in the Computational Fluid Dynamics (CFD) code KIVA-CHEMKIN, is based on four fundamental steps, viz., soot inception through a four ring polycyclic aromatic hydrocarbon species, soot surface growth through acetylene and benzene, soot coagulation, and oxygen and OH-induced soot oxidation. The proposed model can be used not only for soot mass predictions, but also for soot number density and soot particle diameter predictions. Diesel combustion was simulated using reduced n-heptane/primary reference fuel (PRF) chemistry mechanisms and a reduced polycyclic aromatic hydrocarbon (PAH) chemistry mechanism, while the n-heptane chemistry mechanism was used for modeling constant volume n-heptane combustion. Soot model performance was evaluated by comparing the model predictions with available constant volume combustion chamber optical diagnostic experiments, optical engine in-cylinder soot data, and a light-duty single cylinder metal engine-out smoke data. In particular, a variety of combustion regimes were tested, including conventional diesel, premixed charge compression ignition (PCCI), and high exhaust gas recirculation (EGR) low temperature combustion (LTC). In general, the model was able to predict well the trends in soot mass over the range of operating conditions. With increasing degree of premixing, the model was able to predict relatively lower soot concentrations compared to diffusion combustion. In terms of soot particle diameter, the model computed particle size was seen to increase with increasing simulated EGR when the ambient oxygen concentration was varied from 21% to 8% by volume and with increasing in-cylinder density (14.8 kg/m³ vs. 30 kg/m³ at the same in-cylinder O₂ level)

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under steady-state constant volume combustion conditions. It is also observed from the optical and metal engine studies that the soot particle size is larger for high temperature combustion conditions that are marked with high soot formation rates during the combustion process and may or may not lead to smaller particles at exhaust valve opening (EVO) depending on the oxidation rates. The soot size for LTC is generally lower through-out the combustion process including EVO conditions. All but one data-point show uni-modal type soot particle size distribution with respect to both soot mass and particle number.

Keywords

CFD; Soot model; Polycyclic aromatic hydrocarbon (PAH) chemistry; Low temperature combustion; Combustion chemistry; PRF mechanism

1. Introduction

Increasingly stringent pollution mandates have necessitated further study of gasoline and diesel combustion fundamentals. Particularly, mitigating emissions of NO_x and soot in diesel engines has been a long-standing challenge for the internal combustion engine community. After-treatment devices such as selective catalytic reduction (SCRs), NO_x absorbers and diesel particulate filters (DPFs) are actively being employed to control regulated pollutant emissions, but the conversion efficiency, durability, backpressure, and the need for thermal management strategies are some of the challenges posed by these devices. Also, with recent emphasis on corporate average fuel economy (CAFE) and greenhouse gas (GHG) standards, the spotlight in engine research has lately transitioned from emissions to fuel economy. Hence there exists a need to probe alternative combustion strategies, not only to reduce emissions within the engine combustion chamber, but also from an efficiency standpoint. These advanced combustion strategies are significantly different than conventional combustion strategies for both spark-ignited (SI) and compression-ignited (CI) engines. Particularly for diesel engines, strategies such as homogeneous charge compression ignition (HCCI), achieved by significantly early injections, premixed charge compression ignition (PCCI), possibly with two injections (one early and one conventional), and low temperature combustion (LTC) with heavy amounts of exhaust gas recirculation (EGR) are being explored [1,2]. Figure 1, reproduced from Akihama et al. [1] and modified here (shown in bold text) for illustration purposes, shows the combustion regimes in the classic equivalence ratio-temperature (Φ -T) map. The modifications were based on the generic Φ -T plot

shown in Tree and Svensson [2], which are derived from the experimental observations of Pickett and Siebers [3] in a constant volume spray vessel. These regimes of combustion are very different compared to conventional diesel diffusion combustion, which significantly alters the mechanisms of pollutant formation processes. As seen in Figure 1, the drive has been to operate the engine either between the soot and NO_x peninsulas or at much lower temperatures, which provides a wider operating range and hence is desirable. Soot formation can be avoided either by operating at lower temperatures (<1800 K) or higher temperatures (> 2600K) or lower local equivalence ratios (< 2), but lower NO_x warrants lower in-cylinder temperatures. Points 1 (a) and 1 (b) refer to high EGR LTC (~ 10% ambient O₂) with a small nozzle diameter (~50 μ) for enhanced fuel-air mixing. The fuel jet core is represented by point 1a ($\Phi > 1$ and low temperature) while the flame sheet is represented by 1b ($\Phi \sim 1$ and high temperature). As the nozzle diameter is small, which enhances atomization, the entrained air amount is higher, which inhibits soot formation in the fuel jet core. In the flame sheath region the high EGR helps to keep the temperatures low and hence lower in-cylinder NO_x is achieved. Point 2 corresponds to an ambient temperature of 850 K with higher oxygen content (21%O₂) and also using small nozzle diameters (~50 μ). This corresponds to low temperature lean combustion. PCCI falls closer to point 2, which is generally accomplished with multiple injections where the overall equivalence ratio could be much less than local equivalence ratios. The ordinate of the point 'PCCI' in the figure represents the local equivalence ratio. Point 3 refers to high EGR operation (~8% ambient O₂) with a production feasible nozzle diameter (~180 μ) while operating with an oxygenated fuel, which produces the same result of low NO_x and low soot.

This discussion indicates that there are multiple ways of obtaining low NO_x and low soot, at least from a scientific standpoint, and the current techniques rely on longer ignition delay combustion strategies to allow more time for fuel-air mixing. From a modeling perspective, predicting emissions for low NO_x-low soot combustion necessitates less empirical and more physically driven pollutant models that can reasonably replicate the formation and oxidation kinetics over a wide range of operation. Particularly, soot modeling for conventional diesel combustion has been a long standing research subject and, with newer combustion regimes, soot models are further challenged. Also, to prevent the possibility of allowing ultra-fine particles from escaping into the atmosphere, future US, Japan and European standards aim at mandating particulate

number (PN) (Johnson [4]), and thus model predictions of engine-out soot particle size and number density along with soot mass have gained increasing importance.

Finally, the last piece of the modeling puzzle is the fuel itself. Due to limited resources of fossil fuels and with the growing momentum for their conservation, dual fuel combustion such as pilot diesel ignited natural gas, reactivity controlled compression ignition (RCCI) [5], and use of alternative fuels such as E85 (85% by volume of ethanol and 15% by volume of gasoline) and biodiesel are also being explored for diesel and gasoline engines. The pathways of soot formation differ from one fuel to another depending upon the fuel composition. Regular pump gasoline or diesel may have anywhere between 20%-32% aromatics by volume, which are also major contributors to soot formation, in addition to soot formation due to fuel chain breakdown. However, fuels such as biodiesel (mainly composed of fatty acid esters) and natural gas (mainly composed of methane) do not have fuel aromatics and hence the soot formation is directly a function of the fuel breakdown chemistry. Thus, newer emission and fuel efficiency standards along with the continued focus on exhaust particulate reduction motivate the drive toward newer in-cylinder engine combustion strategies and alternative fuels. Consequently, the modeling thrust is to reduce empirical approaches and replace them with more physics-based detailed and versatile combustion and emission models.

In the present work, the soot model of Ref. [8] implemented in the 3-D CFD code KIVA-CHEMKIN was applied to study advanced diesel combustion modes, including conventional diesel combustion, high-EGR diesel LTC and diesel PCCI. Also, the model predictions of soot mass were compared with available experimental data and model predictions of soot diameter were qualitatively evaluated for each combustion mode. The computational mesh, spray, turbulence, and other sub-models are the same as used in Vishwanathan and Reitz [8].

2. Model framework

2.1 Chemistry mechanisms

2.1.1 Fuel and lower carbon chemistry

Two separate chemistry mechanisms were used in the present study. The first mechanism (Mech-1) comprised 37 species and 180 reactions, and is an n-heptane mechanism that was applied for constant volume n-heptane combustion. The second mechanism is a reduced primary reference fuel (PRF) mechanism (Ra and Reitz [7]), which was found to be more suitable for diesel engine combustion simulations (Mech-2). Mech-2 comprised 40 species and 280 reactions. Both these mechanisms were previously used by Vishwanathan and Reitz [8]. Throughout this work diesel was modeled with the fuel properties of tetradecane ($C_{14}H_{30}$) [8].

2.1.2 PAH chemistry using Xi and Zhong mechanism

The reduced PAH mechanism was obtained from Xi and Zhong [9] and integrated with the above two mechanisms. The first aromatic ring formation, i.e., A_1 takes place through the propargyl radical (C_3H_3). Higher PAHs are formed through the hydrogen-abstraction and carbon-addition (HACA) mechanism (Frenklach and Wang [10]). The nomenclature adopted for representing PAHs (e.g., A_1 = benzene, A_2 = naphthalene, A_3 = Phenanthrene, and A_4 = Pyrene) is common in the literature, where A denotes a mono- or poly-aromatic species and the suffix denotes the number of fused aromatic rings. This PAH mechanism (PAH-1) was comprised 21 species and 52 reactions.

2.2 Soot Model

The soot model was implemented in the 3-D CFD code KIVA-CHEMKIN [6] and published by Vishwanathan and Reitz [8] and Vishwanathan [11]. The present modeling framework is summarized as follows:

a) Soot inception through A_4 :

$[A_4]$ represents the concentration of A_4 in mol cm^{-3} . $[]$ indicates concentration of species.

b) C_2H_2 assisted surface growth:

$$\begin{aligned} s + C_2H_2 &\xrightarrow{R_2} 2C(s) + H_2, R_2 = k_2 \cdot C_2H_2, \\ k_2 &= 9.0 \cdot 10^4 \cdot \exp(-12100/T) \text{ s}^{-1}, S = \pi d_p^2 \cdot N \text{ cm}^{-1}, d_p = 6 Y_{c(s)} \rho_{c(s)} N^{1/3} \text{ cm} \end{aligned} \quad (2)$$

S , is the soot surface area per unit volume, which can be obtained from the particle size, d_p , $Y_{c(s)}$ is the soot mass fraction, ρ is the ambient density, $\rho_{c(s)}$ is the soot density for which the density of graphite (2.0 g cm^{-3}) is used in this study, and N is the soot number density in particles cm^{-3} . For each computational cell, the model would predict a unique value of soot species density and soot number density, and hence a soot diameter for each cell. This diameter can be viewed as a local number-averaged soot particle size. Hence, the model is locally mono-disperse (on a computational cell basis) but the diameter can vary from one computational cell to another.

c) Soot coagulation:

$$nC(s) \xrightarrow{\dot{R}_3} \Theta(s)_n, \dot{R}_3 = K(T) \cdot \left(\frac{\rho Y_{c(s)}}{M_{c(s)}} \right)^{\frac{11}{6}} \cdot [N]^{\frac{11}{6}}, K(T) = 2C_a \left(\frac{6M_{c(s)}}{\pi \rho_{c(s)}} \right)^{\frac{1}{6}} \cdot \left(\frac{6K_{bc} T}{\rho_{c(s)}} \right)^{\frac{1}{2}} \left\{ \frac{\text{particles}}{\text{cm}^3} \right\} \quad (3)$$

where, $M_{c(s)}$ is the molecular weight of carbon (12.0 g mol^{-1}), T is the temperature of the CFD cell and K_{bc} is Boltzmann's constant ($1.38054 \cdot 10^{-16} \text{ erg K}^{-1}$). The constant C_a is the agglomeration constant for which a value of 9.0 was used in this study. The particle size d_p is the effective particle diameter accounting for soot coagulation and is termed the 'spherical agglomerate size'. The primary particle size can be obtained by equating the coagulation term to zero.

d) Soot oxidation through Nagle-Strickland and Constable model [12]:

$$C(s) + \frac{1}{2} O_2 \xrightarrow{\dot{R}_4} CO, \dot{R}_4 = \frac{12}{M_{c(s)}} \cdot \left(\left(\frac{K_A P_{O_2}}{1 + K_Z P_{O_2}} \right) \cdot x + K_B P_{O_2} \cdot (1-x) \right) \cdot S \text{ {mol cm}^{-3} s^{-1}} \quad (4)$$

$$\left. \begin{aligned} x &= P_{O_2} / (P_{O_2} + (K_T/K_B)), K_A = 30.0 \cdot \exp(-15800/T) \text{ {g} \over {cm}^2 \text{ s atm}}, K_B = 8.0 \cdot 10^{-3} \cdot \exp(-7640/T) \text{ {g} \over {cm}^2 \text{ s atm}}, \\ K_T &= 1.51 \cdot 10^5 \cdot \exp(-49800/T) \text{ {g} \over {cm}^2 \text{ s}}, K_Z = 27.0 \cdot \exp(3000/T) \text{ {atm}^{-1}} \end{aligned} \right\} \quad (5)$$

Equation (5) gives the net-rate of surface oxidation at the more reactive A sites and the less reactive B sites. The fraction of A sites is given by the term x in Eq. (4) and the fraction of B sites by (1-x). P_{O_2} (atm) is the partial pressure of oxygen in the CFD cell.

e) Soot oxidation through OH radical:

Soot oxidation via the OH radical was considered through the modified Fenimore and Jones [13] mechanism step.

In the above expression, X_{OH} is the OH mole fraction and γ_{OH} is the OH collision efficiency (0.13 in this study).

f) PAH assisted surface growth /PAH coagulation

Di Domenico et al. [14] used an approach for considering PAH-assisted growth on soot, originally developed by Frenklach and Wang [10] for PAH-PAH collision (dimerization or coagulation) leading to soot inception.

where, PAH_{kj} is a PAH species with k carbon and j hydrogen atoms. γ_{ks} is the PAH-soot collision efficiency (0.3 in this study) and β_{ks} is the frequency of PAH-soot collisions, the expression for which is,

$$\beta_{ks} = 2.2 \cdot \sqrt{\frac{\pi \cdot K_{bc} \cdot T}{2\mu_{ij}}} \cdot (d_p + d_{PAH})^2 \{cm^3 s^{-1}\} \quad (8)$$

where d_{PAH} is the PAH collision diameter and μ_{ij} is the reduced mass. Frenklach and Wang [10] used the PAH collision diameter,

$$d_{PAH} = d_A \cdot \sqrt{\frac{2m_i}{3}} \quad (9)$$

where d_A is the size of a single aromatic ring the value for which is $1.395 \cdot 10^{-8} \cdot \sqrt{3} \{cm\}$ and m_i is the mass of aromatic ring class, expressed in terms of number of carbon atoms in the aromatic. μ_{ij} is the reduced mass and in this case, it is taken as the mass of the colliding gaseous PAH species. As the original model was used with the method of moments, the above implementation is simplified as a representative rate process for soot surface growth through aromatics. In the present study only A_1 (generally the highest concentration species among the aromatics and PAH), resulting through the C_3/C_4 pathway, was considered for the growth process. However, it was found that it did not have a pronounced effect on the net soot mass.

g) Transport equations for soot species and number densities:

$$\frac{\partial M\mu}{\partial t} = -\nabla \cdot (M\mu \vec{v}) + \nabla \cdot \left(\frac{\mu}{SC\rho} \nabla \left(\frac{T}{T_0} \right) \right) + \dot{M} \quad \text{for } \mu = 0.75 \cdot \left(1 + \frac{\nabla \cdot \vec{v}}{T} \right) \quad \text{and } \eta_M = 0.9 \quad / \quad \frac{\pi\eta}{8} \quad (10)$$

The above equation represents two additional transport equations that were solved in the CFD code. M stands for either soot species density, $\rho Y_{C(s)}$, in $g cm^{-3}$ or soot number density, N , in $particles cm^{-3}$, v (cm/s) is the CFD gas-phase velocity, SC is the Schmidt number, μ is the computational cell viscosity, T (K) is the cell temperature, and $\dot{M}$ represents source terms. The second term inside the bracket on the right hand side of Eq. (10) is the thermophoretic diffusion term which was modeled as a source term in KIVA. The accommodation factor $\eta = 0.9$. The source terms for the respective equations are,

$$\left. \begin{aligned} SpYC_s &= 16R_1 + 2R_2 + 6R_6 - R_4 - R_5 - MCsgcm3s, \\ SN &= 16R_1 \cdot MCsMnuci - R_3 \text{ particles}cm3s, \end{aligned} \right\}$$

$$M_{nuci} = \pi \cdot d_{nuci}^3 \cdot \rho_C(s) \quad (11)$$

M_{nuci} is the mass of the soot inception particle nucleus, assumed to be of diameter $d_{nuci} = 1.28$ nm (approximately equivalent to 100 carbon atoms). More specific details of the model can be found in Vishwanathan and Reitz [8] and in Vishwanathan [11].

Recently, numerous studies have used this soot model with different PAH chemistry mechanisms for a variety of fuels for diesel combustion (Sukumaran et al. [15]), n-heptane/toluene combustion (Reitz et al. [16]), oxygenated fuel n-heptane-n-butanol combustion (Wang et al. [17]), and dual-fuel gasoline-diesel combustion (Yang et al. [18]). However, most of these studies focused on PAH chemistry improvements for different fuels and soot mass predictions. The current study focuses on soot number density and soot diameter predictions along with soot mass predictions. The model predicted soot mass was compared with various experimental results, and the associated discrepancies have been documented in detail elsewhere [8]. Also, Part-II of the present paper details some of the model prediction discrepancies using sensitivity analysis. In addition, the chemistry mechanisms were tested for predicting 1) Ignition delay times and compared with experimental shock-tube ignition delay data 2) PAH profiles in 0-D perfectly stirred reactors and in the Two-Stage Lagrangian (TSL) model framework against detailed chemistry mechanisms, and finally, 3) n-heptane lift-off lengths were compared against SANDIA constant volume experimental spray data. All this information was well documented in Ref. [8] and has been omitted here to avoid redundancy.

3. Results and discussion

2.1 Spray chamber results

Comparisons were made with Sandia constant volume spray chamber experimental data using a single-hole nozzle (<http://www.ca.Sandia.gov/ecn/cvdata/frameset.html>). Lift-off lengths, total soot mass, and 2-D soot distributions were compared. The lift-off length was defined as the distance from the tip of the injector to the axial location where significant OH radical formation was seen, consistent with Vishwanathan and Reitz [8]. The experimental lift-off lengths were obtained by OH chemiluminescence in the experiments of Pickett and Idicheria [19]. The lift-off length was averaged during the quasi-steady period of combustion

for all cases, i.e., from 3.5 ms to 6.5 ms after start of injection (SOI), and the injection duration was 6.8 ms. Predictions of time-averaged soot mass over the quasi-steady combustion period were also compared to the time-averaged experimental soot mass (estimated from soot extinction and laser induced incandescence (LII)), as shown in Figure 2 for the conditions in Table 1. With the Mech-1 chemistry mechanism, it is seen that at lower density ($\rho = 14.8 \text{ kg/m}^3$), the model does a very good job of predicting the total soot mass in the jet. For higher density ($\rho = 30.0 \text{ kg/m}^3$), and hence for higher in-cylinder pressures, the model captures the effect of pressure dependence on soot formation by predicting higher soot formation, however the total soot mass at moderate O_2 concentration conditions is somewhat under-predicted and over-predicted at low O_2 conditions. Mech-2 consistently predicts more soot than Mech-1 for all the conditions. Also, the predictions of soot mass using Mech-2 are closer to the experiments than Mech-1 for the high density conditions. Mech-1 and Mech-2 chemistry mechanisms are fundamentally different and were reduced and tuned for replicating n-heptane and diesel combustion, respectively. The low temperature reaction rates involving ketohydroperoxyl radical, C_2H_4 and C_2H_2 chemistry are different between the two mechanisms. Particularly, Mech-2 reaction rates were tuned to mimic diesel soot formation and Mech-1 rates were tuned for n-heptane soot formation, and thus, soot formation increased with the Mech-2 chemistry mechanism, where the predominant contribution is from C_2 reactions. Since C_2H_2 contributes to soot surface growth and predominant soot mass, variation in reaction rates of these reactions can cause different soot mass predictions by different mechanisms. Part-II of the paper gives a detailed sensitivity analysis of the reactions and their effect on soot mass predictions

It is also interesting to note that both capture the so-called ‘soot-bump’, i.e., the soot peak at intermediate O_2 concentrations. This peak occurs due to the competition between increased residence time (longer lift-off length) due to lower oxygen entrainment rates, and lower local temperatures for soot formation and oxidation, with lower O_2 concentrations. Also, it is seen from Figure 2 that in terms of the present model implementation, the contribution of PAH induced surface growth is very much less, as seen by the inverted triangles at the 21% O_2 high temperature combustion and at the 8% O_2 LTC condition. Although not noticeable from the figure, the total soot mass as predicted by the model without PAH surface growth for the 21% O_2 condition was about $12 \mu\text{g}$ while it was about $14 \mu\text{g}$ considering PAH surface growth. However, for the LTC condition, the difference is insignificant. As previously mentioned, this sub-model

may have a greater contribution when fuel aromatics are included in a fuel surrogate. All the results presented here consider the PAH induced surface growth sub-model.

Figures 3(a), 3(b) and 3(c) show the steady-state (6.5 ms after SOI) soot particle diameter, d_p vs. number of particles and d_p vs. percent soot mass for 21% O_2 - $\rho = 14.8 \text{ kg/m}^3$, 10% O_2 - $\rho = 14.8 \text{ kg/m}^3$ and 10% O_2 - $\rho = 30 \text{ kg/m}^3$, respectively. The d_p vs. particle number histograms were obtained by grouping all the computational cells with soot diameters in bins. Finally, the total number of particles for each bin was obtained by summing over the number of particles in all the cells belonging to the corresponding bin (i.e., cells within the d_p range). For each cell, the number of particles was obtained from the product of the cell soot number density and the cell volume. The contribution of each bin to the percent soot mass was also obtained similarly. The model predicts that most of the particles, in terms of both mass and number, are around 10-20 nm for the 21% O_2 , $\rho = 14.8 \text{ kg/m}^3$ condition. For the 10% O_2 , $\rho = 14.8 \text{ kg/m}^3$ case (higher simulated EGR), it is seen that most of the particles under steady state are between 20 and 30 nm. Finally, for the 10% O_2 , $\rho = 30 \text{ kg/m}^3$ high sooting condition the sizeable contribution to the mass and particle number is from particles with sizes 20-40 nm with some contribution also from the 40-50 nm size range. In general, it is seen that when the ambient oxygen is reduced (from 21% O_2 to 10% O_2) at the same in-cylinder density, or when the in-cylinder density is increased at the same in-cylinder ambient oxygen level (from 14.8 kg/m^3 to 30 kg/m^3) the particle size increases.

Table 2 shows the constant volume combustion chamber experimental conditions for diesel fuel combustion with different injection pressures and nozzle diameters obtained from the Sandia database (<http://www.sandia.gov/ecn/cvdata/dsearch/frameset.php>). The simulations were performed with the Mech-2 + PAH-1 chemistry mechanism with tetradecane assumed for the fuel physical properties. Figure 4 and Figure 5 show the steady-state soot volume fraction distributions at four different injection pressures and with four different nozzle diameters, respectively, with the dashed lines representing the lift-off lengths. As can be seen, the model is able to predict higher net soot at lower injection pressures and with larger nozzle diameters, in agreement with the experimental trends.

In general, the peak values of the model predicted volume fraction are modestly under-predicted as compared to the experiments, except for the no-sooting 50 μ diameter case. In general, the model predicted width of the soot zones is smaller than the experimental soot zones. This discrepancy may be due to the

radially-narrower rich cores predicted by the model when compared to the experiments, or due to enhanced soot oxidation by the peripheral OH in the flame zone, or both. For the 50 μ diameter case, even though the model shows a soot amount peak of 2 ppm, the total amount of soot per unit injected mass is very low, as seen in Figure 6. Figure 6 also shows that the effect of nozzle diameter is more prominent in soot reduction due to enhanced air entrainment as compared to increasing the fuel injection pressure. Figure 6 also shows that with increasing nozzle size, the soot core increases due to the wider rich zones caused by relatively poorer entrainment.

Figures 7(a) and 7(b) show the d_p vs. soot mass histograms as a function of injection pressure and nozzle diameter, respectively, with 1nm bins just before the end of injection (~ 6.8 ms after SOI). For relatively low-sooting conditions either with higher injection pressures or smaller nozzle-hole diameters, there is a relatively narrow normal distribution of the soot size with respect to the soot mass contribution and there is a distinct diameter with maximum soot mass contribution. For lower injection pressures and larger nozzle holes, the contribution from the predominant diameter is reduced and the distribution is wider. In general, the soot size predictions are unimodal and are also higher for high-sooting conditions as compared to low-sooting conditions due to higher soot inception and surface-growth rates in the soot core region.

2.2 Optical engine results

The performance of the soot model was also evaluated for in-cylinder soot formation and oxidation in the Sandia Cummins N-14 series optical engine with #2 diesel fuel. Four different engine combustion conditions, two high temperature conditions (premixed combustion and conventional diesel diffusion combustion high temperature conditions (HTC)) and two LTC conditions (early injection LTC and late injection LTC) were studied by Singh et al. [20]. The boundary conditions for the numerical simulations are shown in Table 3. Figure 8 shows the in-cylinder pressure and heat release rate (HRR) predicted by the Mech-2 + PAH-1 mechanism as compared to the experiments for all four conditions. It is seen that, in general, the model is able to predict the in-cylinder pressure and heat release history well for the different combustion regimes. The model predicted HRR is the chemical heat release (exclusive of wall heat transfer) and hence is expected to be somewhat higher than the experimental HRR.

Singh et al. [20] used the two-color technique to monitor the in-cylinder soot evolution in the experiments. Two laser wavelengths were used to obtain the soot path length ($f_v L$) and temperature, and the flame area (A_{flame}) could be inferred from the images only in the piston-bowl region, finally yielding an expression for soot mass as:

$$\text{Soot mass } \{\text{g/kg-f}\} = \frac{(f_v L) \rho(A_{\text{flame}}) \cdot C(s)}{\text{Injected fuel mass}} \quad (12)$$

The model predicted soot volume fraction (f_v) and cell temperatures were used to infer the intensity of radiation, using the two-color equations (Singh et al. [21]). Finally, A_{flame} was obtained using the projected computational cell area in the piston bowl with intensity greater than a 15% threshold, as used in the experiments and the soot mass calculated using Equation (12) was compared to the experimental soot mass.

Figure 9 shows the in-cylinder soot history for the four combustion cases. The model is seen to predict the soot formation and oxidation for all the cases quantitatively, except the premixed HTC case. Also, for the HTC diffusion case, a second bump is seen in the experiments that is not predicted by the model. This could be caused by faster soot oxidation predicted by the model. Another reason could be the uncertainty in soot mass obtained by the two-color method, as the two-color procedure is a line-of-sight technique, where the detector signals are biased towards the hottest soot in the combustion chamber [8]. However, the model was able to predict lower soot in going from HTC-diffusion to HTC-premixed combustion, as expected. The model also predicted the large decrease in soot from HTC to LTC, as seen by comparing the peak soot values. Some of the discrepancies may be due to the lack of fuel-aromatic pathways to soot contributions, particularly at LTC conditions. According to Heywood [22], the mechanism of soot formation at higher temperatures (>1900 K) may be dominated by linear unsaturated hydrocarbons like C_2H_2 due to the faster fuel decomposition, while PAHs contribute to the soot at lower flame temperatures, where the LTC regime of operation falls.

For the two conditions, HTC diffusion and LTC early injection condition, where the soot model predictions are closer to the experiments, two-sets of Φ -T maps were generated. The first map (Figure 10) relates cells with maximum Φ , maximum temperature, maximum soot, and maximum NO at different crank angles (transient Φ -T map) while the second map (Figure 11) shows, at the maximum sooting crank angle (4

ATDC for diffusion combustion and -4 ATDC for early injection), computational cells with more than 50% of either the maximum NO value or maximum soot value at that crank angle position (The NO and soot peninsulas in the maps are from Akihama et al. [1]).

As seen from Figure 10, there is a good correlation between the maximum temperature cells (rectangles) and maximum NO cells (inverted triangles), while the maximum Φ cells (solid circles) correlate well with the maximum soot cells (open circles) above 1750 K at the different crank angles. As seen in Figure 11, the operating regime clearly shifts from high soot and high NO_x locations (rectangles) to low soot and low NO_x locations (circles), making this strategy attractive for medium loads. Thus, the soot model predictions seem to qualitatively agree well with conventional understanding.

Figure 12 shows the evolution of d_p vs. particle number and d_p vs. percent soot mass for the HTC diffusion case at different crank angles. As seen in Figure 12(a), soot particles only up to 30 nm are formed initially and most of the mass is accumulated within 10-30 nm soot particles. At 5 CAD ATDC (Figure 12(b)), most of the soot mass is accumulated in the 20-60 nm range with maximum number of particles and mass in the 30-40 nm range. At a later crank angle, CA = 10 ATDC (Figure 12(c)), the soot mass is quite evenly distributed between 30 to 120 nm particles. The larger size soot particle population is due to enhanced surface growth and soot coagulation. Finally, in the soot oxidative phase at 20 CAD ATDC (Figure 12(d)), higher particle size soot is oxidized and the soot population is seen to vary from 20 to 50 nm that contributes significant mass. This is also seen from in-cylinder contours of soot species density (g/cm^3), OH species density (g/cm^3) and d_p (nm) for 10, 15, and 20 CAD ATDC as shown in Figure 13, which is a cut-plane view along the spray axis. It is seen that at 10 CAD, the soot concentration is very high, which is progressively oxidized by the presence of OH radicals and hence the residual soot mass that is left behind at 20 CAD is of smaller size.

Figure 14 shows the evolution of d_p vs. number density and d_p vs. percent soot mass for the LTC early injection case at different crank angles. As seen in the figure, this combustion condition is dominated by in-cylinder soot diameters that do not exceed 50 nm. Initially, predominant particles are in the size range between 10 and 30 nm (-5 CAD ATDC), but at later crank angles higher diameter particles are also populated (CAD ATDC = 0 and CAD ATDC = 5). At a crank angle of 20 ATDC, particles ranging from 0 to 50 nm are populated fairly equally in terms of their mass, but the number of larger particles progressively decreases.

Thus, under the present conditions of study, the HTC-diffusion condition is characterized by high soot, higher in-cylinder particle numbers and larger in-cylinder particles, at least for a short period of time, that eventually oxidize, while the LTC early injection regime is characterized by significantly lower soot, lower in-cylinder particle number and particle diameters that are not widely populated even for a short period of time. These results explored the in-cylinder evolution of soot. The following section explores the evolution of particles closer to EVO.

2.3 Metal engine-out soot predictions

In this section model predicted results are compared with experimental data obtained from a light-duty GM 1.9L single cylinder version engine operated under PCCI conditions. The EVO model predictions are compared with experimental AVL filter smoke number (FSN) converted values using:

$$C \left\{ \text{mg/m}^3 \right\} = \frac{1}{0.405} \cdot 4.95 \cdot \text{FSN} \cdot \exp(0.38 \cdot \text{FSN}) \quad (13)$$

where C is the soot mass per sample exhaust volume. The above correlation was primarily developed for conventional diesel combustion although recent work by Northrop et al. [23] has found that it can also be used for partially premixed LTC and biodiesel combustion without any need for modifications. The model performance was tested at an EGR rate of 52%, and a main injection sweep was done from -8 to -2 CAD ATDC. The first pulse injection pressure (300 bar) at -51 CAD ATDC was significantly less than the second injection pressure (1200 bar) to reduce spray-wall impingement (Swor [24]). Table 4 summarizes the tested conditions and Figures 15(a) and 15 (b) show the model predicted pressure and heat release traces as compared to the experiments for main SOI of -8 CAD ATDC and -2 ATDC. For the SOI = -2 CAD ATDC case (Figure 15(b)), there were significant variations in the in-cylinder pressures for different experimental runs and thus three traces are shown. Figure 15(c) shows the model predicted exhaust soot compared to the average of three engine measurements with the associated error bars and Figure 15(d) shows the in-cylinder soot formation and oxidation histories.

Figures 16 (a), (b) and (c) show particle number vs. d_p and particle number vs. soot mass for the SOI = -8 CAD ATDC case at different crank angles and Figure 16(d) shows for the SOI = -2 CAD ATDC case at EVO. It is seen that the soot diameter corresponding to the maximum soot mass transitions from 20-30 nm

size particles (Figure 16(a)) to 40-50 nm size particles (Figure 16(b)) at 40 CAD. At the time of EVO (Figure 15(c)), most of the particle mass is between 30 to 35 nm and 50 to 65 nm size particles, while a higher number of particles is seen between 15 to 35 nm. Interestingly, two characteristic modes are seen in this case. Figure 16(d) shows the EVO distribution (112 CAD ATDC) for SOI = -2 CAD ATDC in 1 nm bins. In this case, it is seen that higher soot size particles are unpopulated and all the mass is found in particles with size less than 40 nm. The SOI = -8 CAD ATDC case is characterized by higher soot formation as compared to the SOI = -2 CAD ATDC, as seen from Figure 15(d). The higher soot formation rate due to higher inception and surface growth rates coupled with slower oxidation rates after 40 CAD ATDC, lead to the larger particle size population as compared to the SOI = -2 CAD ATDC case at EVO.

These results demonstrate that the present soot model can be effectively used for relative comparisons of exhaust soot diameter and particle number distributions involving different parametric sweeps. This capability is very important for analysis-led engine design and combustion strategy development for future diesel engines.

4. Summary and conclusions

In the current study a previously developed soot model was applied to study a variety of combustion regimes, including conventional diesel combustion, high-EGR LTC, and multiple injection PCCI conditions. The following conclusions were drawn regarding the soot mass predictions:

- For the constant volume chamber combustion simulations, the model was able to reproduce the trend of high soot at lower injection pressures or when larger nozzle diameter holes are used. The effect of decreasing the nozzle hole diameter to about 50 μ is more pronounced than increasing the injection/rail pressure to 190 MPa for reducing soot formation due to the enhanced entrainment. The in-cylinder soot peaks were modestly under-predicted by the model except for the 50 μ nozzle hole diameter case. The widths of the sooting zones were smaller than seen in the experimental sprays, which is possibly due to narrower rich spray cores predicted by the model compared to the experiments, or to enhanced model-predicted soot oxidation.
- For the Sandia optical engine study, the model yielded higher in-cylinder soot formation for high temperature diffusion combustion compared to high temperature premixed combustion. The early and

late injection LTC cases yielded much lower in-cylinder soot formation. The model predictions agreed very well to the experiments for diffusion combustion and early injection LTC.

- Finally, comparisons with engine-out soot data from a metal light-duty engine were made. The model did not need any additional tuning of the calibration constants and the model predicted soot at EVO compared well with PCCI experiments over a sweep of main SOI timings.

In terms of predictions of soot particle size, the following observations were made:

- Generally, the soot particle size predicted by the model was larger for high-sooting conditions compared to low-sooting conditions over the studied parametric sweeps (SOI, EGR etc.).
- For steady-state combustion in the spray chamber, the soot size at the maximum mass fraction location was seen to increase with either reducing in-cylinder O_2 at the same in-cylinder density, or by increasing chamber density at the same in-cylinder O_2 volume fraction. Also, the soot size contribution corresponding to maximum soot mass was seen to marginally decrease with decreasing nozzle diameter (21 nm for 180 μ to 13 nm for 50 μ), or by increasing injection pressures (23 nm for 40 MPa to 18 nm for 190 MPa). Similar trends for soot primary particle diameter with increasing diesel injection pressures ($\sim$ 18.5 nm at 50 MPa, $\sim$ 18 nm at 90 MPa, and $\sim$ 6 nm at 130 MPa) were seen by Ryser et al. [25] using time-resolved laser induced incandescence. In the engine study of Mathis et al. [26], at low load, transmission electron microscope (TEM) analysis yielded primary particle diameters of 32.5 nm, 29.6 nm and 17.5 nm when operated at injection pressures of 50, 110 and 140 MPa, respectively. However, at high load an increase in the rail pressure from 80 MPa to 140 MPa yielded only a small reduction from 21.4 nm to 20.4 nm. Thus, the current model predictions agreed well with these experimental trends in soot diameter.
- For the metal engine with PCCI combustion, the soot particle size was seen to decrease with retarded main SOI (-8 CAD ATDC and -2 CAD ATDC) at EVO. This was due to higher soot formation, coupled with lower soot oxidation beyond 40 CAD ATDC for the early injection case.
- In the current study a unimodal type soot size distribution with respect to soot particle number or soot mass was obtained for all simulations, except for the SOI = -8 CAD ATDC engine case. This may be a function of higher residence time (early injection) available for soot surface growth and coagulation, leading to larger particles and lower soot oxidation rates (inadequate oxidation of smaller size particles).

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Smokeless Rich Combustion by Reducing Temperature

Figure 1: Generalized $\Phi - T$ map showing HCCI, PCCI and LTC regimes of operation (Original figure from Akihama et al. [1] and modified based on Tree and Svensson [2]).

Figure 2: Model predicted total soot mass in the n-heptane jet as compared to the experiments [19].

Figure 3: Histograms of d_p vs. number of particles and d_p vs. soot mass at 6.5 ms after SOI for a) 21% O_2 , $\rho = 14.8 \text{ kg/m}^3$, b) 10% O_2 , $\rho = 14.8 \text{ kg/m}^3$, and c) 10% O_2 , $\rho = 30 \text{ kg/m}^3$. The plots shows the effect of in-cylinder oxygen and density.

Figure 4: Comparison of model predicted soot volume fraction (ppm) distributions (right) with experiments (left) using 100 μ nozzle diameter at injection pressures of a) 40 MPa, b) 90 MPa, c) 140 MPa, and d) 190 MPa. Experimental results can be found in <http://www.sandia.gov/ecn/cvdata/dsearch/frameset.php>

Figure 5: Comparison of model predicted soot volume fraction (ppm) distributions (right) with experiments (left) for 140 MPa injection pressure with nozzle diameters of a) 50 μ , b) 71 μ , c) 100 μ , and d) 180 μ . Experimental results can be found in <http://www.sandia.gov/ecn/cvdata/dsearch/frameset.php>

Figure 6: Variation of model predicted steady-state total in-cylinder soot mass (per unit injected fuel mass) with injection pressure and nozzle diameter.

Figure 7: Variation of d_p vs. soot mass histograms at a) different injection pressures and b) different nozzle diameters

Figure 8: Model predicted pressure and heat release traces as compared to the experiments for a) high temperature premixed combustion b) late injection low temperature combustion c) early injection low temperature combustion and d) high temperature diffusion combustion.

Figure 9: Model predicted total in-cylinder soot histories as compared to the two-color experiments for four different combustion strategies.

Figure 10: Transient Φ -T map for a) HTC diffusion combustion and b) LTC early injection over a CA sweep. The Φ -T contours are the generalized Φ -T contours from Figure 1

Figure 11: Comparison of the operating regimes of HTC diffusion and LTC early injection in the Φ -T map at 4 and -4 CA ATDC, respectively. The Φ -T contours are the generalized Φ -T contours from Figure 1

Figure 12: Histograms of d_p vs. number of particles and d_p vs. soot mass at a) CAD ATDC = 0, b) CAD ATDC = 5, c) CAD ATDC = 10 and d) CAD ATDC = 20 for HTC condition.

Figure 13: Contours of soot and OH species density (g/cm^3), and d_p (nm) at 10 CAD ATDC (top row), 15 CAD ATDC (center row) and 20 CAD ATDC (bottom row). The circumferential edges represent the liner.

Figure 14: Histograms of d_p vs. number of particles and d_p vs. soot mass at a) CAD ATDC = -5, b) CAD ATDC = 0, c) CAD ATDC = 5, and d) CAD ATDC = 20 for LTC condition.

Figure 15: Model predictions of a) pressure and heat release rate at SOI = -8 CAD ATDC, b) pressure and heat release rate at SOI = -2 CAD ATDC, c) EVO soot mass as compared to experiments and d) In-cylinder soot formation and oxidation histories.

Figure 16: Histograms of d_p vs. number of particles and d_p vs. soot mass at a) CAD ATDC = 10 for SOI = -8 CAD ATDC, b) CAD ATDC = 40 for SOI = -8 CAD ATDC, c) CAD ATDC = 112 (EVO) for SOI = -8 CAD ATDC, and d) CAD ATDC = 112 (EVO) for SOI = -2 CAD ATDC

Table 1: SANDIA n-heptane constant volume spray chamber combustion conditions.

Vol.% O ₂	Vol.% N ₂	Vol.% CO ₂	Vol.% H ₂ O	Ambient density (ρ) – kg/m ³
21	69.33	6.11	3.56	14.8
15	75.15	6.22	3.63	14.8
12	78.07	6.26	3.65	14.8
10	80.00	6.33	3.67	14.8
8	81.95	6.36	3.69	14.8
15	75.15	6.22	3.63	30.0
12	78.07	6.26	3.65	30.0
10	80.00	6.33	3.67	30.0
8	81.95	6.36	3.69	30.0

Ambient temp. (T) = 1000 K, nozzle diameter (d) = 0.1 mm, injection profile – top-hat, injection pressure = 1500 bar, fuel = n-heptane, injection temperature (T_{inj}) = 373 K.

Table 2: SANDIA diesel constant volume spray chamber combustion conditions

Inj. pressure (Mpa)	Nozzle dia.(μ)	Inj. quantity (mg)	Inj. time (ms)
140	50	11.9	17.0
140	71	8.2	5.9
140	180	39.8	4.7
140	100	14.7	5.4
40	100	7.5	4.8
90	100	11	4.9
190	100	16.2	5.1

Ambient temp. (T) = 1000 K, 21% O_2 , ambient density (ρ) = 14.8 kg/m³, injection profile – top-hat, fuel = #2 diesel with 33.8% by vol. aromatics, injection temperature (T_{inj}) = 436 K.

Table 3: Sandia Cummins N14 optical engine conditions.

Parameters	High temperature diffusion combustion	High temperature premixed combustion	Low temperature early injection	Low temperature late injection
Ambient vol. O ₂ %	21	21	12.7	12.7
SOI ATDC	-7	-5	-22	0
Intake press. (MPa)	0.233	0.192	0.214	0.202
Intake temp. (C)	111	47	90	70
Injected fuel (mg)	61	61	56	56
Inj. Pressure (bar)	1200	1200	1600	1600
IMEP (bar)	4.4	4.5	3.9	4.1

Table 4: GM metal engine operating conditions

% EGR	52
First SOI (CAD ATDC)	-51
Main SOI (CAD ATDC)	-8, -6, -4, -2
Intake press. (MPa)	0.162
Intake temp. (C)	60
Fuel in first pulse (mg)	2.07
Fuel in second pulse (mg)	11-11.5
First injection pressure (bar)	300
Second injection pressure (bar)	1200
IMEP (bar)	5.5