Efficient simulation and auto-calibration of soot particle processes in Diesel engines

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Abstract

Parameters describing soot particle processes are generally derived from a limited number of experimental studies. These parameters then have to be carefully calibrated for different operating conditions in internal combustion engine applications. This paper presents an innovative calibration procedure for soot simulation in Diesel engines. A Diesel engine is simulated using the Stochastic Reactor Model engine code, which is implemented with the Moment Projection Method for handling the soot particle dynamics. The main advantage of the engine-soot model is its low computational cost. The model is then coupled with an advanced statistical toolkit, Model Development Suite, where the Hooke-Jeeves algorithm is adopted to calibrate seven soot model parameters automatically based on the measurement data. The ability of the integrated code for soot model calibration is evaluated by simulating the soot formation and oxidation processes in a heavy-duty Diesel engine which is operated under 18 different conditions. Results suggest that the integrated code is able to calibrate

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the soot model parameters effectively. A significant improvement in the match between the simulation results and experimental soot emission is obtained after calibration.

Keywords: Diesel engine, soot, calibration

1. Introduction

Compared with gasoline engines, the merits of Diesel engines are lower fuel consumption and higher thermal efficiency [1, 2, 3, 4, 5]. Diesel engines are therefore heavily relied upon for power generation, seaborne and land transport for freight as well as for passenger cars [6]. For years to come, the Diesel engines will remain as a major energy source for our daily life and industrial activities [7, 8, 9]. However, due to the existence of rich high temperature zones leading to fuel pyrolysis, Diesel engines tend to produce more soot and other particulate matter (PM) in the exhaust gas which challenges its viability as more stringent emissions standards have been imposed [10, 11, 12, 13, 14, 15]. In order to improve our understanding of the soot formation and oxidation mechanisms and to meet the demand for cleaner combustion strategies, modelling and simulation of soot formation and oxidation in Diesel engines are of increasing importance.

The formation and oxidation of soot is a rather sophisticated physical and chemical process involving gas-phase chemistry, heterogeneous reactions on the particle surface and particle dynamics depending on a wide range of parameters [16, 17, 18, 19, 2]. The major processes considered important for soot formation in Diesel engines include: inception, *i.e.* formation of the primary soot particles through collision of two polycyclic aromatic hydrocarbon (PAH) molecules [20]; condensation, *i.e.* deposition of the PAH molecule on the soot surface leading to soot mass growth [21]; coagulation, *i.e.* formation of large soot particles due to collision and sticking among the small ones [22, 23, 24]; surface growth, *i.e.* increase of soot mass following the hydrogen-abstraction and acetylene-addition (HACA) mechanism [25], and oxidation where the soot particle is oxidised through heterogeneous reactions with molecular oxygen and

hydroxyl radicals [25]. The evolution of the soot particle size distribution (PSD) can be modeled by using population balance equations (PBEs) which are in mathematics a set of partial differential equations [26]. However, parameters describing these soot process rates are derived from a limited number of experimental or theoretical studies. There is a lack of consensus within the literature in terms of these parameters and the uncertainty ranges. Therefore, these model parameters have to be carefully calibrated for different engine operating conditions in order to achieve high prediction accuracy.

The purpose of calibration is to determine the model parameters to achieve the smallest difference between the experimental measurements and the numerical simulation results with minimal consumption of computation time. Manual calibration is time-consuming, impractical and may not always give optimal results. To realise efficient model-based engineering, it is vital to combine physical-chemical models with advanced optimisation methods to achieve auto-calibration of model parameters. One of the challenges in this endeavour is to find a balance between the predictive power of the model and computational expense due to the large numbers of model evaluations required for calibration. For example, a global calibration across many operating points using computational fluid dynamics with detailed chemistry and soot models is largely infeasible at present. In [27], a self-calibration model for Diesel engine simulation is presented. The model consists of a stochastic reactor model (SRM) for Diesel engine in-cylinder processes simulations and a package of optimisation algorithms (OPAL) for various optimisation problems. Genetic algorithms (GA) were adopted to determine the model parameters selected for calibration against the measured in-cylinder pressure history and engine-out emissions of nitrogen oxides and unburned hydrocarbons. It has been shown that the presented approach is able to efficiently calibrate the Diesel engine model, and that a higher calibration accuracy is achieved compared to manual calibration. In [28], a combination of GA and ant colony algorithms for calibrating combustion and heat transfer parameters including radiation coefficient, the combustion speed and terminal angle is proposed. This new approach has been shown to satisfy the

required accuracy with reduced simulation times. In [29], a Hybrid Calibration Method (HCM) for internal combustion engine (ICE) simulation is proposed. This method consists of two steps. The first step comprises the determination of calibration parameters for ICE sub-systems based on the direct search procedure. In the second step, calibration is performed with a reduced number of calibration parameters for the entire ICE simulation. The main advantage of HCM is its ability for the calibration of a large number of tuning parameters in a robust, repeatable and time-efficient manner.

The current literature on the engine model calibration is largely targeted at the engine in-cylinder pressure. Very little is focused on soot emissions. In the few publications [30, 31] where the soot emission is the calibration target, the adopted soot models are restricted to over-simplified empirical ones [32, 33]. The main advantage of these empirical soot models is the low computational cost. However, they do not account for the detailed soot particle dynamics. As a result, limited information on the soot formation and oxidation processes can be provided. By contrast, the detailed soot model is able to provide a deep insight into the soot particle dynamics. The problem is that the PBEs describing the soot particle processes usually take lots of computational resources, making the calibration highly time-consuming given the fact that usually multiple simulation times are needed to complete the calibration process. An efficient engine-soot code is therefore desired for calibration of the detailed soot model parameters. Generally, there exist three approaches to handle the soot PBEs: stochastic method [34, 35], sectional method [36, 37, 38] or method of moments (MOM) [39, 40, 41]. The stochastic method and sectional method are accurate and able to provide the detailed soot PSD but suffer from a high computational cost. MOM is much superior to other methods in terms of computational efficiency. With MOM, the soot PBEs are transformed into a set of moment equations and only a few lower-order moments are solved for, thus reducing the computational cost significantly. However, MOM suffers from some closure problems arising from the application of realistic collision kernels and particle depletion due to oxidation. Over the last three decades, numerous moment

methods have been proposed [39, 40, 41, 42, 17, 43, 44] trying to handle these problems. Among them, the Moment Projection Method (MPM) [45, 46] is very promising, having been shown to be able to handle all particle processes with satisfactory accuracy.

The main purpose of this paper is to present a new calibration procedure for soot simulation in Diesel engines which is able to determine the optimal soot model parameters automatically. To achieve this goal, the Stochastic Reactor Model (SRM) engine code [47, 48, 49, 50, 51, 30, 31] is used to simulate the combustion process, including the formation of soot, inside Diesel engines. The Moment Projection Method is implemented to handle the soot particle dynamics, and it is demonstrated that this does not substantially increase simulation times. The SRM code is then coupled with the Model Development Suite (MoDS) [52, 53] where a direct search algorithm is adopted to calibrate the soot model parameters automatically based on the measured soot emission data over a range of different operating conditions.

The paper is structured as follows. In the next section, the engine and soot models are presented. The incorporation of the Moment Projection Method into the SRM framework is explained followed by an outline of the calibration algorithm. Section 3 briefly describes the experimental measurements. In section 4, the performance of the integrated code for calibration of soot model parameters is tested by simulating a four-cylinder heavy-duty Diesel engine operated under 18 different conditions which cover a wide range of soot emissions. Finally, some key conclusions are summarised.

2. Model description

2.1. Engine model

The Stochastic Reactor Model (SRM) engine code is a spatially zero dimensional model for physical and chemical processes, which is applicable for simulations of in-cylinder combustion processes [49, 47]. The model is inspired by the Probability Density Function (PDF) transport methods [54]. It employs

detailed chemical kinetics to describe the combustion process inside the engine but at low computational cost [51, 31]. An important concept in the SRM is the so-called “stochastic particle” which can be taken as a fluid parcel in the cylinder. One stochastic particle represents a point in phase-space for the local scalar variables such as species concentrations, temperature, and pressure [48]. The engine cylinder charge can then be split into an ensemble of stochastic particles to represent the distribution of these variables [50]. The time evolution of these stochastic particles and hence the in-cylinder species concentrations and temperature distributions as a consequence of processes such as heat transfer, turbulent mixing, piston movement, and fuel injection are described by appropriate sub-models which are given in [55]. The precision of the engine model predictions is dependent on the number of stochastic particles. Higher accuracy is achieved with a larger number of stochastic particles but at the expense of computational cost. Usually 100 stochastic particles are sufficient for engine simulations.

2.2. Soot model

Within each stochastic particle, the soot model proposed by [56, 57] is adopted in this work to describe the soot formation and oxidation processes. The soot is modelled as a population of spherical particles composed of merely carbon atoms. The physical and chemical processes considered to be important for the formation of soot particles are inception, coagulation, condensation, surface growth, and oxidation. The detailed description of these processes can be found in [56, 57]. Here we only focus on the soot reaction kernels. We have introduced a set of multipliers which are applied to these kernels so that the soot reaction rates can be calibrated against the experimental soot emissions.

For inception, the reaction rate is expressed by using a collision efficiency model with the collision kernel given as:

$$\beta_{i,j}^g = x_{in} \varepsilon_{i,j} \sqrt{\frac{8\pi k_B T}{\mu_{i,j}}} (r_i + r_j)^2. \quad (1)$$

$\varepsilon_{i,j}$ is a size-dependent enhancement factor due to attractive or repulsive inter-particle forces. k_B is the Boltzmann constant, T is the temperature, $\mu_{i,j}$ is the reduced mass of the collision species, and r_i is the radius of the species of size i . x_{in} is the multiplier introduced to tune the soot inception rate.

Coagulation refers to the formation of large particles through collision and sticking among the population of soot particles. The process is modelled using the Smoluchowski's equation [58]. The collision frequency is dependent on the Knudsen number. For the collisions occurring under the continuum regime the coagulation kernel is given as

$$\beta_{i,j}^c = x_{\text{cont}} K_c \left[\frac{1}{i^{1/3}} + \frac{1}{j^{1/3}} + K'_c \left(\frac{1}{i^{2/3}} + \frac{1}{j^{2/3}} \right) \right] (i^{1/3} + j^{1/3}), \quad (2)$$

and for the collisions under the free-molecular regime the kernel becomes:

$$\beta_{i,j}^f = x_{\text{free}} K_f \left(\frac{1}{i} + \frac{1}{j} \right)^{1/2} (i^{1/3} + j^{1/3})^2. \quad (3)$$

K_c , K'_c and K_f are the kernel factors which are functions of temperature, gas viscosity and mean free path. x_{cont} and x_{free} are the multipliers used to calibrate the coagulation rates. It is suggested [59] that the collisions of soot particles under Diesel engine conditions are usually in the transition region between the free molecular regime and the continuum regime. Therefore, the harmonic mean value of the coagulation rates determined in the two regimes is adopted as the coagulation rate in this work.

Table 1 lists the mechanism used for soot surface reactions. The soot surface growth is modelled using the HACA mechanism [25]. The oxidation of soot particles via O_2 is described using the concept of soot surface radical sites while the oxidation by OH is modelled through a collision theory between OH radical and soot particles. Condensation is modelled as the deposition of a pyrene molecule from the gas-phase on the surface of soot particles. For all these processes, the multipliers are introduced as listed in Table 1 to tune the corresponding reaction rates.

As already mentioned, these soot particle processes can be modelled by using PBEs. Since the soot particles usually span a wide size range, an infinite number

Table 1: Soot surface reactions. The rate parameters are taken from [56].

No.	Reaction	$k = AT^n \exp(-E/RT)$			Multiplier
		A [cm ³ /mol/s]	n [-]	E [kcal/mol]	
S1	$C_{\text{soot-H}}^i + H \rightleftharpoons C_{\text{soot-}}^i + H_2$	4.2×10^{13}		13.0	-
		3.9×10^{12}		11.0	-
S2	$C_{\text{soot-H}}^i + OH \rightleftharpoons C_{\text{soot-*}}^i + H_2O$	1.0×10^{10}	0.734	1.43	-
		3.7×10^8	1.139	17.1	-
S3	$C_{\text{soot-*}}^i + H \rightarrow C_{\text{soot}}^i$	2.0×10^{13}			-
S4	$C_{\text{soot-*}}^i + C_2H_2 \rightarrow C_{\text{soot-H}}^{i+2} + H$	8.0×10^7	1.56	3.8	x_g
S5	$C_{\text{soot-*}}^i + O_2 \rightarrow 2CO + C_{\text{soot-H}}^{i-2}$	2.2×10^{12}		7.5	x_{O_2}
S6	$C_{\text{soot}}^i + OH \rightarrow CO + C_{\text{soot}}^{i-1}$	reaction probability = 0.13			x_{OH}
S7	$C_{\text{soot}}^i + PAH \rightarrow C_{\text{soot}}^{i+n_{PAH}}$	reaction probability = 1.0			x_{cond}

of partial differential equations are needed to describe these processes. In order to reduce the computational cost, the method of moments is adopted to handle the soot model. The moments are defined as

$$M_r = \sum_{i=i_0}^{\infty} i^r N_i, \quad r = 0, 1, \dots, \infty, \quad (4)$$

where N_i is the number density of soot particles that contain i carbon atoms. i_0 refers to the smallest particle size. M_r is the r^{th} order moment. The lower-order moments have physical meanings, for example M_0 represents the total soot number and M_1 is related to the total soot mass. One only needs to solve for a few lower-order moments which are sufficient to provide the general information on soot particles. However, the soot moment equations are usually unclosed. The application of the collision kernels leads to the presence of fractional or even negative order moments that are not directly tracked and have to be properly estimated. Another closure problem arises from the soot oxidation process where the number of the smallest soot particles has to be approximated to evaluate

the depletion of soot particles. In this work, the Moment Projection Method (MPM) is adopted to handle these problems. The basic idea of MPM is to approximate the PSD with a set of weighted particles and directly track the number of the smallest particles by fixing one of the weighted particle sizes at the smallest size. Please refer to [45, 46] for the detailed numerical algorithm for MPM.

2.3. Integrating the models

Figure 1: A schematic of the coupled engine-soot model.

Figure 1 shows the schematic of the integrated engine-soot model. The engine cylinder charge is split into an ensemble of stochastic particles that represent the distribution of temperature, gas-phase species concentrations and soot moments. The main step of integrating the soot model into the SRM code consists of associating the soot moment equations with each of the SRM stochastic particles and solving them in every computational time step. Another key step is to link the gas-phase chemistry to the soot particle processes by identifying one or more species in the chemical mechanism as inception and soot surface

reaction species. The contributions to these gas-phase species due to soot inception and surface reactions are fed back to the gas-phase chemistry to adjust the corresponding gas species production rates. After these unknowns are updated within each stochastic particle, a mixing among the stochastic particles is conducted to generate new distributions of temperature, gas-phase species concentrations, and soot moments inside the engine cylinder which are fed as inputs into each stochastic particle and repeat the above steps until simulation time ends. It should be noted that the selection of a proper mixing model is very important. An inappropriate mixing of moments may violate the realisability of the moment set, leading to unphysical results. Realisability refers to the existence of an underlying particle size distribution (PSD) which corresponds to a set of moments [60]. All the moments are linked to each other under very complex mathematical relationships. If these relationships are not preserved due to the mixing of moments, the moment set can be unrealisable, indicating that no PSD can be represented by these moments or they lead to unphysical distributions [61]. In this work the mixing of the stochastic particles is described by Curl’s model [62] in which a pair of stochastic particles is chosen at a time according to a certain probability law characteristic of the particular model and mixed to produce two new stochastic particles. The properties of the two post-mixed stochastic particles are the mean of the properties of the original ones. Curl’s mixing model has been proven in [62] to be able to maintain the consistency between the PSD and the corresponding moments. As a result the realisability of the moment set is ensured.

2.4. Soot model calibration

The seven soot model multipliers described in the previous section were calibrated automatically through MoDS based on the experimental soot emissions obtained at $N = 18$ operating points. The accuracy of the model multipliers was evaluated using a least-squares objective function:

$$f(x) = \sum_{i=1}^N \left[\frac{m_i - \tilde{m}_i(x)}{\sigma_i} \right]^2, \quad (5)$$

where $x = (x_{\text{in}}, x_{\text{cont}}, x_{\text{free}}, x_{\text{g}}, x_{\text{O}_2}, x_{\text{OH}}, x_{\text{cond}})$ refers to the vector of soot model multipliers. m_i and $\tilde{m}_i$ are, respectively, the experimental and simulated soot emissions for each test case.

The local optimisation was done with the Hooke-Jeeves algorithm [63] which is a type of direct search method. This algorithm is an extension of the basic discrete grid method, both of which are described in [64]. In the basic version, at the k^{th} iteration the algorithm searches for

$$x_k^* \in \mathcal{N}(x_k) : f(x_k^*) < f(x_k), \quad (6)$$

where $\mathcal{N}(x_k)$ is the set of points containing x_k and its immediate neighbours on a grid with spacing δx_k , *i.e.*

$$\mathcal{N}(x_k) = \left\{ x \mid \exists i \in \{1, \dots, D\}, a \in \{-\delta x_k, 0, +\delta x_k\} : x = x_k + a e_i, \right\} \subset \mathbb{R}^D, \quad (7)$$

where e_i is the unit vector in the i^{th} direction, and $D = 7$ in our case. If a better point cannot be found in $\mathcal{N}(x_k)$ then the search distance δx is reduced by multiplying it by $s < 1$. That is, the next point is given by

$$x_{k+1} = \begin{cases} x_k^*, & \text{if } f(x_k^*) < f(x_k), \\ x_k \quad (\text{and } \delta x_{k+1} = s \delta x_k), & \text{otherwise.} \end{cases} \quad (8)$$

Hooke and Jeeves' version extends this by suggesting that if x_k is better than x_{k-1} then taking another step in the same direction to $y_k = 2x_k - x_{k-1}$ and searching for

$$y_k^* \in \mathcal{N}(y_k) : f(y_k^*) < f(x_k), \quad (9)$$

where $\mathcal{N}(y_k)$ is defined as in Eqn. (7), might provide a further improvement. This gives

$$x_{k+1} = \begin{cases} x_k^*, & \text{if } f(x_k^*) < f(x_k) \text{ and } f(y_k^*) \geq f(x_k^*), \\ y_k^*, & \text{if } f(x_k^*) < f(x_k) \text{ and } f(y_k^*) < f(x_k^*), \\ x_k \quad (\text{and } \delta x_{k+1} = s \delta x_k), & \text{otherwise.} \end{cases} \quad (10)$$

Figure 2: The calibration workflow for the soot model multipliers in the coupled SRM-MoDS model.

The algorithm terminates if either the number of iterations, k , reaches $k_{\max}$ or δx_k is reduced such that $\delta x_k < \epsilon$, where ϵ is a user-defined tolerance.

Figure 2 shows the calibration workflow for the soot model multipliers in the coupled SRM-MoDS model. The values of the multipliers are initially set to 1.0. The predicted soot emissions of the SRM engine-soot model are compared with the experimental measurements to evaluate the accuracy of the soot model multipliers. If the predicted soot emissions are within the user-defined error tolerance, the calibration process stops and outputs the current multipliers as the final results. Otherwise, these multipliers are modified using the Hooke-Jeeves algorithm in order to minimise Eqn. (5). A set of new multipliers are then generated and sent to the SRM engine-soot model to repeat the above procedures until the desired error tolerance for the soot emissions is achieved.

3. Experiments

Table 2: Specification of Diesel engine geometry.

Quantity	Value
Bore	105 mm
Stroke	127 mm
Connecting rod length	219 mm
Wrist pin offset	0.5 mm
Compression ratio	16.5:1
Displacement	4400 cm ³

Table 3: Range of considered engine operating conditions.

Quantity	Value		Unit
	Min	Max	
Pilot SOI	-18.0	-7.3	CAD ATDC
Main SOI	-10.0	-0.1	CAD ATDC
Main injection fuel mass	100.5	110.0	mg
Injection pressure	1550	2500	bar

The engine considered in this work is a Caterpillar C4.4 ACERT four-cylinder, turbo-charged, heavy-duty Diesel engine. It has eight valves, a displacement of 4.4 litres, and is operated with a dual injection strategy. Further details of the engine specification are listed in Table 2.

In total, 18 different operating points, hereafter referred to as ‘test cases’, were adopted, at a range of operating conditions that are summarised in Table 3. The Diesel engine was running at a constant speed of 2200 rpm. The pilot injection fuel mass is always 2.9 mg for all the cases while different injection timings, injection pressures, and main injection fuel mass are adopted. The

cases have been chosen such that they cover a wide range of soot emissions from Diesel engines, and hence to ensure that the determined soot model parameters have a wide range of applicability.

Measurement data from the test bed obtained at steady-state operation includes pressure profiles, averaged over multiple cycles, and emissions of soot and NO_x , both of which are measured engine-out. NO_x emissions are measured in units of parts per million by volume (ppmv) with a tolerance target of 3%. Soot is measured using an industry-standard AVL Smoke Meter, with a nominal statistical uncertainty of 5%.

4. Results and discussion

In this section, the integrated codes were used to simulate combustion in the engine test cases described above. A detailed kinetic model for Primary Reference Fuel (PRF) [49] is employed to describe the gas phase chemistry. The chemical mechanism contains 208 species and 1002 reactions. The formation of soot precursors including Polycyclic Aromatic Hydrocarbons (PAHs) and other soot surface reaction related species such as acetylene are all included. For all the cases, simulations were run with a time step of 0.2 CAD. 100 stochastic particles were adopted in the SRM engine code. Figure 3 compares the simulated in-cylinder pressure profiles with the experimental measurements for six representative test cases. As can be seen the simulation results match the experimental data well. The predicted ignition timings and cumulative heat releases are in good agreement with the experiments. These results suggest that the combustion process inside the Diesel engine cylinder was successfully simulated with the integrated code.

In order to evaluate the computational efficiency of the integrated code for soot simulation in Diesel engines, for each test case the simulation was run twice: one considers both the gas phase chemistry and the soot particle dynamics while the other one only considers the gas phase chemistry. The CPU time spent for these cases is shown in Fig. 4. Note that for each test case the CPU time is

Figure 3: Comparison between simulated and measured in-cylinder pressures as a function of crank angle. For each case, the pressure values have been normalised by the maximum measured pressure $P_{\max}$.

Figure 4: Evaluation of the additional CPU costs induced by including the soot model in the Diesel engine simulation. For each test case, the CPU time is normalised by the result obtained where no soot particle dynamics are considered.

normalised by the result obtained where only gas phase chemistry is considered. As can be seen, the inclusion of the detailed soot model in the engine code increases the CPU cost only by about 20% for all the test cases. The main CPU cost for the engine simulations stems from solving the chemical kinetic equations. MPM transforms the soot population balance equations into only five moment equations, which can be solved alongside the chemistry equations, thus allowing a highly computationally efficient solution. This result should be seen in light of the fact that, traditionally, the treatment of soot models using stochastic methods tends to make Diesel engine simulations prohibitively expensive.

Figure 5 shows the distribution of temperature and equivalence ratio in a plot at several crank angles throughout the cycle for case 1. At $\text{CAD} = -50$, it

Figure 5: Time evolution of equivalence ratio and temperature distribution as a function of crank angle for case 1 obtained using the integrated code. The soot and NO_x iso-lines are adopted from [65].

can be seen that all the stochastic particles are located at a low temperature with a fuel/air equivalence ratio of zero since no fuel has been injected yet and the cylinder has been charged with air. At CAD = -20, these particles are found to be moved to a higher temperature region due to compression while the equivalence ratio still remains zero. Then at CAD = -5 where the pilot injection has already started, the burning of the fuel leads to a fast increase in temperature. Note that the particles with equivalence ratios close to stoichiometric reach the highest temperature as expected. Some stochastic particles start to be present in the NO_x peninsula. As the injection continues, the mixture becomes richer. Starting from Top Dead Center at CAD = 0, some stochastic particles start to enter the soot peninsula and become the source of soot formation. The stochastic particle with the highest equivalence ratio belongs to the area that surrounds the fuel injector where very little air exists. During the main combustion period from CAD = 0 to CAD = 30, the stochastic particles span a wide range of equivalence ratios and temperatures, leading to the continuous formation of both NO_x and soot. Then from CAD = 50, a shift of these stochastic particles towards lower temperature and smaller equivalence ratio is observed as the fuel is burned out. These particles start to leave the soot and NO_x peninsula. The formation rates for soot and NO_x are expected to decrease.

The simulation results and experimental measurements for NO_x and soot emissions are compared in Figs. 6 and 7, respectively. To indicate the calibration accuracy, the normalised root mean square error between the simulation results and experimental data has been calculated as follows:

$$\text{NRMSD} = \frac{\sqrt{\frac{\sum_{i=1}^N (m_i - \tilde{m}_i)^2}{N}}}{\tilde{m}_{\max}}, \quad (11)$$

where $\tilde{m}_{\max}$ is the maximum measured engine-out soot mass. As can be seen the predicted NO_x emissions by the integrated code are in good agreement with the experimental measurements both qualitatively and quantitatively for all 18 test cases. By contrast, the soot emissions are largely over-predicted by the uncalibrated soot model where all the soot process multipliers are set as 1.0.

Figure 6: Comparison between the simulation results and experimental measurements for NO_x emissions for the 18 test cases, with normalised root mean square error. Values are normalised by the maximum experimental NO_x emission value. The solid line indicates a 1:1 relation.

This indicates that the formation and growth rates for soot particles under Diesel engine combustion conditions have been overestimated with the original soot model parameters.

The calibration of the soot model multipliers was completed after 156 simulation runs. The finally accepted multipliers determined by the Hooke-Jeeves algorithm are shown in Fig. 8. It can be seen that only the multipliers for soot surface growth, oxidation via molecular oxygen and free-molecular coagulation are changed significantly while the multipliers for other soot particle processes remain almost unchanged. In particular, x_{free} has been increased by around 327 times. Figure 7 shows a comparison between the experimentally measured engine-out soot mass with the simulation results before and after calibration of

Figure 7: Comparison between the experimental soot mass and the simulated results before and after calibration of soot model parameters, with normalised root mean square error. Values are normalised by the maximum experimental value for the soot emission. The solid line indicates a 1:1 relation.

Figure 8: Change of the soot model multipliers after calibration.

the soot model parameters. As can be seen, compared to the uncalibrated soot model the calibrated model exhibits a much better match between the simulation results and the experimental data, suggesting that soot formation rates have been tuned to more reasonable values with the newly proposed calibration technique. However, in Fig. 7 we could still observe some discrepancies between the experimental measurements and the simulation results obtained with the calibrated soot model. For example the soot emissions in cases 2, 4, 8, 10, and 12 are over-predicted by around two orders with the new multipliers. These discrepancies might be a consequence of the inherent defects of the adopted soot model for describing the soot formation and oxidation processes. The detailed soot formation mechanism is far from being fully understood. Take inception as an example, the inception mechanism adopted in the current soot model is a very simple one which assumes the pyrene molecule to be the only soot precursor. The inception rate is determined through a simple collision equation. However, several studies [56, 66] indicate that PAHs larger than pyrene might act as the soot inception species. Furthermore, it has been suggested that the soot inception process should be reversible which is not the case in the adopted soot model. A better agreement between the experimental data and the simulation results can be expected if a more accurate soot model is adopted. It should be noted that the purpose of this paper is not to develop a new soot model but to present an efficient calibration procedure for soot simulation in Diesel engines. The proposed engine-soot model in this work has the advantage of low computational cost. The soot model multipliers have been successfully calibrated via the coupled SRM-MoDS code. We note that, while there is some statistical variability in the SRM simulations, we found that this does not noticeably affect the results of the calibration.

Next, the influence of the soot model multipliers on the soot formation and oxidation processes are discussed. Figure 9 compares the time evolution of the predicted total soot mass as a function of crank angle for case 1 obtained before and after the calibration of the soot model multipliers. As can be seen, at the beginning inception dominates the soot particle processes, leading to a fast

Figure 9: Comparison of the time evolution of the total soot mass as a function of crank angle obtained before and after calibration of the soot model for case 1.

build-up of soot mass inside the engine cylinder. The total soot mass keeps increasing until around $\text{CAD} = 50$ after which oxidation starts to dominate over the soot formation process. During the whole combustion process less soot mass is predicted using the calibrated soot model compared with the original one. Especially during the late combustion stage the soot mass predicted with the calibrated soot model exhibits a sharp decrease while the uncalibrated model leads to a slow change of the soot mass. As a result, a satisfactory agreement between the simulated engine-out soot mass and the experimental measurement is achieved with the calibrated soot model, while the uncalibrated model over-predicts the engine-out soot emission.

The predicted soot formation and oxidation rates for case 1 obtained before and after calibration of the soot model are shown in Fig. 10. The coagulation rate for the total soot mass is zero, therefore it is not shown in the figure. From

Figure 10: Time evolution of the soot inception and surface reaction rates as a function of crank angle for case 1 obtained using the integrated code.

Fig. 10 it can be seen that the oxidation rate via OH is much larger than that via O_2 , suggesting that the soot oxidation process is mainly governed by the external burning process. During the main combustion stage from 0 CAD to 60 CAD, the soot inception rates, condensation rates and surface oxidation rates predicted by the calibrated and uncalibrated soot models are almost the same. By contrast, the calibrated soot model leads to a lower soot surface growth rate compared with the uncalibrated model, which is the main contributor for the less soot mass obtained by the calibrated soot model as observed in Fig. 9. During the

late combustion stage (> 60 CAD), the calibrated soot model predicts a lower rate for not only the surface growth but also the inception and condensation processes compared with the uncalibrated soot model, which leads to a faster decrease in the total soot mass as observed in Fig. 9.

One significant weakness of the method of moments is that it can only provide the bulk quantities of soot particles while the detailed soot PSD is lost. In order to perform a comprehensive study on the soot particle dynamics, the maximum entropy (ME) approach [67] is adopted in this work as a post-process technique to reconstruct the soot PSD based on the obtained moments at each simulation step. The basic idea of ME reconstruction is to approximate the PSD using a series of exponential polynomials, which is given as

$$N(i) = \exp\left(-\sum_{j=0}^K \lambda_j i^j\right), \quad (12)$$

where λ_j ($j = 0, \dots, K$) are Lagrange multipliers. The reconstructed PSD has to meet the requirement that the first $K+1$ moments are equal to the moments of the real PSD:

$$\sum_{i=i_{\min}}^{i_{\max}} i^r N_K(i) = \sum_{i=i_{\min}}^{i_{\max}} i^r \exp\left(-\sum_{j=0}^K \lambda_j i^j\right) = M_r, \quad r = 0, \dots, K. \quad (13)$$

The key problem is to determine these Lagrange multipliers. To achieve this, a convex potential is introduced:

$$\Delta = \sum_{i=i_{\min}}^{i_{\max}} \left[\exp\left(-\sum_{j=0}^K \lambda_j i^j\right) - 1 \right] + \sum_{j=0}^K M_j \lambda_j. \quad (14)$$

One just needs to minimise this potential to find its stationary points. Please refer to [67] for the detailed ME algorithm.

The reconstructed soot PSDs as a function of crank angle for case 1 obtained before and after calibration of the soot model multipliers are compared in Fig. 11. As can be seen, at the beginning inception dominates the soot formation process, the PSD exhibits a delta distribution. An increase in the total soot number is observed from CAD = -5 to CAD = 0 as injection continues. From CAD = 4, the soot PSD starts to exhibit a bimodal distribution. The

Figure 11: Predicted time evolution of the soot particle size distributions for case 1 obtained before and after calibration of the soot model multipliers.

first mode arises from the continuous particle inception while the second mode is due to the particle growth and coagulation processes. As more and more particles are formed, the PSD is becoming wider. At the same time, the inception process is weakened as most fuels have been burned out and a shift of the PSD from bimodal to unimodal distribution is observed at the late combustion stage accompanied by a continuous decrease of the total soot number. Compared with the uncalibrated soot model, the calibrated soot model predicts a narrower soot PSD during the main combustion period from $CAD = 4$ to $CAD = 35$. This is mainly attributed to the decrease in the soot surface growth multiplier after calibration, which leads to a slow accumulation of soot particles at large sizes. In addition, the increase of the soot oxidation multiplier after calibration results in an overall lower soot number through the whole particle size range compared with the uncalibrated soot model. During the late combustion stage, the coagulation process starts to dominate the particle process as the soot surface reactions become weakened. Since the coagulation multiplier has been increased after calibration, a faster increase in the soot particle size range is induced by the calibrated soot model compared with the uncalibrated one.

The predicted evolution of the soot PSD within the engine cylinder seems reasonable. Unfortunately there is no available experimental measurement for engine in-cylinder soot PSD for comparison. Once again, it should be noted that the purpose of this paper is not to develop a new soot model to account for the soot formation and oxidation processes precisely. The aim here is to present an efficient approach for soot simulation in Diesel engines over a wide range of operating conditions. The integrated code proposed here can be used to handle the soot model with little CPU cost. It enables an auto-calibration of the model parameters which represents a significant improvement over the manual calibration which is time-consuming and impractical. Furthermore, a comprehensive analysis on the soot particle dynamics inside the engine cylinder can be conducted. The evolution of the soot particle size distribution can be studied, which resolves a long-standing problem of the method of moments.

5. Conclusion

In this work, an efficient calibration procedure for soot simulation in Diesel engines is developed and presented. The procedure is based on the integration between the engine-soot model and an advanced optimisation method. The Stochastic Reactor Model engine code is adopted to describe the combustion process inside Diesel engines. The Moment Projection Method is implemented into the Stochastic Reactor Model to account for the detailed soot particle dynamics including inception, condensation, coagulation, surface growth, and oxidation. The integrated engine-soot model is then coupled with the Model Development Suite where the Hooke-Jeeves algorithm is adopted to automatically calibrate the soot model parameters against the experimentally measured soot emissions.

The performance of the integrated code has been tested by simulating the soot formation and oxidation processes in a four-cylinder heavy-duty Diesel engine operated under 18 different conditions which cover a wide range for soot emissions. Results suggest that the integrated code is very efficient. The inclusion of the detailed soot model in the engine code induces an additional CPU cost of only about 20% – a relatively minor increase compared to the treatment of soot particle dynamics using stochastic methods, which tends to make simulations prohibitively expensive. The auto-calibration of the soot model parameters is completed after 144 simulation runs. Compared to the original soot model, the calibrated model exhibits a much better agreement between the simulation results and the measured soot emissions. A deeper investigation reveals that the soot model parameters undergoing major changes due to calibration are the multipliers for soot surface growth, O_2 oxidation, and free-molecular coagulation. The influence of these multipliers on the soot formation and oxidation processes in Diesel engines is discussed. Another advantage of the integrated code is that it is able to provide not only the bulk soot quantities such as total soot number and mass but also the evolution of the detailed soot particle size distribution, thus enabling a comprehensive study of the soot particle dynamics

inside the Diesel engine cylinder.

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Nomenclature

Upper-case Roman

A	Pre-exponential factor
C	Gas species concentration
E	Activation energy
K	Kernel
M	Moment
N	Number
P	Pressure
R	Universal gas constant
T	Temperature

Lower-case Roman

e	Unit vector
k	Rate constant

k_B	Boltzmann constant
r_i	Radius of particle of size i
m	Mass
x	Multiplier

Greek

β	Collision kernel
Δ	Convex potential
ϵ	Tolerance
ε	Enhancement factor
λ	Lagrange multiplier
μ	Reduced mass

Superscripts

g	Gas
c	Continuum regime
f	Free-molecular regime

Symbols

$\tilde{x}$	Approximation of x
x^*	New guess of x

Abbreviations

CAD	Crank angle degree
EOI	End of injection
EVC	Exhaust valve closing
EVO	Exhaust valve opening

GA	Genetic algorithm
HACA	Hydrogen abstraction and acetylene addition
HCM	Hybrid calibration method
ICE	Internal combustion engine
IVC	Inlet valve closing
IVO	Inlet valve opening
ME	Maximum entropy
MoDS	Model development suite
MOM	Method of moments
MPM	Moment projection method
OPAL	Optimisation algorithm
PAH	Polycyclic aromatic hydrocarbon
PBE	Population balance equation
PDF	Probability density function
PM	Particulate matter
PRF	Primary reference fuel
PSD	Particle size distribution
SOI	Start of injection
SRM	Stochastic reactor model

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