

A Numerical Study on Detailed Soot Formation Processes in Diesel Combustion

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ABSTRACT

This study simulates soot formation processes in diesel combustion using a large eddy simulation (LES) model, based on a one-equation subgrid turbulent kinetic energy model. This approach was implemented in the KIVA4 code, and used to model diesel spray combustion within a constant volume chamber. The combustion model uses a direct integration approach with a fast explicit ordinary differential equation (ODE) solver, and is additionally parallelized using OpenMP. The soot mass production within each computation cell was determined using a phenomenological soot formation model developed by Waseda University. This model was combined with the LES code mentioned above, and included the following important steps: particle inception during which acenaphthylene (A_2R_5) grows irreversibly to form soot; surface growth with driven by reactions with C_2H_2 ; surface oxidation by OH radical and O_2 attack; and particle coagulation.

The results obtained using our new model are compared to those generated using a RANS (RNG k-epsilon) model, and also to experimental data from the engine combustion network (ECN) of Sandia National Laboratories. The sensitivity of the LES results to mesh resolution is also discussed. The results show that both RANS and LES simulations predict the dispersion and vapor penetration of the injected fuel fairly well. LES generally provides flow and spray characteristics in better agreement with experimental data than RANS. It is also shown that the phenomenological soot model is useful for investigating soot particle production and distribution. The LES model was better than the RANS model at describing instantaneous soot concentration contour.

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INTRODUCTION

Diesel engines for automobiles are required to comply with increasingly stringent emission regulations and fuel economy standards. Computational fluid dynamics (CFD) has emerged as an effective tool for the development of higher efficiency and lower emission diesel engine combustion systems. Direct numerical simulation (DNS) is one of the most precise CFD methods, able to accurately describe organized vortex structures within turbulent flows. Using DNS to predict a diesel spray characteristic requires excessive computation time, therefore currently DNS is not a useful turbulent flow approach for practical applications.

There are two types of turbulent modeling approaches: Reynolds averaged Navier-Stokes simulation (RANS), which is based on ensemble and time averaged governing equations; and large eddy simulation (LES), based on spatial filtering. Because RANS relies on time averaging, detailed information about the instantaneous anisotropic characteristics of the

turbulent flow is inevitably lost when using this method. RANS simulations are therefore not optimal for studying highly transient flows, such as those encountered in engine cylinder. Owing to this, LES is becoming more popular in the field of fluid mechanics as a practical method for analyzing in-cylinder turbulent phenomena.

Recently a number of studies on the numerical analysis of fuel sprays using LES have been reported. Hori et al. [1] modeled non-evaporative and evaporative diesel sprays in a constant volume chamber by using an LES model. Their results show that the accuracy of evaporative spray simulation using LES is strongly grid-dependent. Furthermore, comparisons with RANS approaches have shown that LES modeling can give improved agreement with experimental data. Celic et al. [2] explored LES approaches in engines relatively early; their group implemented an LES turbulence model into the KIVA code [3] and calculated intake and compression flows. Some researchers used LES to simulate the fuel atomizing process in a direct

injection spark ignition engine with wide cycle-to-cycle variations. These results showed that an LES approach is able to describe highly unsteady, anisotropic fuel-air mixing processes and dynamically evolving fine scale vortical and scalar structures. Rutland and his group [5, 6, 7, 8, 9, 10] performed considerable research on the combustion modeling of internal combustion engines with the LES approach. Their model, called the dynamic structure model, works very well in engine applications, and provides accurate results for the subgrid turbulence kinetic energy (TKE) for use in combustion, scalar mixing, and spray models.

Soot models developed over the last few decades range from relatively simple phenomenological models to more complex kinetic models. The two-stage empirical soot model of Hiroyasu [11] is widely used due to its simplicity and convenience. In the Hiroyasu model, soot formation proceeds via two steps, namely the formation step and the oxidation step.

The mass rate of soot formation is assumed to be directly proportional to the amount of fuel vapor. The Arrhenius formation and oxidation rates are obtained from experimental data. It is known, however, that soot formation is a very complex physical phenomenon, involving not only gas-phase chemical processes but also physical processes such as particle dynamics. These considerations motivated Fusco et al. [12] to propose an eight-stage phenomenological soot model for studies of diesel combustion. This model considers eight global reaction steps, including particle inception, particle coagulation, surface growth, and surface oxidation, as well as global reaction steps for the formation and oxidation of intermediate species. Tao et al. [13] coupled a phenomenological soot model with a detailed chemical kinetic mechanism that incorporates polycyclic aromatic hydrocarbon (PAH) chemistry for gas phase soot precursor formation and oxidation. The predictions obtained by Tao et al. showed that this approach provides a superior description of diesel flame structure than simpler phenomenological approaches. Their simulations also provide additional useful information such as the soot precursor abundance, soot number density and soot particle size.

The CHEMKIN-II package [14] was used to model the combustion chemistry in this work. It is well known that simulating multi-dimensional combustion with detailed kinetics often requires solving a large number of ODE problems at each global time step. In many cases, the ODE integrations account for more than 90% of the wall-clock time for such simulations. In order to reduce the computational run time, a fast explicit stiff ODE solver, based on the existing CHEMEQ2 code [15], was employed for this study. Shi et al. [16] found that simulated data obtained with CHEMEQ2 showed good agreement with the traditional implicit stiff solver, DVODE [17]. Their results indicated that explicit integration methods are more efficient

than implicit methods for ODE systems that are only moderately stiff or non-stiff. Moreover, explicit algorithms typically involve simple logic flows which can be more easily understood. Note that smaller time steps are required to retain accuracy and avoid instability.

This paper presents a reduced chemical mechanism that couples a phenomenological soot formation model with complex kinetic chemical mechanisms for gas phase soot precursor formation and oxidation. The diesel oil surrogate mechanism developed at Chalmers University of Technology consists of 74 species and 320 reactions [18]. This has been used to simulate evaporative n-dodecane sprays and n-heptane combustion. The gas phase chemical mechanism is composed of the formation and oxidation of PAHs, which is up to two aromatic rings. The relatively small-scale hydrogen abstraction C_2H_2 addition (HACA) mechanism is used in this scheme. The phenomenological soot formation model was developed at Waseda University [19, 20] and is based on the model of Tao et al. [13]. The model's framework is based on four fundamental steps: particle inception via the five-membered ring PAH species acenaphthylene (A_2R_5), surface growth by the addition of C_2H_2 , surface oxidation by OH radical and O_2 attack, and particle coagulation.

The authors also parallelized the explicit CHEMEQ2 solver with OpenMP to utilize the multi-core processors. The production rate of chemical reactions within a computation cell is determined by each thread. The OpenMP implementation is well suited to CFD codes because they contain many loops over cells and vertices, and many of the calculations within these loops require information from neighboring cells and vertices. Communication and load balancing between threads is handled internally by the operating system.

NUMERICAL APPROACH

The CFD methodology used in this study is based on the KIVA4 code with improved subgrid turbulence, liquid spray, chemical combustion, and soot models. This section presents a brief overview of the governing equations and modeling methods that have been implemented in the KIVA4 code.

LES Equations

The filtering operator is applied in the form of a box filter to the conservation equations of species, momentum and energy. The LES equations for two-phase flows, in accordance with the KIVA4 [21] formulation, are described below.

Continuity equation:

$$\frac{\partial \bar{\rho}}{\partial t} + \frac{\partial \bar{\rho} \bar{u}_j}{\partial x_j} = \bar{\rho}^s \quad (1)$$

Momentum conservation equation:

$$\frac{\partial \bar{\rho} \tilde{u}_i}{\partial t} + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{u}_i \tilde{u}_j - \bar{\tau}_{ij} - \tau_{ij}^{sgs}) = \bar{F}_i^s \quad (2)$$

Energy conservation equation:

$$\frac{\partial \bar{\rho} \tilde{e}}{\partial t} + \frac{\partial \bar{\rho} \tilde{u}_j \tilde{e}}{\partial x_j} + \bar{p} \frac{\partial \tilde{u}_j}{\partial x_j} + \frac{\partial \tilde{h}_j^{sgs}}{\partial x_j} + \frac{\partial \tilde{q}_j}{\partial x_j} - \bar{\sigma}_{ij} \frac{\partial \tilde{u}_i}{\partial x_j} - \theta^{sgs} = \bar{Q}^s \quad (3)$$

Species conservation equation:

$$\frac{\partial \bar{\rho} \tilde{Y}_m}{\partial t} + \frac{\partial}{\partial x_j} (\bar{\rho} \tilde{Y}_m \tilde{u}_j - \bar{D}_m \frac{\partial \tilde{Y}_m}{\partial x_j} + \phi_{j,m}^{sgs}) = \bar{\dot{\rho}}_m^s \quad (4)$$

where $\bar{\rho}^s$, $\bar{F}_i^s$, $\bar{Q}^s$ and $\bar{\dot{\rho}}_m^s$ are the mass transfer, momentum transfer and energy transfer source term for the continuity, momentum, energy and species transport equation. The subgrid stress tensor τ_{ij}^{sgs} is modeled using the resolved strain rate, a subgrid eddy viscosity and a subgrid kinetic energy defined as $K^{sgs} = \frac{1}{2}(\tilde{u}_i \tilde{u}_i - \tilde{u}_i \tilde{u}_i)$. The Boussinesq eddy viscosity approximation is described as:

$$\tau_{ij}^{sgs} = -2\nu_t \left(\tilde{S}_{ij} - \frac{1}{3} \tilde{S}_{kk} \delta_{ij} \right) + \frac{2}{3} K^{sgs} \delta_{ij} \quad (5)$$

where the filtered rate of strain tensor $\tilde{S}_{ij}$ is defined as:

$$\tilde{S}_{ij} = \frac{1}{2} \left(\frac{\partial \tilde{u}_i}{\partial x_j} + \frac{\partial \tilde{u}_j}{\partial x_i} \right) \quad (6)$$

The subgrid eddy viscosity ν_t is calculated using a Smagorinsky-based model:

$$\nu_t = (C_s \Delta)^2 \tilde{S} \quad (7)$$

where Δ is based on the local grid size, the value of the coefficient C_s was set to 0.1 in this study, and the magnitude of the rate of strain tensor is expressed as:

$$\tilde{S} = 2(\tilde{S}_{ij} \tilde{S}_{ij})^{1/2} \quad (8)$$

Subgrid Kinetic Energy Equation

In this case, the subgrid kinetic energy K^{sgs} is obtained by solving the following equation proposed by Menon et al. [22].

$$\frac{\partial \bar{\rho} K^{sgs}}{\partial t} + \frac{\partial \bar{\rho} \tilde{u}_j K^{sgs}}{\partial x_j} = P^{sgs} - D^{sgs} + \frac{\partial}{\partial x_j} \left(\bar{\rho} \frac{\nu_t}{Pr_t} \frac{\partial K^{sgs}}{\partial x_j} \right) + \bar{W}^s \quad (9)$$

Here, the production term is closed by:

$$P^{sgs} = -\bar{\rho} \tau_{ij}^{sgs} \left(\frac{\partial \tilde{u}_i}{\partial x_j} \right) \quad (10)$$

And the subgrid kinetic energy dissipation rate is defined as:

$$D^{sgs} = C_\varepsilon \bar{\rho} (K^{sgs})^{3/2} / \Delta \quad (11)$$

The coefficient value C_ε was set to 0.916 in this study. For more details see reference [22].

Phenomenological Soot Formation Model

A phenomenological soot model [19, 20] is used to predict soot emissions in this study. The reaction scheme used in this model is listed on Table 1. The principle framework of the soot model is based on the detailed approach developed by Tao et al. [13]. Four competing processes, soot inception, surface growth, surface oxidation and soot coagulation, are simulated to determine the rate of change of soot mass and particle number density. These can be expressed by equations (12) and (13); a more detailed description of the model is given below.

$$\frac{d[C_{(s)}]}{dt} = \omega_{nuc} + \omega_{sg} - \omega_{O_2} - \omega_{OH} \quad (12)$$

$$\frac{dN}{dt} = \frac{dN_{nuc}}{dt} - \frac{dN_{coag}}{dt} \quad (13)$$

If the soot mass concentration and the particle number density are known, the particle diameter can be determined by:

$$d_p = \left(\frac{6}{\pi} \frac{C_s}{\rho_{soot} N} \right)^{1/3} \quad (14)$$

Table 1. Reaction scheme in the soot model

Step	Reaction	$k = A T^b \exp(-E/RT)$		
		A	b	E
		cm/(mol s)		kcal/mol
Nuc	$A_2 R_5 \Rightarrow 12 C_{(s)} + 4 H$	2.15e+04		5.0
Sg	$C_{(s)} + C_2 H_2 \Rightarrow 3 C_{(s)} + H_2$	3.50E+03	0.78	3.8
So	$C_{soot} H + O_2 \Rightarrow C_{soot-2} H + 2 CO$	Nagle and Strikland-Constable model		
So	$C_{soot} H + OH \Rightarrow C_{soot-1} H + CO + H$	Neoh et al. model, $\gamma = 0.13$		
Coag	$\kappa C_{soot} \Rightarrow C_{soot}$	Modified Smoluchowski equation		

Particle Inception/Nucleation

The first critical step in soot formation is soot particle inception or nucleation.

$$\frac{dN_{nuc}}{dt} = \omega_{nuc} \frac{dN_A}{C_{min}} \quad (15)$$

$$\omega_{nuc} = 12k_{nuc} [A_2R_5] \quad (16)$$

where ω_{nuc} is nucleation rate, N_A is Avogadro constant, C_{min} is the carbon number for nucleation, k_{nuc} is reactivation coefficient of nucleation. Soot particles are taken to be dry, graphite-like particles containing solely carbon atoms. Based on this assumption, the incipient soot particles are assumed to have a diameter of 12.5 nm, which corresponds to about 100 carbon atoms.

Surface Growth

Soot particle surface growth is assumed to be C_2H_2 -assisted:

$$\omega_{sg} = 2k_{sg} P_{C_2H_2} \left(\alpha \frac{\chi}{N_A} A_s N \right) \quad (17)$$

where ω_{sg} is surface growth rate, and k_{sg} is reactivation coefficient of surface growth, $P_{C_2H_2}$ is partial pressure of C_2H_2 , χ is active site density defined as Surovkin's coefficient [23], A_s is surface area of particle, and N is particle number density.

In this expression, α is an inactivation factor representing the fraction of the surface area that is available for surface growth reactions. It is defined using the expression proposed by Kazakov et al. [24]

$$\alpha = \frac{1}{2} \left[\tanh \left(\frac{8186}{T} - 4.57 \right) + 1 \right] \quad (18)$$

Surface Oxidation

The concentrations and effects of two oxidizing agents are considered in this work. Oxidation by O_2 attack is modeled using the Nagle and Strickland-Constable model [25]. This model assumes the existence of two types of reactive site on the carbon surface, with the rate of the surface chemistry being determined by their relative abundance. Oxidation by OH radicals is modeled by the method of Neoh et al. [26], using a value of 0.13 for the collision efficiency of OH radicals with the soot surface.

Particle Coagulation

Soot particle coagulation is a significant process that is known to take place before the exhaust stroke. In the present model this is described using the Smoluchowski equation:

$$\frac{dN_{coag}}{dt} = \frac{6}{5} \kappa f_v^{1/6} N^{11/6} \quad (19)$$

The rate constant of coagulation is expressed as

$$\kappa = \frac{5}{12} \left(\frac{3}{4\pi} \right)^{1/6} \left(\frac{6k_B T}{\rho_p} \right)^{1/2} G C_a \quad (20)$$

where f_v is the particle volume fraction, k_B is the Boltzmann constant, ρ_p is the particle density, G is the spherical particle factor, C_a is the self-preserving size distribution factor, and T is the ambient temperature. A more detailed discussion about the coefficients of the soot model can be found in the work of Adachi et al. [27].

Explicit Stiff ODE Solver

To facilitate the modeling of complex gas-phase chemistry processes, CHEMKIN-II has been integrated into the KIVA4 code. The implicit stiff solver DVODE is used to integrate ODEs in CHEMKIN. Since the Jacobian matrix is computed for each cell, however, significant computational time is spent on the chemistry. Therefore, to reduce the computational time spent modelling the chemical processes, a fast explicit stiff solver based on CHEMEQ2 code was coupled to the CFD code.

The CHEMEQ2 solver, developed by Mott et al. [15], is a single-step stiff ODE integrator that uses the α -QSS method of integration. It has the advantage that it requires only information relating to the current time step in order to integrate and advance to the next time step, which is not the case for many implicit stiff ODE solvers, such as DVODE. This unique feature of the explicit stiff solver minimizes start-up costs in multi-dimensional combustion simulations. Detailed descriptions of the CHEMEQ2 method have been presented by Mott et al. [15] Qureshi et al. [28] and Shi et al. [16]. In brief, however, the α -QSS method is based on the following two expressions:

$$\frac{\omega_i^n}{\rho} = \frac{q_i^n - p_i^n y_i^n}{1 + \alpha_i^n p_i^n \Delta t^n} \quad (21)$$

$$\alpha_i^n (p_i^n \Delta t^n) = \frac{1 - (1 - e^{-p_i^n \Delta t^n}) / (p_i^n \Delta t^n)}{1 - e^{-p_i^n \Delta t^n}} \quad (22)$$

where ω_i is the production rates of species i , q_i and p_i are the creation and destruction rates of species i , α is a function of $p\Delta t$ as in equation (22).