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Modeling of NO_x and Soot Formation in Diesel Combustion

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Résumé — Modélisation de la formation des NO_x et de la suies en combustion Diesel — Cet article présente une approche de modélisation de la formation et de l'oxydation ou de la réduction des suies et des No_x dans les flammes de diffusion turbulente. Le modèle repose sur l'étude des données d'une bibliographie de « flamelets », et intègre la prise en compte des pertes radiatives de chaleur dans la flamme. Comme les processus de formation des suies et NO sont relativement lents, une nouvelle approche modifiée est proposée. Au lieu d'extraire les concentrations de suies et NO directement de la bibliothèque, leurs termes sources sont calculés et une équation de transport pour les masses moyennes est résolue dans le code de mécanique des fluides. Les termes sources sont obtenus à partir de calcul en chimie détaillée sur une configuration de type flamme à contre-courant. Les équations de transports de la variable de mélange et de sa variance sont résolues, et le terme source chimique est obtenu par une technique de PdF présumé avec des fonctions bêta et une distribution log-normale pour la dissipation scalaire.

Le modèle a été testé sur des flammes de jet laminaires et turbulentes. En appliquant une stratégie de réduction des termes sources, il a été rendu applicable au calcul de la simulation des suies Diesel pour plusieurs températures et pressions de l'oxydant. De plus, différentes formulations d'équations de « flamelets » ont été testées et leur précision évaluée par rapport à une expérience de flamme turbulente.
Mots-clés : suies, No_x, flamelets.

Abstract — Modeling of NO_x and Soot Formation in Diesel Combustion *— An approach to model the formation and oxidation or reduction of soot and NO in turbulent diffusion flames is presented. The model is based on the flamelet library approach and extended to account for radiative heat losses in the flame. Due to the rather slow processes leading to soot and NO a modified flamelet library approach is used. Instead of taking the mass fractions directly from flamelet libraries the different source terms for soot and NO formation are calculated and a transport equation for the mean mass fractions is solved in the CFD calculation. The source terms are obtained from laminar counterflow-flame calculations using a detailed chemistry model for the gas phase species and the formation and oxidation of soot. Transport equations for the mean mixture fraction and the mixture fraction variance are solved and the chemical source term is closed by presuming a beta-function like distribution of mixture fraction and a log-normal distribution of the scalar dissipation rate.*

The model was first tested in laminar and turbulent jet flames. By applying a reduction strategy for the flamelet libraries of the source terms it was made applicable to the simulation of soot formation in a Diesel spray taking different oxidizer temperatures and pressures into account. Additionally, different formulations of the flamelet equations have been tested and their accuracy has been evaluated by comparing them to turbulent flame experiments.
Keywords: soot, No_x, flamelets.

INTRODUCTION

There are numerous model approaches used for the calculations of soot formation and oxidation in Diesel engines. The most recent approach is based on the laminar flamelet concept [1], solving so-called Representative Interactive Flamelets (RIF) on line to the CFD code [2,3]. A full description of the chemical processes as well as soot and

NO_x formation can be afforded without having to simplify the fluid dynamics, but it is time consuming, and might be difficult to incorporate in existing CFD codes.

Therefore a simpler approach based on stationary flamelet libraries of the sources of soot and NO formation and destruction was formulated [6]. This model represents, combined with a reduction strategy of the calculated flamelet libraries giving the flamelet libraries as algebraic functions, an attractive alternative for the simulation of pollutant formation in Diesel engines in terms of CPU time and computer storage requirement [7].

1 SOOT MODELING

1.1 Concept

The approach contains detailed submodels for the gas phase, growth and oxidation of the Polycyclic Aromatic Hydrocarbons (PAH), as well as particle inception, PAH condensation on the soot surface and particle growth, fragmentation and oxidation due to heterogeneous soot gas-phase reactions. The formation of aromatic species in two-dimensional diffusion flames has been calculated on the basis of stationary flamelets [4]. However, the accuracy of this approach was limited due to the fact that the time scale of benzene formation was of the same order as the time scale of the convective terms perpendicular to the flamelets. The typical time scales for soot formation are even longer. Therefore either a Lagrangian description of the entire flamelet is needed [2, 5] or additional transport equations for the moments of the soot size distribution functions have to be solved [6]. For the latter approach the source terms of soot in a two-dimensional diffusion flame can be calculated with a detailed soot model on the basis of the classical flamelet concept. However, the growth of soot particles is slow in comparison with other chemical reactions or transport processes in the flame. An additional transport equation has to be solved in the CFD code for the soot volume fraction. Both approaches make use of the fact that the formation of soot still occurs in thin layers, so that the flamelet assumption is valid. The numerical effort for the Lagrangian description of the flamelets in turbulent flames is high, limiting the number of representative flamelets used in the calculations. These flamelets will then be distributed statistically in the computational domain.

The numerical effort of the source term library approach can be minimized by the fact that the growth and oxidation of soot can be calculated with the same accuracy either proportional to the outer surface of spherical particles, or proportional to the inner surface of porous particles. This effect is due to the finding that the surface of soot particles has a fractal dimension. Hence the accuracy of calculations with only one statistical moment of the soot size distribution function, soot volume fraction, assuming that growth and oxidation are proportional to soot volume is as high as the accuracy of calculations with two statistical moments, soot volume fraction and soot number density assuming that the growth and oxidation are proportional to the soot surface of spherical particles.

The source terms of particle inception, surface growth and fragmentation, scaled by the local soot volume fraction, are stored in the flamelet library. Conventional flamelet libraries include the local scalar dissipation rate, the pressure, the composition of the fuel and the temperature on the air side of the flamelet or the enthalpy of the place of stoichiometric mixture. However, the libraries and interpolation schemes needed for the application in three-dimensional calculations set too high demands on computer storage and CPU time. Therefore the flamelet libraries for the source terms are reduced by fitting them to multi-parameter functions, resulting in simple algebraic equations and a parameter library. These are included as a source term in the transport equation of the soot volume fraction that is part of the CFD code.

1.2 The Detailed Chemical Soot Model

The soot model used in the calculations relies on a detailed description of the physical and chemical processes developed by Frenklach [8] and further developed by Mauss [9] (see Fig. 1). It can be subdivided into the gas phase reactions, the growth of PAH and the processes of particle inception, surface growth, condensation and oxidation.

The gas phase reactions are modelled using a detailed chemical reaction [9]. mechanism for the combustion of heptane. It includes 89 species and 855 reactions and, is based on the mechanism of Chevalier *et al.* modified with recent kinetic data. The different formation and destruction pathways of NO are included in the mechanism.

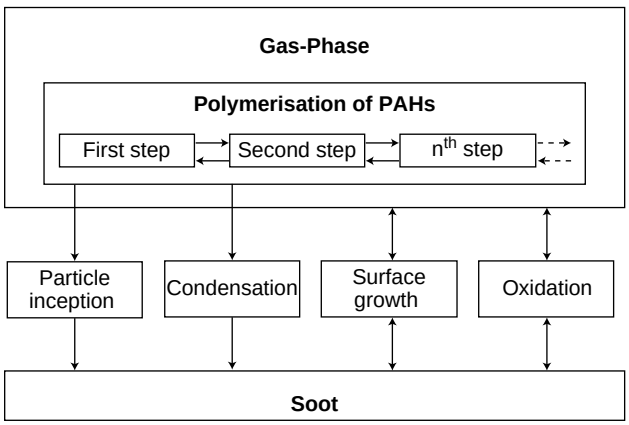

Figure 1

Illustration of the different processes involved in the formation and oxidation of soot.

Starting with benzene, a repeating cycle of hydrogen abstraction and acetylene addition that can be modelled as a fast polymerization process [9] describes the growth of PAH. Processes for particle inception, soot growth and oxidation are described by chemical and physical models. Physical models are applied to all coagulation processes, e.g. the coagulation of two PAH forming the first soot particles, the coagulation of PAH and soot particles describing the condensation of PAH on the soot particle surface. A chemical model is applied to the heterogeneous reactions occurring on the soot surface. Most of the soot mass growth is completed by surface growth of the soot particles by reaction with acetylene adding new aromatic rings in a similar manner as the PAH growth (HACA mechanism). The soot is oxidized by reaction with molecular oxygen and hydroxyl radicals.

The size distribution function of soot is calculated with the methods of moments proposed by Frenklach, where the moments are defined as:

$$ {}^{s}M_r = \sum_{i0,1}^{\infty} i^r N_i \qquad r = 0,1,\cdots,\infty \tag{1} $$

with:

N_i the density of particle *i* and

${}^{s}M_r$ the r^{th}- moment of the soot size distribution.

${}^{s}M_0$ represents the particle density and ${}^{s}M_1$ is proportional to the volume fraction of soot.

1.3 Flamelet Calculations

The flamelet concept proposed by Peters [1] was applied to uncouple the flow calculations from the combustion processes. Equations for energy and species conservation are formulated assuming fast chemistry and a thin reaction zone as a function of the mixture fraction and the scalar dissipation rate that accounts for non-equilibrium effects. The flamelet equations are obtained by a co-ordinate transformation of the equations from the physical into the mixture fraction-scalar dissipation rate space. These equations can be simplified by determining the leading order terms with the help of an asymptotic analysis. To evaluate the accuracy of different assumptions leading to the classical flamelet equations the source terms of soot and NO formation and oxidation calculated for the fully transformed set of flamelet equations were compared to the solutions of simplified equations. Best results were obtained assuming unity Lewis numbers in the outer layer of the flamelet and non-unity Lewis numbers in the thin reaction layer. Solutions obtained with different flamelet equations for the source term of surface growth are given in Figure 2.

Radiation from soot particles is of importance in the modeling of jet diffusion flames. Therefore the flamelet approach was extended to capture this important feature (details in [11]). The source term in the flamelet enthalpy equation accounting for radiation was multiplied with a factor α. Flamelet libraries for a variation of α were calculated accounting for different radiation levels. Radiation

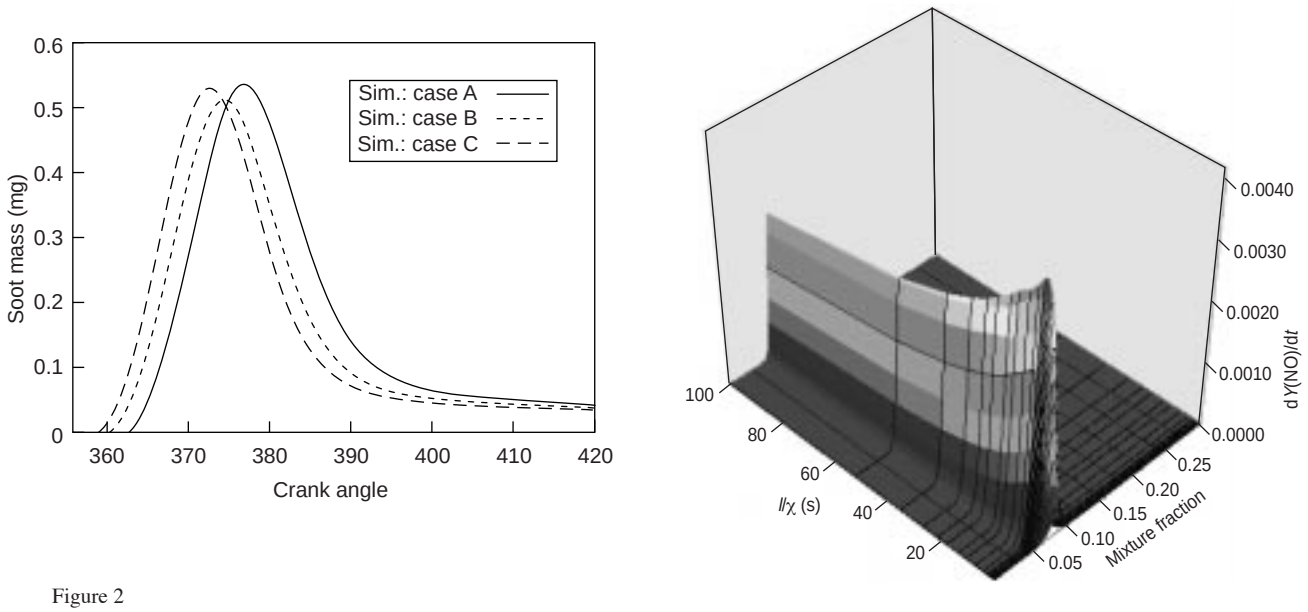

Figure 2

The source terms of surface growth for different flamelet formulations, the source term for NO in the mixture fraction-scalar dissipation rate space.

heat losses were calculated in the flow field and by comparing the enthalpy at each point in the flow field with the enthalpy in the flamelet libraries the correct library could be chosen.

The soot model was incorporated in the flamelet program and flamelet libraries were calculated using a modified arclength-method. The source terms of soot volume fraction, e.g. particle inception, surface growth, oxidation and fragmentation, as well as the source terms for NO (see Fig. 2) were stored in flamelet libraries as function of the mixture fraction and the scalar dissipation rate and additional parameters required, e.g. pressure and radiation factor.

To reduce the storage requirements and the CPU time needed for the interpolation of the source terms from the library, a simplification strategy was established [7]. The source terms are fitted successively to the parameters of interest, e.g. mixture fraction, scalar dissipation rate, pressure and oxidizer temperature, and formulated as algebraic functions of these variables. The accuracy of this approach was shown to be high by Mauss *et al.* [7] comparing the different source terms and the soot volume fraction in a calculation of soot formation in a laminar methane/air diffusion flame.

The normalized source terms of surface growth, oxidation and fragmentation and the particle inception are fitted to the mixture fraction in the first step of the simplification procedure. The beta function proved to reproduce the bell shaped curves of the source terms over the mixture fraction quite adequate.

Thus the different sources are formulated in multi-parameter functions and a set of parameters obtained by applying the simplification procedure to the flamelet library. Calculating the source from the multi-parameter function not only reduces the storage needed for the library but also speeds up the calculation within the CFD code.

2 CFD CALCULATION

Different CFD codes using different combustion models were used together with the submodel for soot and NO formation. Additional transport equations for the Favre-averaged mean and the variance of mixture fraction were added to the system of equations. The mean and the variance of the scalar dissipation rate were modelled according to [1]. The source terms for the transport equations of soot volume fraction and NO massfraction were closed by assuming a beta function like distribution for the mixture fraction and a log-normal distribution for the PDF of the scalar dissipation rate. The soot model could be implemented in the CFD codes without major changes being necessary to the main codes. The soot model part of the code consists of a separate subroutine in which the detailed flamelet library is summarized in the form of simple analytical functions as described above. These modifications to the code resulted in only a minor increase, approximately 20%, of the overall CPU time required for a run. The calculations become even faster by integrating the source terms algebraically since both the PDF of mixture fraction and the source terms are given as beta functions.

3 RESULTS

The flamelet library approach for the sources of soot formation and oxidation has been tested in laminar C_2H_2/air jet flame first [6]. The accuracy of the reduced flamelet library approach was evaluated in a CH_4/air laminar jet flame [7]. The approach was consecutively applied to a turbulent C_2H_4/air jet flame [11] including the radiative effects in the flamelet calculations as mentioned above. Turbulence was modeled with the κ-ε model while combustion was modelled via the flamelet approach. The soot volume fraction is calculated to be in good agreement with the measurements as shown in Figure 3. Major differences between prediction and measurements occur at low heights above the burner.

Soot formation and oxidation was simulated in a spray burning under Diesel engine conditions [10]. The software package used for spray and combustion calculations was the TAFF code. This code and models therein have been described in detail elsewhere. Three engine operating conditions, differing mainly by the injection timing, were used in order to study the performance of the soot model.

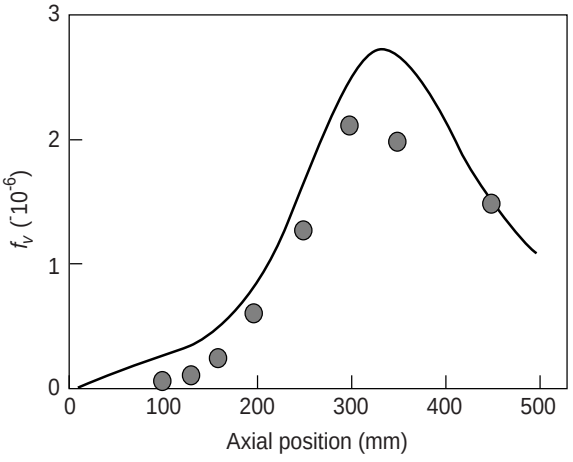

Figure 3

The calculated soot mass as function of CAD for different injection timings (left), calculated soot volume fraction at different heights above the burner compared to measurements in a turbulent C_2H_4/air jet flame (right).

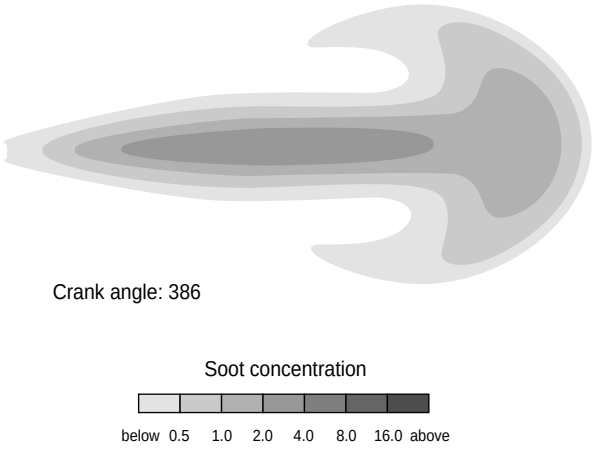

Figure 4

Distribution of the soot mass in the spray at CAD 386.

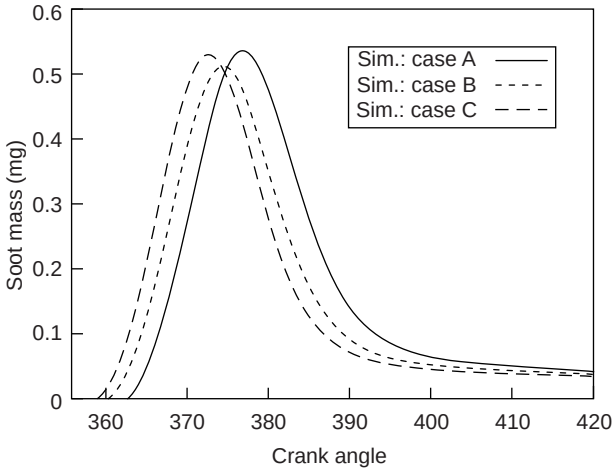

Figure 5

Time evolution of soot mass fraction.

These conditions all correspond to part load conditions for a Volvo D12A truck engine. The experimental data used in this work were obtained during in-house measurements using a one-cylinder research version of the Volvo D12A engine. The engine has a wide and shallow bowl design, "Mexican hat", and utilizes a low swirl combustion concept. A unit injector allows high injection pressures and gives good control over the injection timing and duration.

It is interesting to note that the results obtained by calculations are in qualitative agreement with the soot distributions measured by Dec and coworkers for similar engine conditions, see [13] and references therein. This means that maximum soot volume fraction is found in a broad area in the tip of the spray. At later times, however, the maximum soot volume fraction occurs at positions where the spray was evaporated, see Figure 4. Here the gas temperatures are lower than in the tip and the oxidation of soot is slower.

Further, the typical droplet penetration lengths calculated agree with those observed experimentally. Results shown in Figure 5 illustrate the time evolution of the total soot mass for the three operating conditions. The calculations were terminated at a crank angle of 420°. However, qualitatively by extrapolation, it can be observed that the calculated amounts of soot are in the correct range. Although the correct trends are predicted, the calculated final soot masses seem to converge towards a single final soot mass. This is in contrast to the measured results showing significant differences in exhaust soot masses. Since the presence of walls and other 3D effects are not taken into account in the calculations full geometry calculations have to be performed to get a better insight on the predictive capacities of the model.

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