Unsteady Flamelet Modeling of Soot Formation in Turbulent Diffusion Flames

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Unsteady flamelet modeling of steady turbulent diffusion flames has been shown in recent studies to yield good predictions for temperature, concentrations of major chemical components, including intermediates, as well as OH radical and NO concentrations [1, 2]. It has been shown that transient effects in steady jet diffusion flames have to be considered, if slow physical processes, such as radiation, or slow chemical processes, like the formation of NO_x , are important.

In the present study the formation of soot in a steady turbulent C_2H_4/air jet diffusion flame is modeled using the unsteady flamelet concept. The chosen configuration has experimentally been investigated by Kent et al. [3, Flame A]. In this experiment the nozzle diameter was 3 mm and the jet exit velocity 52 m/s, which leads to a fuel Reynolds number of 14660. Since the formation of soot is a slow process and both gas and soot radiation have a strong influence on the temperature in this flame unsteady effects are expected to be important in the investigated configuration.

The unsteady flamelet model applied in the present study is described in detail in Ref. [1]. The flamelet equation for the temperature is

$$\rho \frac{\partial T}{\partial t} - \rho \frac{\chi}{2} \left(\frac{\partial^2 T}{\partial Z^2} + \frac{1}{c_p} \frac{\partial c_p}{\partial Z} \frac{\partial T}{\partial Z} \right) + \frac{1}{c_p} \left(\sum_{k=1}^N h_k \dot{m}_k + \dot{q}_R^{\prime\prime\prime} - \mathcal{H} \right) = 0, \qquad (1)$$

where t denotes the time, Z the mixture fraction, T the temperature, χ the scalar dissipation rate, ρ the density, c_p the specific heat capacity at constant pressure, $\dot{q}_R^{\prime\prime\prime}$ the rate of radiative heat loss per unit volume. N is the number of chemical species, h_k the enthalpy, and \dot{m}_k the chemical production rate per unit volume of species k. \mathcal{H} accounts for the enthalpy flux by mass diffusion. The flow field has been calculated using the FLUENT code. To incorporate transient effects into the flamelet calculations, an unsteady flamelet has been solved interactively with the CFD solution. Since the boundary conditions of the flamelet, which are the temperatures and the composition of the fuel and the oxidizer stream, as well as the pressure, remain constant throughout the calculation, the only varying parameter influencing the flamelet solution is the scalar dissipation rate describing the impact of the turbulent flow field on the diffusion flame structure.

The unsteady flamelet has been calculated as a function of the flamelet time, which is related to the distance from the nozzle x as

$$t = \int_{0}^{x} \frac{1}{u(x') \left| \left(\tilde{Z} = Z_{\rm st} \right) \right|} dx', \qquad (2)$$

where \widetilde{Z} is the Favre average of the mixture fraction and $u(x) \left| \left(\widetilde{Z} = Z_{st} \right) \right|$ is the axial velocity component at the radial position, where $\widetilde{Z} = Z_{st}$, and the index st refers to stoichiometric conditions. Following Ref. [1]

the development of the scalar dissipation rate as a function of the nozzle distance is obtained by determining its conditional mean value in each computational cell and spatially averaging for each radial cell layer. The solution for the turbulent diffusion flame is then obtained by recalculating the unsteady flamelet after a certain number of FLUENT iterations. Convergence is achieved if the change of the scalar dissipation rate used for the flamelet calculations is smaller than a prescribed tolerance. Typically, two flamelet calculations yield a satisfying accuracy.

Since it has been found in many experiments in turbulent jet diffusion flames [4, 5, 6] that differential diffusion effects on species concentration and temperature appear only very close to the nozzle, where transient effects are unimportant because of the high scalar dissipation rates [1], the numerical simulations have been carried out with unity Lewis numbers for all chemical components.

The calculations have been performed with a detailed chemical reaction scheme compiled by Mauss [7]. The mechanism describes the oxidation of the fuel, the formation of higher aliphatic hydrocarbon species and benzene, and the growth of aromatic compounds up to pyrene.

The further growth of polycyclic aromatic hydrocarbons (PAH) is assumed to follow a fast H abstraction carbon addition (HACA) reaction sequence and is described with a model given by Mauss et al. [8]. The coagulation of PAH molecules forming three-dimensional structures is regarded as particle inception.

The further growth, the oxidation and the motion of particles is described by the solution of differential equations. For illustration purposes the Flamelet equations for the number density N_j of particles consisting of j mass units are here given in a simplified form as

$$\rho \frac{\partial N_j / \rho}{\partial t} - \rho \frac{\chi}{2 \text{Le}_j} \frac{\partial^2 N_j / \rho}{\partial Z^2} - 0.55 \frac{\chi}{2} \operatorname{Pr} \frac{N_j}{T} \frac{\partial^2 T}{\partial Z^2} + \frac{1}{4} \left(1 - \frac{1}{\text{Le}_j} \right) \frac{\partial \rho \chi}{\partial Z} \frac{\partial N_j / \rho}{\partial Z} - \frac{0.55}{4} \operatorname{Pr} \left(\frac{\partial N_j \chi / T}{\partial Z} \right) \frac{\partial T}{\partial Z} - \dot{N}_j = 0.$$

The chemical source term N_j includes contributions by particle inception, particle coagulation, condensation of PAH on the particle surface, and heterogeneous reactions of the particles with the gas phase leading to soot mass growth and oxidation. The diffusion coefficient of the particles, $D_{p,j}$, varies in the free molecular regime with d_j^{-2} [9], where d_j is the particle diameter of size class j. Thus, we can write $D_{p,j} = j^{-2/3}D_{p,1}$, and in terms of the Lewis number $\text{Le}_j = j^{2/3}\text{Le}_1$. To preserve generality we write

$$\operatorname{Le}_{j} = j^{\delta} \operatorname{Le}_{1}, \qquad (3)$$

where $\delta = 2/3$ and Le₁ is the Lewis number of the smallest particle if differential diffusion effects are considered. Unity Lewis numbers of all particles lead to $\delta = 0$ and Le₁ = 1

In order to solve Eq. 3, following Frenklach [11] the method of statistical moments is used here. Introducing the size dependent Lewis number for the particles, Eq. 3, and the statistical moments defined by

$$M_r = \sum_{j=1}^{\infty} j^r N_j \quad r = 0, \dots, \infty$$
(4)

into Eq. 3 the flamelet equations for the statistical moments can be obtained as [10]

$$\rho \frac{\partial M_r/\rho}{\partial t} - \rho \frac{\chi}{2\mathrm{Le}_1} \frac{\partial^2 M_{r-\delta}/\rho}{\partial Z^2} + \frac{1}{4} \frac{\partial \rho \chi}{\partial Z} \left[\frac{\partial M_r/\rho}{\partial Z} - \frac{1}{\mathrm{Le}_1} \frac{\partial M_{r-\delta}/\rho}{\partial Z} \right] - \dot{M}_j = 0,$$
(5)

which reduces to a flamelet equation of the species type if differential diffusion effects are neglected ($\delta = 0$). If non-unity Lewis numbers of the particles are considered, the terms with $M_{r-\delta}$ are small and only the first convection term remains as transport term.

Results of the calculation are compared to experimental data by Kent et al. [3] along the axis of symmetry and in three different radial slices. The first figure shows the mean temperature distribution along the centerline. The numerical results are in good agreement with the experimental data. Also shown are the results of numerical calculations, in which either soot radiation (dashed line) or soot and gas radiation (dotted line) has been neglected. The results show that both soot and gas radiation effect the temperature distribution by significant and comparable amounts, decreasing the maximum temperature each by approximately 200 K. This order of magnitude has already been reported by Kent et al. [3]. In the second figure radial profiles of the temperature are compared to experimental data, showing very good agreement at x/D = 46. Further downstream, the predictions close to the centerline still match the experimental data quite well. However, the radial temperature distribution in the outer lean part of the flame shows some discrepancies.

The following figures show predicted axial and radial distributions of the soot volume fraction compared to experimental data from laser light extinction measurements. The calculations have been performed accounting for non-unity Lewis numbers of the soot particles. The overall agreement is quite good. The maximum centerline value is almost identical. However, the predicted profile seems to be slightly shifted upstream, which has not been observed in the temperature profile. The agreement of the radial soot volume fraction profiles is also reasonable, especially, if the axial shift of the profiles is considered. The centerline soot volume fraction profile from a calculation with unity soot particle Lewis numbers is also depicted as the dash-dotted line. The maximum value is more than one order of magnitude too low, which seems to indicate that non-unity Lewis numbers of the soot particles have to be considered.



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