Effects of non-unity Lewis number of gas-phase species in turbulent nonpremixed sooting flames

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ABSTRACT

Turbulence statistics from two three-dimensional direct numerical simulations of planar n-heptane/air turbulent jets are compared to assess the effect of the gas-phase species diffusion model on flame dynamics and soot formation. The Reynolds number based on the initial jet width and velocity is around $15,000$, corresponding to a Taylor scale Reynolds number in the range $100 \leq \mathrm{Re}_\lambda \leq 150$. In one simulation, multicomponent transport based on a mixture-averaged approach is employed, while in the other the gas-phase species Lewis numbers are set equal to unity. The statistics of temperature and major species obtained with the mixture-averaged formulation are very similar to those in the unity Lewis number case. In both cases, the statistics of temperature are captured with remarkable accuracy by a laminar flamelet model with unity Lewis numbers. On the contrary, a flamelet with a mixture-averaged diffusion model, which corresponds to the model used in the multi-component diffusion three-dimensional DNS, produces significant differences with respect to the DNS results. The total mass of soot precursors decreases by 20–30% with the unity Lewis number approximation, and their distribution is more homogeneous in space and time. Due to the non-linearity of the soot growth rate with respect to the precursors' concentration, the soot mass yield decreases by a factor of two. Being strongly affected by coagulation, soot number density is not altered significantly if the unity Lewis number model is used rather than the mixture-averaged diffusion. The dominant role of turbulent transport over differential diffusion effects is expected to become more pronounced for higher Reynolds numbers.

1. Introduction

A strong consensus has been reached in the turbulence community regarding the effect of viscosity and molecular diffusivity on energy and scalar dissipation in turbulent flow. It has become empirically clear that, away from the walls, the mean dissipation rate of turbulent kinetic energy is independent of the fluid viscosity if the Reynolds number is high [1–6]. This hypothesis includes also the limiting case of zero viscosity (dissipation anomaly). The same concept applies to molecular diffusivity and scalar transport [7]: the mean mixing and scalar dissipation do not depend on the molecular diffusivity. This property suggests also that the details of molecular diffusion might be inconsequential for the diffusive transport in turbulent flows and have negligible impact on the scalar statistics provided the Reynolds number is sufficiently high.

On the other hand, the details of the molecular diffusion process, and in particular differential diffusion among different chemical species, play a remarkably important role in laminar flames and, more generally, in low-Reynolds number combustion [8–11].

In order to understand the physics of turbulent combustion and formulate appropriate reduced models, it is important to understand the effect of molecular diffusion processes on the dynamics of nonpremixed flames at high Reynolds number and assess whether the generally accepted paradigm in incompressible turbulence can be extended to turbulent combustion or not. The main question is whether the effects of differential diffusion of gas-phase species observed in laminar nonpremixed flames are overshadowed by turbulence at sufficiently high Reynolds numbers.

The issue is of primary importance in turbulent nonpremixed combustion modeling. The majority of models rely on the concept of mixture fraction, for which an advection–diffusion equation is solved together with a complex state relationship to compute the

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local thermochemical state from the mixture fraction. One of the main advantages is a reduction in the problem size. The number of partial differential equations to be solved decreases to no more then three or four from a number that can easily be of the order of fifty for a hydrocarbon flame in air. In addition, it is possible to formulate accurate models for the interaction between chemistry and the unresolved scales of turbulence in Reynolds Averaged Navier–Stokes (RANS) approaches and Large Eddy Simulation (LES).

There are a number of issues in these approaches related to differential diffusion. The definition of an appropriate mixture fraction might not be trivial in the presence of differential diffusion [12]. In addition, the state relationships between the chemical state and the transported quantities (mixture fraction and progress variable) are often built in laminar settings such as the counterflow configuration, and it is not clear if differential diffusion should be taken into account in these laminar calculations or not. While it could be concluded that including differential diffusion produces results closer to reality in laminar flames, it has often been argued that this may not be the correct choice for turbulent combustion closure. In flamelet models, it has become customary to neglect differential diffusion in the construction of the state relationship, in particular for temperature and major species [13,14]. This is justified by improved comparisons with experiments when unity Lewis number models are used as shown for example by Barlow et al. [15]. For a series of jet flames with increasing Reynolds number, they show an evolution from a scalar behavior governed by molecular diffusion to one dominated by turbulent transport.

Finally, it has been shown that a number of chemical species, such as the polycyclic aromatic hydrocarbons (PAH) responsible for soot formation and growth, are very sensitive to the local unsteady turbulent mixing process [16] and to differential diffusion effects, at least in laminar nonpremixed flames [17]. In addition, due to the strong sensitivity and nonlinearity of the soot formation processes to the local precursor concentration, even small effects observed on the precursor is magnified in the soot yields. These observation have important consequences in the modeling of turbulent sooting flames. In the context of Large Eddy Simulation of turbulent sooting flames, [18] adopt a transport equation for the naphthalene mass fraction to model the interaction between unsteady mixing and soot precursor dynamics. This approach allows to describe the unsteadiness of PAH kinetics directly and, if needed, to treat their transport using an appropriate diffusion coefficient.

A conclusive assessment of differential diffusion effects in turbulent nonpremixed flames, both sooting or not, does not appear in the literature due to the complications related to measuring the effect of differential diffusion in experiments and to the difficulty of achieving high Reynolds numbers in simulations of turbulent combustion [19].

In the present work, the effects of differential diffusion are analyzed by comparing two direct numerical simulations (DNS) that employ a detailed diffusion model with differential diffusion and a unity Lewis number approximation, in which differential diffusion is neglected. The analysis includes a comparison of temperature and major species statistics between the two DNS cases and with two laminar flamelet models computed with the same diffusion models employed in the DNS. Finally, the effects of species differential diffusion on the yield and morphology of soot, and on the dynamics of soot precursors are quantified.

2. Physical models and numerical methods

The gas-phase hydrodynamics are modeled with the reactive, unsteady Navier–Stokes equations in the low Mach number limit [20]. Combustion is modeled using a reduced mechanism for the oxidation of *n*-heptane comprising 47 species and 290 reactions, based on the detailed mechanism developed by Blanquart

et al. [21]. The details on the mechanism reduction and various validation cases relevant to *n*-heptane oxidation and benzene formation are available in [22].

The main focus of this work is the investigation of the effects of the diffusive transport model of the gas-phase species on the flame and soot statistics. Two cases with different modeling approaches are compared. The first simulation employs a mixture-averaged diffusion model. The Hirschfelder and Curtiss approximation to the diffusive fluxes [23] is used together with a velocity-correction approach [24] to enforce mass conservation. The species mass fraction and temperature equations read

$$\frac{\partial \rho Y_i}{\partial t} + \nabla \cdot (\rho \mathbf{u} Y_i) = -\nabla \cdot (\rho Y_i \mathbf{V}_i) + \dot{\omega}_i \tag{1}$$

and

$$c_p \left[\frac{\partial \rho T}{\partial t} + \nabla \cdot (\rho \mathbf{u} T) \right] = \nabla \cdot (\lambda \nabla T) - \sum_{i=1}^{M} c_{p,i} (\rho Y_i \mathbf{V}_i) \cdot \nabla T + \dot{\omega}_T. \tag{2}$$

$\mathbf{V}_i = \mathbf{V}_i^0 + \mathbf{u}^c$ is the mass-based species velocity and

$$\mathbf{V}_i^0 = -D_i \frac{\nabla X_i}{X_i}, \tag{3}$$

$$\mathbf{u}^c = -\sum_{i=1}^{M} Y_i \mathbf{V}_i^0, \tag{4}$$

where X_i and D_i are the mole fraction and mixture-averaged diffusion coefficient of species *i*, respectively. The thermal conductivity is λ; c_p is the mixture specific heat at constant pressure; $c_{p,\,i}$ are the specific heats for each species; and *M* is the number of species. The terms $\dot{\omega}_i$ and $\dot{\omega}_T$ indicate source terms for species mass fractions and temperature, respectively.

A number of analyses have been already reported for this simulation [16,25–28], and the reader is referred to these works for additional details on the flame and soot dynamics in the present flow.

For the second simulation, a simplified approach is used. The Lewis number, i.e., the ratio between the diffusivity of heat and that of species mass, is assumed to be unity for all gas-phase species. Because conductivity and density (and thermal diffusivity) are functions of temperature and mixture composition, the diffusivities of all species are equal, but depend on temperature and mixture composition. Soret and Dufour effects are neglected in both simulations.

Soot particles and aggregates are described by their volume (*V*) and surface area (*S*) [29], and the Hybrid Method of Moments (HMOM) of [30] is adopted to describe the evolution of soot. The bivariate soot moment $M_{x,\,y}$ is defined as

$$M_{x,y} = \sum_j V_j^x S_j^y N_j, \tag{5}$$

where *x* and *y* are the moment orders for volume and surface, and V_j, S_j, and N_j are the volume, surface area, and number density of soot aggregates belonging to size class *j*. Moments evolve according to

$$\frac{\partial M_{x,y}}{\partial t} + \nabla \cdot (\mathbf{u} M_{x,y}) = \dot{M}_{x,y}, \tag{6}$$

where $\dot{M}_{x,y}$ is a source term describing aerosol internal processes described below. Soot transport is characterized by a high Schmidt number and diffusive mass fluxes are therefore neglected. It is worth noting that this approximation is employed in both cases presented and the differences in the diffusion model pertain only to the gas-phase, while the differential diffusion between soot and the gas-phase is always present together with its strong