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A reduced multicomponent diffusion model

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ABSTRACT

The diffusion models for multicomponent mixtures are investigated in planar premixed flames, counterflow diffusion flames, and ignition of droplet flames. Discernable discrepancies were observed in the simulated flames with the mixture-averaged and multicomponent diffusion models, respectively, while the computational cost of the multicomponent model is significantly higher than that of the mixture-averaged model. A systematic strategy is proposed to reduce the cost of the multicomponent diffusion model by accurately accounting for the species whose diffusivity is important to the global responses of the combustion systems, and approximating those of less importance. The important species in the reduced model are identified with sensitivity analysis, and are found to be typically among those in high concentrations with exception of a few radicals, e.g. H and OH, that are known to participate in critical reactions. The reduced model is validated in simulating the propagation of planar premixed flames, extinction of counterflow non-premixed flames and ignition of droplet flames. The reduced model was shown to feature similar accuracy to that of the multicomponent model while the computational cost was reduced by a factor of approximately 5 for an *n*-heptane mechanism with 88 species.

1. Introduction

Accurate modeling of diffusion is essential in the simulation of the structure and response of flames, which consist of a multitude of chemical species strongly interacting with each other in narrow spatial domains of steep concentration gradients The task is monumental as the diffusion of a single species would depend on the properties and concentrations of all the species in the mixture. For a mixture of N perfect gas components, the Maxwell-Stefan multicomponent diffusion model, is derived from the Boltzmann's equation of kinetic theory [1–11] and recently revisited by Lam [12], offers the most rigorous, state-of-the-art description for combustion simulations. Its implementation, however, incurs significant computational cost due to the required matrix inversion, particularly when large reaction mechanisms are involved. As such, it is difficult to employ the multicomponent model in large-scale flame simulations. A simplified description is that of Curtiss and Hirschfelder [1], who obtained the solution to the first-order perturbations of the Boltzmann equation following the Chapman-Cowling procedure [3], and developed a mixture-averaged diffusion model to describe the diffusion of species in low concentrations. For many combustion systems, the mixture-averaged model offers rather good accuracy with substantially lower computational cost, and thus has long been the *de facto* standard in combustion simulations as reviewed by Smooke [13]. The multicomponent model is typically needed in such special situations when the Soret effect is of interest, for non-dilute mixtures or when high accuracy is required [14–23].

The effects of using the mixture-averaged vs. the multicomponent model on flame simulations have been extensively studied. For example, Bongers and De Goey [18] examined the laminar planar premixed flames for H_2–air, H_2–O_2 and CH_4–air, showing that the differences between the two models are discernable in predicting the flame speed. Gopalakrishnan and Abraham [19] investigated the ignition of *n*-heptane–air diffusion flames, and reported a 10% difference in the transient temperature and major species profiles. Dworkin et al. [24] studied soot formation in C_2H_4–air counterflow and coflow flames, and found that although the different diffusion models induce only small differences in the temperature profiles, they could lead to 10–15% deviation in the peak soot volume fraction. For premixed turbulent flames, Charentenay and Ern [25] simulated the interaction of H_2–O_2 flames with turbulence, and found that the two models result in rather different instantaneous profiles of temperature and species concentrations,

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especially for highly curved flames. However, turbulence fluctuations appear to diminish these effects when considering the mean quantities, leading to a rather small change of 7% in the thickness of the flame brush. Kumar and Mazumder [26] studied the heterogeneous combustion of H_2–air and CH_4–air mixtures in a monolithic channel coated by platinum, and found a 2% difference in the fuel concentrations at the exit of the channel. It is also noteworthy that Giovangigli and Ern [14–15,27] reduced the cost of the Maxwell-Stefan model from the mathematic point of view. They developed the iterative methods for solving the Maxwell-Stefan model. All transport coefficients are expressed as convergent series and approximated by the truncation of these series, as implemented in the EGLIB software package [28].

The objective of the present study is to develop a systematic approach to reduce the cost of the multicomponent model, hereafter referred to as the reduced multicomponent (RM) model, based on the physical understanding of the combustion systems. Its efficiency and fidelity is demonstrated for mixtures of *n*-heptane–air. Results show that the RM model retains the high accuracy of the multicomponent model and features a computational cost linearly proportional to the number of species.

2. Comparison of the diffusion models in combustion simulations

2.1. Simulation configuration

To compare the mixture-averaged and multicomponent models in different flames, three combustion systems of heptane–air mixture, namely the planar premixed flame, the counterflow diffusion flame and ignition of the droplet flame, are simulated, covering premixed and non-premixed, stretched and unstretched, and steady and unsteady conditions. The planar premixed flame and counterflow diffusion flame are simulated by using the algorithms of Kee et al. [29] and Nishioka et al. [30], respectively, while ignition of the droplet flame is simulated with the Adaptive Simulation of Unsteady Reacting Flows (A-SURF) code [31–35]. The chemistry of heptane oxidation is described by a skeletal mechanism of 88 species [36] derived from a detailed mechanism [37], which is available online [38].

The planar premixed flame is simulated over the range of parameters with equivalence ratios of 0.6–1.5, pressure of 1–40 atm, and freestream temperature of 300 K. Simulation of the counterflow diffusion flame is conducted for pure heptane against air with temperature of 300 K at both inlets, and pressure of 1–3 atm. Such parameters are selected to match the operation conditions of the flame speed measurement in [39] and the counterflow flame experiment in [40]. Each simulation starts with coarse grids and the mixture-averaged model. The grid is then refined iteratively until the solution converges. The diffusion model is then switched to the multicomponent model while the mesh is fixed, such that comparison of the models will not be biased due to changes in the mesh. For the unsteady droplet ignition, the droplet has initial radius of 1 mm and uniform temperature of 300 K. The ambient air is quiescent at temperature of 1400 K and pressure of 40 atm, being relevant to the working condition of IC engines [41]. The initial boundary layer at the surface of the droplet spans approximately 100 grid points, with temperature and species concentrations linearly interpolated from the droplet surface condition to the ambient condition. The evolution of the system is simulated by solving the 1-D, unsteady, compressible *N–S* equations, species equations and energy equation for the multicomponent reacting flow in the spherical coordinate. Thermal diffusion is not considered in all the simulations.

2.2. Simulation results with different diffusion models

Figure 1 presents the relative error of the mixture-averaged model in predicting the burning flux of the laminar premixed flame, f^0, as a function of equivalence ratio, ϕ, under different pressures. The relative error is 0.5–1% at atmospheric pressure, and increases to 2% at 40 atm. Compared with the typical uncertainty of 5% in experimental measurements of the flame speed [39], the mixture-averaged model results in only a minor deviation for the planar premixed flame.

Figure 2 illustrates the maximum temperature, T_{max}, in counterflow flames of pure heptane against air as a function of strain rate, κ, at pressures of 1–3 atm. It is seen that the extinction strain rates at the turning points predicted by the mixture-averaged and multicomponent models differ by 4% at atmospheric pressure and 6% at elevated pressures. Consequently, the counterflow diffusion flame is more sensitive to the selection of the diffusion model as compared to the premixed flame, in Fig. 1. Furthermore, based on the results in Figs. 1 and 2, deviation of the mixture-averaged model tends to increase at higher pressures.

In the simulation of unsteady droplet ignition, the fuel and air are mixed within a thin layer near the droplet surface through diffusion. Radicals build up during the induction period until approximately t = 20 ms, when thermal runaway is triggered. From 20 to 28 ms, the maximum system temperature rapidly increases

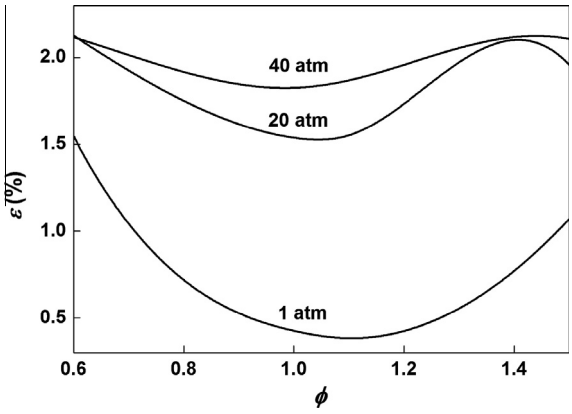

Fig. 1. Relative error of the mixture-averaged model in predicting the burning flux of planar premixed heptane–air flame as a function of equivalence ratio, at pressures of 1–40 atm and freestream temperature of 300 K.

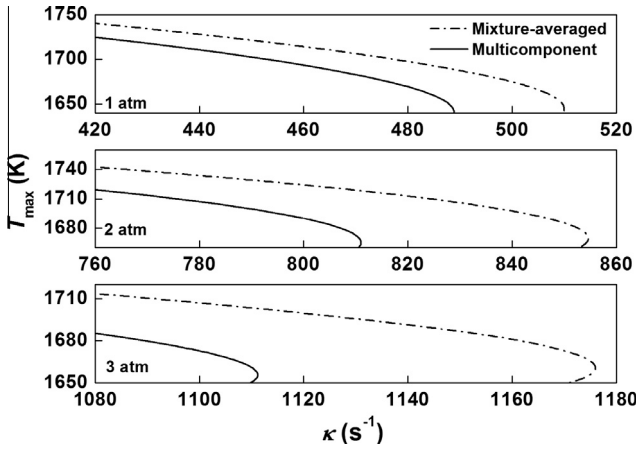

Fig. 2. The maximum temperature in the counterflow flame as a function of the strain rate, for pure heptane flowing against air at pressure of 1–3 atm and boundary temperature of 300 K at both inlets, calculated with different diffusion models. Dash dot line: mixture-averaged model; solid line: multicomponent model.