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**EFFECTS OF MOLECULAR TRANSPORT ON TURBULENCE-CHEMISTRY INTERACTIONS
IN A HYDROGEN-ARGON-AIR JET DIFFUSION FLAME**

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ABSTRACT

A numerical simulation of entrainment, turbulent advection, molecular transport and chemical kinetics in a turbulent diffusion flame is used to investigate effects of molecular transport on turbulence-chemistry interactions. A full finite-rate chemical mechanism is used to represent the combustion of a hydrogen-argon mixture issuing into air. Results based on incorporation of differential diffusion and variable Lewis number are compared to cases with the former effect, or both effects, suppressed. Significant impact on radical species production and on NO emission index (based on a reduced mechanism for thermal NO) is found. A reduced mechanism for hydrogen-air combustion, omitting both effects and incorporating other simplifications, performs comparably except that its NO predictions agree well with the case of full chemistry and molecular transport, possibly due to cancellation of errors.

INTRODUCTION

There is increasing experimental evidence [1-3] of the sensitivity of combustion chemistry in turbulent diffusion flames to molecular transport mechanisms in the flame zone. Previous models of the three-way coupling of flow, chemistry, and molecular transport mechanisms have involved idealized laminar configurations [4,5] or turbulence models [1,6,7] that do not account for the local time scales governing molecular mixing. To represent this coupling quantitatively in a model, the minimal requirements are (i) a validated chemical mechanism, (ii) a mechanistically literal treatment of molecular transport, and (iii) an accurate representation of the length and time scales governing the formation and strain history of the flame.

A model conforming to these requirements is applied here to the combustion of a turbulent jet of hydrogen-argon issuing into air. The chemical mechanism for this system is well established [8]. The treatment of turbulent flow and molecular transport is based on the linear-eddy model [9,10]. The formulation applied here incorporates the entrainment of air parcels into the jet, their breakdown by turbulent eddies, the molecular mixing of fuel and air, chemical conversion, thermal transport, and the coupling of density variations in the jet interior to the entrainment process [9]. Representation of all these processes is achieved by performing a stochastic simulation on a line corresponding to the jet centerline, with sufficient spatial and temporal resolution to allow mechanistically literal implementation of species and enthalpy transport. Limitation of the simulation to one spatial dimension allows computationally affordable simulation at full resolution for Reynolds numbers (Re) corresponding to typical laboratory experiments (jet Re of order 10^4).

To investigate the three-way coupling of flow, chemistry, and molecular transport, results based on a full implementation of the model are compared to cases in which the molecular transport mechanisms are simplified, either by suppressing differential molecular diffusion or by suppressing both differential diffusion and Lewis-number (Le) variations. A reduced chemical scheme [8] that omits differential diffusion and Le variations is also considered. Sensitivity of species concentrations, temperature, and NO emission index to the alternative formulations is examined.

MODEL FORMULATION AND NUMERICAL METHOD

The simulation model is based on Kerstein's linear-eddy model [9-11] which preserves the basic mechanistic distinction between convective transport and molecular transport by incorporating them as two concurrent processes along a one-dimensional line representing the centerline of the jet. Molecular transport (and chemical production) is incorporated by the finite-difference solution of the diffusion equation:

$$\frac{\partial Y_k}{\partial t} = \frac{w_k W_k}{\rho} - \frac{1}{\rho A} \frac{\partial}{\partial x} (\rho A Y_k V_k) \quad (1)$$

where Y_k , W_k , V_k and w_k are respectively, the k th species mass fraction, molecular weight, diffusion velocity and molar production rate. Here, ρ is the mixture density and A is the jet area. The diffusion velocity is approximated by [12]: $V_k = -(D_k/X_k) \partial X_k / \partial x$ where, X_k and D_k are respectively, the k th species mole fraction and mixture-averaged diffusion coefficient. For the cases with finite-rate kinetics, a one-dimensional thermal diffusion equation is also solved along with the molecular diffusion equation. Since chemical reactions and diffusion/mixing occur at the small scales, the strategy is to resolve all relevant length scales by using a one-dimensional representation of the streamwise evolution of the jet at its centerline. More details of this approach are given elsewhere [11,13].

Turbulent convection is represented by randomly occurring rearrangement events that punctuate the ongoing molecular transport and chemical kinetic processes. Each event is the model analog of a turbulent eddy. The event is a 'triplet map' applied to a randomly chosen segment of the computational domain, representing the folding and compressive strain effects of an eddy of that size on the concentration and thermal fields. The rearrangement process is formulated so that the sequence of events reproduces the eddy size-versus-frequency distribution for inertial-range turbulence, based on the Kolmogorov cascade picture. Again, for brevity, details discussed in the cited references are omitted.

The basic processes of convective and molecular (mass and thermal) transport and chemical kinetics are supplemented by application-specific processes that are designed to incorporate the specific aspects of the problem under study. The present formulation incorporates two application-specific processes: air entrainment and streamwise flow. Air is entrained in discrete parcels introduced at randomly chosen times and locations.

The dependence of parcel size and entrainment event frequency on location is determined by an empirical entrainment law [9] based on data correlations developed by Becker and Yamazaki [14]. The entrainment law incorporates buoyancy effects caused by combustion-induced density variations, thus introducing dynamic coupling of chemical heat release and turbulent mixing. The streamwise motion caused by thermal expansion (due to heat release), air entrainment, and fuel feed from the jet is incorporated as described earlier [9,11].

This formulation embodies the principal mechanisms causing large-scale and small-scale unsteadiness of the flow and of the combustion process, and the couplings among these mechanisms. The model is implemented as a Monte Carlo simulation, starting from an initial jet of air into air, switching to fuel feed at time zero, and igniting a few cells near the nozzle. After an initial transient, statistical stationarity is achieved. Species and temperature data are subsequently collected and reduced in a format akin to data collection and reduction in laboratory experiments.

Hydrogen-air kinetics is modeled [8] using a 10-step reaction mechanism supplemented by a three-step Zeldovich mechanism for thermal *NO* production. In all, there are 11 species (H_2 , O_2 , O , H , H_2O , OH , HO_2 , N_2 , N , NO , and Ar) diffusion equations and one equation for temperature that are solved simultaneously. Using simplifications such as equal diffusivity, $Le = 1$, and partial equilibrium of the bimolecular reactions, a reduced mechanism [8] in terms of a mixture fraction and progress variable has also been investigated.

The solution procedure for simulating both the full and reduced kinetics has been discussed elsewhere [13]. The reduced mechanism was implemented using a look-up table approach [8], while for the full kinetics, the CHEMKIN software package [12] was employed. An implicit predictor-corrector scheme was employed to advance the solution in time. To reduce the computational time, the code was implemented efficiently on a massively parallel computer, Kendall Square Research (KSR1) [13]. All finite-rate calculations were carried out on a 32-processor KSR1.

RESULTS AND DISCUSSION

The effects of molecular transport on the finite-rate kinetics of a H_2 -22%Ar jet flame is investigated. The test geometry is similar to a previous experiment [15] with fuel nozzle diameter $d_o = 0.0052$ m, fuel jet velocity of 75 m/sec and Reynolds number (based on nozzle diameter) of 9000. The fuel and ambient air temperature are initially 300 K and the pressure is 1 atmosphere. A streamwise domain extending to $x/d_o = 228$ has been discretized into 1250 cells, with the resolution scale increasing linearly with x . This discretization does not resolve the cold-flow Kolmogorov scale, but is adequate for the combustion application involving temperature-dependent viscosity. Adequacy of resolution is verified by examining instantaneous spatial profiles of species concentration and temperature in flame zones within the simulation.

Four cases are discussed here: (i) the reduced mechanism, and three cases with full finite-rate chemical mechanisms: (ii) $Le = 1$, no differential diffusion (Case A), (iii) variable Le , no differential diffusion (Case B), (iv) variable Le and differential diffusion (Case C). Thus, the effect of the partial equilibrium assumption in the reduced mechanism can be addressed by comparing with Case A. Lewis number (i.e., thermal conductivity) effects can be investigated using Case B results, while both thermal and molecular differential diffusion effects can be studied using Case C results.

Both time-averaged axial profiles and scatter plots at a chosen location are examined. Care needs to be taken when interpreting the time-averaged data due to the complex nonlinear interaction between air entrainment, mixing, heat release and thermal expansion processes simulated here. Differences between heat release in different cases must precede differences in mixing and entrainment, because variations in the mixing process are caused by thermally induced viscosity changes and entrainment rate variations are caused by thermally induced density changes [9]. Thus, when flow properties are time-averaged, differences among cases at small x must reflect differences in time-averaged heat release due to different transport mechanisms (Cases A-C) and chemistry (reduced case).

Keeping this in mind, we consider the streamwise variation of the time-averaged temperature (Figure 1a) and density (Figure 1b). High temperature (for Cases A and B) causes lower density (Figure 1b) which in turn causes enhanced buoyancy and entrainment. The mechanism leading from this to a faster temperature falloff for cases with

higher peak temperature (Cases A and B) can be interpreted as follows. The air entrainment flux at locations farther downstream is increased, increasing the dilution rate of the fuel and thus moving the crossover point from overall fuel-rich to overall fuel-lean mixture to smaller x . After this crossover, the main effect of dilution is to lower temperature and increase density. This explains the more rapid falloff of temperature for Cases A and B than for Case C. However, in Figure 1b, for large x , there is a recrossing because the higher density for Cases A and B in the range $40 < x/d_o < 120$ eventually decreases the entrainment such that Case C catches up and overtakes them. This occurs because the empirical entrainment law used here is nonlinear and time-lagged, and an additional time lag is introduced by the mixing process [9]. Although the entrainment law is based on time-averaged correlations that may not fully reflect the unsteady dynamics, it is at least indicative of the flow-chemistry interactions that are likely to occur.

Thus, the near-field sensitivity of the flow properties to transport is a direct effect of the molecular transport and chemical kinetics assumptions in Cases A-C and the reduced mechanism, but the far-field sensitivity largely reflects differences in overall stoichiometry due to entrainment effects.

The temperature profile for Case B reaches a higher maximum than the other cases. The difference from Case A is due to a reduction in the thermal conductivity in Case B, indicated by a rapid decrease in Le from 0.95 at $x=0$ to about 0.6 at $x/d_o = 40$. Slower diffusion of heat than for the $Le = 1$ case causes higher temperatures in flame zones, accelerating the exothermic reaction steps. This acceleration increases the average temperature in the near field, affecting entrainment and mixing rates and thereby inducing the far-field effects discussed earlier. Case C, with both variable Le and differential diffusion, has a lower maximum of the temperature than Case B, indicating that differential molecular diffusion tends to cancel the thermal conductivity effects.

The density (Figure 1b) and Y_{O_2} (Figure 1c) profiles indicate that Le and differential diffusion sensitivities, though magnified in the far-field due to turbulence-chemistry interaction, are not as important in the near field as the difference between full and reduced chemical-kinetic mechanisms. In the reduced mechanism, fuel and O_2 can react at 300 K, so mixing with hot products is not required for ignition. This causes underprediction of Y_{O_2} concentration in the near field. In fact, the mixing of O_2 and fuel is fast enough so that the fast reaction causes the Y_{O_2} concentration to be negligible

initially and causes the density to decrease initially as a function of x . In contrast, both of these quantities increase initially in Cases A-C as cold, dense O_2 is entrained and mixed, but is not consumed until it subsequently mixes with hot products. Significant deviations persist until about $x/d_o = 50$, where convergence of the reduced mechanism and Case C profiles occurs. In view of the near-field artifacts of the reduced mechanism, and their likely magnification due to turbulence-chemistry interaction, this convergence indicates a cancellation of errors rather than a mechanistically based performance improvement. This interpretation is supported by the persistence of temperature deviations (Figure 1a) in the far field despite a crossing of the reduced mechanism and Case C temperature profiles near $x/d_o = 50$.

A variety of other properties were analyzed. For brevity, only representative results are discussed. The profiles for the principal product Y_{H_2O} (not shown) follow the trend seen in the temperature profiles. In the near field, profiles for Cases A-C are nearly identical while the reduced mechanism case shows a larger concentration of water due to the instantaneous conversion of the reactants upon mixing.

Radical formation shows much greater sensitivity to molecular transport as well as chemical kinetic assumptions. The reduced mechanism overpredicts by a large amount the formation of H , O and OH while underpredicting significantly the formation of HO_2 . The overprediction of H (not shown) by the reduced mechanism is a result of the partial equilibrium assumption, because H radicals are instantaneously generated when fuel and oxidizer are mixed, irrespective of the temperature. This overprediction of H radicals is consistent with earlier partial equilibrium calculations [8].

Figures 2a and 2b show the OH and O mass fraction profiles, respectively. The OH profiles reflect the temperature differences seen earlier. The reduced mechanism overpredicts OH by the largest amount even where it underpredicts the temperature. This is consistent with the overabundance of H and O radicals for this case. Relaxing the partial equilibrium assumption (Case A) results in a reduction of OH radical, while relaxing the $Le = 1$ assumption (Case B) has a counteracting effect. This is consistent with the increased flame zone temperatures caused by reduced thermal conductivity. OH production is decreased when both Le and differential diffusion effects are allowed (Case C), indicating a cancellation of the effects of thermal and molecular diffusion on radical formation. The O profiles in Figure 2b show significant overproduction by the reduced

mechanism. Relaxation of partial equilibrium (Case A) reduces O production significantly. Case C predicts lower O radical than Case B partly due to reduced temperature and perhaps, as noted above, due to a counterbalance between thermal conductivity (Le) effects and molecular diffusion effects.

Scatter plots at $x/d_o = 30$ were also analyzed. All variables were conditionally averaged as described elsewhere [1] and the mixture fraction ξ was calculated according to the formula [1,8]:

$$\xi = \frac{2[H_2] + 2[H_2O] + 39.9[Ar]}{2[H_2] + 18[H_2O] + 32[O_2] + 28[N_2] + 39.9[Ar]} \quad (2)$$

where $[Ar]$ is assumed to be given by $[Ar] = \frac{.22}{.78}([H_2] + [H_2O])$ as done earlier [1].

With this definition, $\xi = 1$ corresponds to pure fuel and $\xi = 0$ to pure air, with $\xi_{stoic} = 0.1632$.

Temperature averages conditioned on ξ for Case B indicate a significant superequilibrium of temperature on the fuel rich side relative to the adiabatic equilibrium line (for $Le = 1$ and no differential diffusion). With differential diffusion included (Case C), the temperature values fall below equilibrium [13]. These results suggest that the superequilibrium of temperature is a direct Lewis number effect rather than a differential diffusion effect. For opposed flow, nonpremixed laminar flames, Barlow et al. [1] attributed superequilibrium temperature to differential diffusion effects, but they did not separate Lewis number (i.e., thermal conductivity) effects from species diffusion effects.

Strong differential diffusion effects are also seen in conditional averages of Y_{H_2O} , [13] with a large departure from equilibrium on the rich side for Case C. Figure 3a for OH mole fraction shows that all cases exhibit the experimentally observed [1,3] superequilibrium, with the peak OH occurring near stoichiometric mixture fraction. Superequilibrium occurs on both the lean and rich side and significant OH is found away from ξ_{stoic} , even at locations where the adiabatic equilibrium curve predicts zero OH mole fraction. Case B with variable Lewis number shows the largest departure. This is consistent with the higher temperature seen in this case. Although the reduced mechanism and Case A are in good agreement near stoichiometry, they begin to diverge in the lean limit. This is consistent with previous work [1], in which it was determined that partial equilibrium should be a good approximation near ξ_{stoic} while becoming inaccurate as the

local temperature drops below 1600 K for off-stoichiometric conditions.

Superequilibrium of OH is observed even farther downstream. Figures 3b and 3c show the scatter plot for Case C at $x/d_o = 30$ and 50, respectively. Farther downstream, superequilibrium occurs mostly on the lean side and the data ~~is~~ closer to equilibrium. These results are consistent with experimental observation [1] and with the notion that the increase of the local turbulent time scale for mixing and of the residence time with axial location allows more time for the radical to approach equilibrium.

Differential diffusion effects can be seen in the plots of mixture fraction based on atomic species H and O as a function of ξ . If there ~~is~~ no differential diffusion then all data would fall along the diagonal line of slope unity. The differential diffusion effect is minimal with respect to hydrogen [13]; however, there is a strong differential diffusion effect with respect to oxygen, especially on the rich side of stoichiometry (Figure 4). These results indicate the need for caution in interpreting data conditioned on mixture fractions defined by atomic hydrogen and oxygen.

The emission index for NO (EINO) is shown in Figure 5. Both Cases A and B show large values of EINO primarily due to the high peak temperature noted earlier and an abundance of O radical. EINO for reduced kinetics and Case C are surprisingly close. Further analysis shows that the assumption $[NO]/[NO]_{equil} \ll 1$ used to reduce the Zel-dovich mechanism [8] in the reduced kinetics case is satisfied in Case C but not in Cases A and B. More importantly, the reduced kinetics is based on partial equilibrium assumptions leading to overprediction of O radical (essential to thermal NO production), compensating for the temperature suppression (Figure 1a) caused by rapid entrainment (reflected in the rapid rise of the Y_{O_2} profile). Thus it appears that a cancellation of effects accounts for the agreement between the reduced kinetics case and the mechanistically more complete formulation.

CONCLUSIONS

A computational model incorporating simplified but plausible representations of molecular transport, convective transport, and chemical kinetic processes in a turbulent jet diffusion flame has been used to investigate the interactions among these processes. For hydrogen-air combustion, it is found that Lewis-number and differential diffusion effects impact the heat release rate in the near field, affecting the buoyancy of the jet and

thus the entrainment of air farther downstream. A two-way coupling between molecular processes and the large-scale development of the jet is obtained. The back-coupling is due to the effect of the entrainment rate on the overall stoichiometry at a given stream-wise location.

It is shown that this feedback process can either amplify or suppress errors introduced through model simplification. It is not obvious whether the specific trends noted here extrapolate, e.g., to higher Reynolds numbers for which buoyancy effects are reduced. Nevertheless, the results indicate the likelihood that the magnitude of the effects can be significant in practical situations.

Significant superequilibrium of OH mole fraction is obtained, as in experiments. The results for OH and other radicals are sensitive to molecular transport assumptions. Adoption of a reduced chemical kinetic scheme based on partial equilibrium leads to higher concentrations of H , O and OH radicals and lower HO_2 concentration than with obtained with full molecular transport and chemical kinetics (Case C). Overprediction of O tends to cause overprediction of NO . Predictions of NO emissions index based on the full and reduced formulations, respectively, are nevertheless in good agreement due to temperature suppression in the latter formulation, a consequence of the coupling between heat release and entrainment. This cancellation of O overprediction and temperature underprediction is sensitive to the strong couplings among the molecular, convective and chemical processes determining NO emission.

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FIGURE CAPTIONS

Figure 1. Time-averaged axial profiles of flow properties in a H_2 -22%Ar-Air turbulent jet diffusion flame. Fuel jet velocity is 75 m/sec and Reynolds number (based on jet diameter, $d_o = 0.0052$ m) is 9000. Four cases are shown: (i) reduced mechanism, (ii) $Le = 1$, no differential diffusion (Case A), (iii) variable Le , no differential diffusion (Case B), and, (iv) variable Le , differential diffusion (Case C).

(a) Temperature variation as function of x/d_o .

(b) Density variation as a function of x/d_o .

(c) Molecular oxygen mass fraction (Y_{O_2}) variation as a function of x/d_o .

Figure 2. Time-averaged axial profiles of radical species in a H_2 -22%Ar-Air turbulent jet diffusion flame. Conditions and cases same as in Figure 1.

(a) Hydroxyl mass fraction (Y_{OH}) as a function of x/d_o .

(b) Atomic oxygen mass fraction (Y_O) as a function of x/d_o .

Figure 3. Conditional-average and scatter plots of hydroxyl mole fraction as a function of mixture fraction (Eq. 2) for conditions and cases noted in Figure 1. Also shown is the adiabatic equilibrium curve (obtained with $Le = 1$ and no differential diffusion).

(a) Conditional-average of hydroxyl mole fraction at $x/d_o = 30$.

(b) Scatter plot of hydroxyl mole fraction at $x/d_o = 30$.

(c) Scatter plot of hydroxyl mole fraction at $x/d_o = 50$.

Figure 4. Mixture fraction based on atomic oxygen as a function of mixture fraction (Eq. 2) for variable Le and differential diffusion (Case C). Data obtained for a fuel jet speed of 75 m/sec and at $x/d_o = 30$.

Figure 5. Emission Index of NO (gm / Kg of fuel) as a function of axial distance for conditions and cases noted in Figure 1.

Figure 1a

Figure 1b

Figure 1c

Figure 2a

Figure 2b

Figure 3a

Figure 3b

Figure 3c

Figure 4

Figure 5

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