Brief Communication

Sensitivity of Calculated Extinction Strain Rate to Molecular Transport Formulation in Nonpremixed Counterflow Flames

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The counterflow flame is a commonly used geometry for experimental and chemical kinetic modeling studies of nonpremixed combustion. Because the flame structure is quasi-one-dimensional and more computationally tractable than inherently multidimensional flame geometries [1], detailed chemical kinetic modeling codes for counterflow flames have come into widespread use.

An important characteristic of counterflow flames is the extinction strain rate, the maximum velocity gradient which a flame can support and still burn. At strain rates below the extinction value, there exist three solutions to the governing equations which determine the flame structure. Two of the solutions correspond to a stable flame and an essentially non-reacting cold flow, while the third branch is an unstable solution which cannot be physically realized [2]. Various computational approaches have been used to extend stable solutions onto the unstable branch, and thus determine the limits of bistability corresponding to flame extinction and autoignition [2, 3].

The extinction strain rate is important for modeling turbulent combustion using the laminar flamelet approach [4], and is a figure of merit for the effectiveness of fire suppressants in nonpremixed flames [5]. It is desirable for calculations to accurately predict extinction strain rates. Substantial variations in predicted extinction strain rates of nonpremixed methane/air counterflow flames have been noted between different chemical kinetic mechanisms [6]. Here, the predicted extinction strain rate is found to also be sensitive to the treatment of molecular transport in the computational model.

The effect of transport on premixed flame structure and extinction has been recently investigated by Paul and Warnatz [7], and by Ern and Giovangigli [8, 9]. To our knowledge, the effect of transport formalism on extinction of nonpremixed counterflow flames has not been previously reported in the literature. Furthermore, earlier studies reporting computational predictions of extinction strain rates have not always specified the manner in which the calculation handled species transport.

In the present study, we employ two computer programs developed for counterflow flames incorporating the CHEMKIN program packages developed at Sandia National Laboratories. Initially, the chemical kinetics [10] and molecular transport [11] routines were applied to modeling one-dimensional premixed flames [12], and more recently, to counterflow flames [13]. The software developed at Sandia, including the counterflow code, OPPDIF, is available commercially [14]. Independently, Nishioka et al. adapted the Sandia premixed flame code for counterflow flames [2]. The code of Nishioka et al. has the capability to extend the stable flame solution past the extinction point onto the unstable branch, which the OPPDIF code does not. Extinction strain rates can be estimated using OPPDIF by slowly increasing the flow rates, relative to a previous solution, until the calculation returns a non-reacting solution [15].

The counterflow flame codes use routines which calculate molecular diffusivities and viscosities of multicomponent gas mixtures [11]. The two codes employ different versions of the same routines, both of which permit different options for treatment of molecular transport. One choice available to the user is use of mixture averaged diffusivities (weighted average of the binary diffusion coefficients) or of

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multicomponent values obtained by matrix diagonalization. Also, the influence of thermal diffusion on species transport (Soret effect) may be included or neglected for the species H and H_2.

In the results shown below, we have computed the structure of methane/air flames, on a domain of 1.5 cm using the code of Nishioka et al. Plug flow boundary conditions were used (radial velocity gradients set to zero at the boundaries). Axial velocities of the fuel and air flows were chosen to be equal at the two boundaries, with temperatures at both boundaries set to 300K. Central differencing was used on the convective term in the species conservation equations.

The reaction mechanism, species thermodynamics functions, and transport parameters of the Gas Research Institute GRI-Mech 2.11 [16] were used for all calculations. All nitrogen species except for N_2 were deleted from the mechanism.

Four series of calculations were performed: 1) multicomponent diffusivity formulation, with thermal diffusivity considered, 2) multicomponent diffusivity formulation, thermal diffusivity neglected, 3) mixture-averaged diffusivity formulation, thermal diffusivity considered, 4) mixture-averaged diffusivity formulation, thermal diffusivity neglected. For the mixture-averaged calculations, the "correction velocity" formalism was employed, which subtracts from the calculated diffusion velocities of each species any net convective velocity produced by the accumulated errors [11], to ensure species conservation.

Near the extinction condition, the "one-point temperature-controlling continuation" method described by Nishioka et al. [2] was used. The flame temperature at a single point within the reaction zone was specified as an input condition, and the flow velocities of reactants were allowed to vary (but were constrained to be equal to one another) to satisfy the temperature at the specified point. The specified temperature in each continuation run was typically lowered by some 5 K from the value obtained at the same grid point in the previous solution.

Figure 1 shows the peak flame temperature as a function of strain rate for a nonpremixed methane/air counterflow flame. The four series of calculations shown in the plot are identical except in the treatment of molecular transport. The calculations were performed on meshes having between 160 and 260 grid points, which were periodically refined as the strain rate was varied. For all calculations, local strain rates were determined by numerical differentiation of the calculated axial velocity with respect to position. We estimate the uncertainty of the strain rate determination due to the pointwise differentiation and the finite mesh at ± 5 s^{-1}.

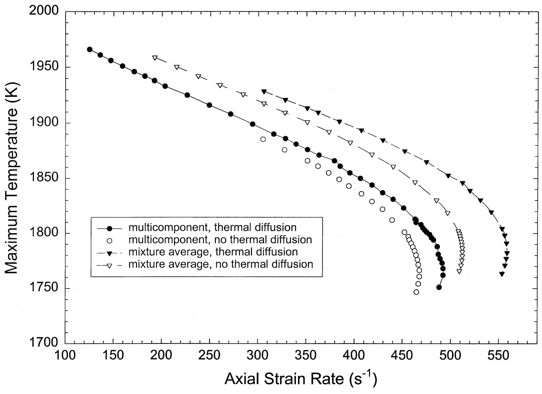

Fig. 1. Relationship between maximum flame temperature and peak air-side strain rate using the counterflow flame code of Ref. [2] with different options specified in the transport formulation. The extinction strain rates are given by the maximum values obtained as the peak temperature decreases.

Use of different transport treatments produces significant differences in the predicted extinction strain rates. The lowest and the highest of the values differ by almost 100 s^{-1}, ranging from approximately 450 s^{-1} to 550 s^{-1}. For comparison, most experimental measurements cluster around 400 s^{-1} [17]. The calculations using the multicomponent transport formulation predict a lower extinction strain rate than do those using the mixture-averaged approximation. For both types of calculations, inclusion of the Soret effect for H and H_2 increases the predicted extinction strain rate. Additionally, we have compared methane/air extinction strain rates estimated using the OPPDIF code for multicomponent and mixture-averaged transport formulations (neglecting the Soret effect), and find a similar sensitivity of extinction strain rate to transport treatment.

Figure 2 shows plots of temperature and H atom mole fraction as a function of position for

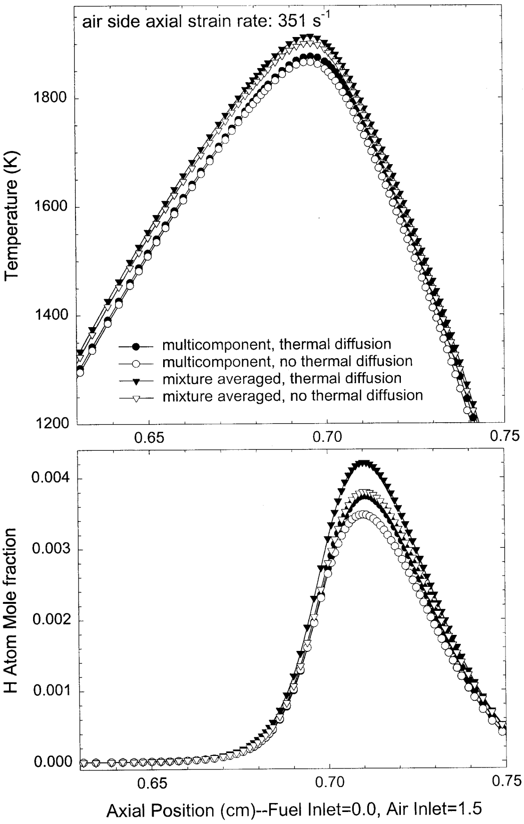

Fig. 2. Profiles of temperature and atomic hydrogen mole fraction in the vicinity of the reaction zone using the different molecular transport treatments. All four calculations used identical reactant flows; the maximum air-side strain rate for all calculations was 351 s^{-1}.

each of the four transport treatments at a strain of 351 s^{-1}. The temperature profile has a similar peak location and shape for all the transport treatments, as does the H atom profile. The peak H atom concentration varies substantially between the calculations with a difference of about 20% between the highest value (mixture averaged transport with thermal diffusivity) and the lowest value (multicomponent transport, no thermal diffusivity). The other two cases have peak H atom concentrations midway between these two values.

The present results demonstrate that use of different transport formulations has a substantial effect (on the order of 20%) on the predicted extinction strain rate of a nonpremixed methane/air flame. This implies that accurate treatment of molecular transport is a prerequisite to obtaining credible computational predictions of extinction strain rates of nonpremixed flames. The extension of these findings to other fuel/oxidizer systems is beyond the scope of the present work, but comparison of hydrogen and methane fuels in premixed flames [8, 9] shows that the former is far more sensitive to transport treatment.

The effect of transport also appears to be, in general, much more significant in a nonpremixed than in a premixed flame. A series of calculations (using the PREMIX code) of the burning velocity of a mixture of 15.8% O_2, 7.9% CH_4, and 76.3% N_2 yielded values which varied by less than 3% (between 22.5 and 23.1 cm/s) between the four treatments of transport considered here. Similarly, Ern and Giovangigli [9] found differences of only about 2% between the calculated extinction strain rates of premixed methane/air counterflow flames using mixture-averaged, multicomponent, and multicomponent-Soret transport formulations. The only nonpremixed flame modeled in Ref. [9] was an axisymmetric methane-air co-flow flame, in which the Soret effect was reported to have only a minor influence on flame structure. This result may suggest an influence of strain rate (typically higher in a counterflow than in a co-flow flame) on the relative importance of transport.

Using the mathematical formulation employed by the CHEMKIN codes, there is a significant penalty in computational time in using the multicomponent transport formulation. Both Ern and Giovangigli [8, 9] and Blasenbrey et al. [18], however, have proposed alternate formulations of multicomponent transport to reduce the computational expense.

The appropriate treatment of the Soret effect is a more problematic issue. The transport routines used for the CHEMKIN codes can include the Soret effect only for H and H_2, and neglect the Dufour effect (influence of species diffusion on thermal transport) entirely. The approach used by Ern and Giovangigli is to include both the Soret and Dufour effects for all species. In Ref. [9] Ern and Giovangigli comment that under certain conditions, the effect of thermal diffusion considered for all species can be opposite the effect when considered only for H and H_2. On the other hand, Paul and Warnatz [7] include the Soret effect only for H and H_2 on the grounds that the uncertainties are too large to justify its inclusion for other species. If the Soret effect does need to be included for all species, the transport formulations available in the CHEMKIN routines will have to be augmented in order to obtain credible predictions of extinction strain rates of nonpremixed flames.

Since the extinction strain rate of nonpremixed flames appears to be a property particularly sensitive to molecular transport, care is required in its modeling. Both the multicomponent formulation and the Soret effect significantly influence the predicted value, and the computational methodology must be specified to allow meaningful comparisons between modeling studies. The need for detailed consideration of the accuracy of the transport coefficients and the computational methodology, as done in Refs. [7–9], is underscored by the present results.

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