Combined Experimental and Computational Study of Laminar Axisymmetric H$_2$+N$_2$/Air Diffusion Flames
5.1 Introduction

The quantitative comparison of measured and computed major species and temperature distributions in laminar axisymmetric H\textsubscript{2}/N\textsubscript{2}-air diffusion flames is presented in this chapter. As in Ch. 4, the major species concentrations are measured using spontaneous Raman scattering, and the temperature both derived from the Raman data and measured independently using CARS. The governing equations are solved with detailed chemistry and transport; the sensitivity of the solutions to selected changes in transport properties, chemical reaction mechanism and flow conditions are considered. The experimental conditions were chosen to match the inlet boundary conditions used in the computations. In particular, heat transfer to the fuel tube was minimized by diluting the hydrogen fuel with nitrogen; this moved the reaction zone downstream so that the gradients in temperature and concentration at the exit of the burner are small. In Ch. 4, we attempted to obtain insight into flame structure by comparison with equilibrium approximations. Here, the focus is on assessing the adequacy of the computational predictions of the major species and temperature distributions in the diluted flames, and the interpretation of the results in light of some of the ideas discussed in Ch. 4. The discussion of the OH radical distribution in these flames will be presented separately in Ch. 6.

5.2 Combined experimental-numerical investigations of laminar H\textsubscript{2} diffusion flames: a brief review

Laminar axisymmetric coflow hydrogen-air flames have been used for the experimental study of Lewis number effects, such as the opening of flame tips [1], and for developing laser diagnostic methods for species and temperature determination in flame studies [2-4]. The emphasis in the diagnostic studies was on the adequacy of the diagnostic method, rather than on insight into flame structure, although one report [4] discussed possible premixing of fuel and oxidizer based on oxygen and nitrogen mole fractions measured in the stabilizing region of a hydrogen-nitrogen jet flame. Only a small number of studies have presented combined experimental and detailed computational results for the steady structure of these flames [5-8]. Measurements of OH concentration and rotational temperature, obtained by direct UV absorption [5], in axisymmetric H\textsubscript{2}-air flames were compared with simulations of flame structure using detailed chemistry, neglecting radiation and thermal diffusion, to assess the one-step-chemistry approximation, and to examine the stabilizing region. Another study [6] compared maximum radial temperatures obtained by solving the governing equations,
using complex chemistry and transport, with coherent anti-Stokes Raman scattering (CARS) measurements to assess the effects of thermal diffusion. Temperature profiles in H$_2$/N$_2$-air flames obtained using Rayleigh scattering were compared with numerical simulations neglecting axial diffusion [7] to examine differential transport in inverse diffusion flames (see also [9]). A recent report [8] compared species data obtained by laser-induced plasma spectroscopy with those using a simplified transport model; although, here too the object was to quantify the measurement technique. The analysis of the species fields in conjunction with the temperature field are essential to provide checks on the internal consistency of the experimental results, to yield insight into the structure of these flames and to appraise the adequacy of the computational results, as shown in studies on methane flames [10-13].

5.3 Numerical formulation and method of solution

The model of an axisymmetric diffusion flame considers an unconfined laminar flame in which a cylindrical fuel stream is surrounded by a coflowing oxidizer jet. Computationally, a velocity-vorticity approximation in which the elliptic two-dimensional governing equations are discretized on a two-dimensional mesh is used. The resulting nonlinear equations are then solved by a combination of time integration and Newton’s method. The Newton equations are solved by a preconditioned Bi-CGSTAB iteration. The grid points of the two-dimensional mesh are determined by equidistributing positive weight functions over mesh intervals in both the $r$ and $z$ directions. The size of the time steps is chosen by monitoring the local truncation error of the time discretization process (see also, Ref. [10]). Due to the cost of forming the Jacobian matrices with detailed transport and finite rate chemical kinetics, a modified Newton’s method is implemented along with several theoretical estimates that determine when a new Jacobian should be reformed. These theoretical results help increase the overall efficiency of the algorithm. The binary diffusion coefficients, the viscosity, the thermal conductivity of the mixture, the chemical production rates as well as the thermodynamic quantities are evaluated using vectorized and highly optimized transport and chemistry libraries [14]. Thermal diffusion is modeled in the trace light component limit (more comprehensive treatments are given in [15]). Radiative losses are included in an optically thin model. For the flames we consider, we assume that the only significant radiating species is H$_2$O [16, 17].

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Computations were made on adaptively determined grids containing approximately 15,000 nodes. To obtain this level of resolution on an equispaced grid, approximately $10^8$ grid points would be required. The computations were performed on a 13GB-RAM IBM RS6000 Model 44P-270 workstation. Each flame typically took several minutes of CPU time. The hydrogen-oxygen submechanism (nine species) contained in GRI 2.11 [18] was used in the computations. As will be seen below, additional calculations utilizing the hydrogen-oxygen submechanism in [10] produced small variations in the results.

5.4 Experimental Approach

The details of the burner setup, gas flow arrangement and optical configurations of the spontaneous Raman scattering, CARS and LIF experiments used in this study are provided in Ch. 2. For these diluted flames, three average fuel exit velocities (18, 27 and 50 cm/s) were chosen for the experiments. A parabolic velocity profile at the exit of the fuel tube was verified by hot-wire anemometry. In all the experiments, the velocity of the coflow air was set to the average velocity of the fuel jet.

5.5 Results and discussion

To illustrate the overall flame structure, two-dimensional false-color plots of the computed distribution of temperature at the three exit velocities are presented in Fig. 5.1. Here we see the characteristic high-temperature “wishbone” of the jet flame [10]. Similar to the methane/nitrogen flame in [11], the temperature rise in the diluted hydrogen flames occurs very close to the burner outlet. Further, the unreacted core of the jet is seen clearly, whose length also increases with velocity. It is interesting to note that the computed maximum temperature just above the exit plane occurs at a radial distance of nearly 6 mm, 1.5 mm outside the fuel tube; this indicates the extent
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Figure 5.1 Two-dimensional false-color plots of the computed distribution of temperature in the flames with three different exit velocities.

to which the radial diffusion of hydrogen causes the flame to spread, as seen earlier in the pure H$_2$ flames in Ch.4. As also observed in Ch. 4, we observe that the maximum

Figure 5.2 Adiabatic equilibrium composition and temperature calculated as a function of equivalence ratio for H$_2$ (50%)+N$_2$ (50%) fuel/air combustion
temperature increases substantially with exit velocity, yet the temperature never approaches the adiabatic stoichiometric temperature (~ 2023 K) calculated for this mixture, as can be seen from the equilibrium compositions and temperatures as a function of φ for this fuel in Fig. 5.2.

To follow the development of the flame, and allow more points of comparison with the computations, we present the radial profiles (r) of temperature and major species in the 50 cm/s flame, at axial positions (z) of 3, 10, 20 and 30 mm in Fig. 5.3-5.6, respectively. Both the experimental profiles and the computed profiles using GRI-Mech 2.11 are presented; since the addition or neglect of thermal diffusion had the largest effects on the computed results, both results are given. In these figures, the full experimental radial profiles, measured between r = -13 and 13 mm is shown, while the computed profiles are shown per half plane: the results obtained neglecting thermal diffusion are plotted for r ≤ 0, while the results including thermal diffusion for r ≥ 0. In the data shown, the symmetry of the experimental profiles around the centerline is sufficient for comparison with the computations.

At 3 mm above the exit of the fuel tube (the lowest axial position achievable for the Raman measurements), shown in Fig. 5.3a, the peak in the temperature, ~ 1800 K is at r = 6 mm, 1.5 mm outside the radius of the fuel tube, as mentioned above. The high temperature zone is thin: the width at half maximum is ~ 3 mm. Here, the core of the jet, where the temperature and composition are essentially those of the original mixture (Fig. 5.3b), extends for more than 1.5 mm from the centerline. As expected, the maxima in the temperature and water profiles coincide. The computed results reproduce the experimental profiles well; the results excluding thermal diffusion are nearly quantitative, while those with thermal diffusion are shifted to slightly larger radial distances. Results obtained in a high-velocity laminar jet [6] show the experimental data lying closer to calculations including thermal diffusion. In both cases in Fig. 5.3, the fuel-rich inner side of the jet is predicted quantitatively, while the results suggest that mixing on the fuel-lean side of the jet is underpredicted. This is evident in the water and oxygen profiles, for which both models overpredict the water
Figure 5.3 Radial profiles of the major species and temperature at 3 mm distance from the fuel tube exit in the flame with the average fuel exit velocity 50 cm/s. The solid and the dashed lines denote the calculations without and with thermal diffusion, respectively.

mole fractions and underpredict the oxygen on the lean side of the flame, although the data including thermal diffusion are more displaced into the coflow. Particularly interesting in Fig. 5.3b is the non-monotonic change in the nitrogen mole fraction. While simple mixing would suggest a monotonic change in the mole fraction from 0.5 in the jet to 0.79 in the coflow air, differential transport and the change in the number of moles within the flame combine to yield a complex profile. Both transport models capture this behavior well.
At $z = 10$ mm, shown in Fig. 5.4a, the width of the high temperature zone has increased to ~ 4.5 mm. Although the fuel on the centerline is still at room temperature, diffusion has reduced the $\text{H}_2$ mole fraction at this position to 0.43 and increased the $\text{N}_2$ mole fraction to 0.57 (Fig. 5.4b). Recalling the effects of differential transport shown in Ch.4, and anticipating the discussion below, the enrichment of the nitrogen mole

Figure 5.4 Radial profiles of the major species and temperature in at 10 mm distance from the fuel tube exit in the flame with the average fuel exit velocity 50 cm/s. In Fig. 5.4a, the symbols □ and ■ show the Raman and CARS temperatures, respectively. The solid and the dashed lines denote the calculations without and with thermal diffusion, respectively.
fraction by transport will have significant consequences for the attainable flame temperature when the fuel burns: the adiabatic stoichiometric temperature for this H₂/N₂ ratio is 1921 K, 100 K lower than the original fuel. In contrast with the pure fuel in Ch. 4, even though the H₂/N₂ ratio on the centerline has changed, there has been no transport of heat or water to the centerline. As was the case at z = 3 mm, the fuel-rich side of the flame is predicted nearly quantitatively, but the profiles without thermal diffusion are now ~ 0.6 mm wider than the measurements, while the predictions with thermal diffusion are ~ 1.2 mm wider than the measurements. Although both differences are outside the measurement uncertainty, the maximum discrepancy in the width of the profile is only 25%. At this axial position, differences in the predicted mole fractions at the centerline are also visible, though slight. Here too, the models capture the essential features of the nitrogen profile. The CARS data is also presented together with the Raman temperatures, showing the excellent agreement in the profiles, both in form and magnitude.

At z = 20 mm, in Fig. 5.5a, the centerline temperature has risen to 620 K, while the H₂ mole fraction has dropped to below 0.2 (Fig. 5.5b), and water has diffused to the centerline from the reaction zone. Whereas the centerline major species and temperature were well predicted at z = 3 and 10 mm, both models begin to show differences with the experimental data and with each other at this axial position. Interesting is that the centerline temperature excluding thermal diffusion is closer to the experimental data, but that the results for H₂ and N₂ are better predicted using thermal diffusion. As at the other axial locations, mixing on the fuel-lean side of the flame appears to be underpredicted by both models, more strongly by incorporation of thermal diffusion.
Figure 5.5 Distribution of the major species and temperature as measured and simulated at 20 mm axial distance from the fuel tube exit in the flame with average fuel exit velocity of 50 cm/s. The solid and the dashed lines denote the calculations without and with thermal diffusion, respectively.

These trends are continued at \( z = 30 \) mm, shown in Fig. 5.6a. Now the centerline temperature has reached 1100 K, and the \( \text{H}_2 \) mole fraction has decreased to nearly 0.05. Both transport models predict the centerline mole fraction of water well, and the neglect of thermal diffusion still yields better temperature predictions than those with
its inclusion. Again, the clear dip in the nitrogen profile in the high temperature region is reproduced very well by the computations. The combustion of hydrogen decreases the number of moles, so that the nitrogen concentration should actually increase in the reaction zone. Further, the peak temperature at this position, ~ 1800 K, is too low for a significant radical pool to increase the total number of moles. We also note that the peak temperature is 100 K lower than that at $z = 20$ mm; the fuel is nearly exhausted.
and dilution with coflow air is beginning to dominate the further development of the flame structure.

The overall consequences of the mixing field are illustrated in the axial centerline profiles for the 27 cm/s flame in Fig. 5.7. Here we see that the fuel is completely consumed by \( z = 25 \text{ mm} \), coinciding with the maximum in the water concentration. The temperature continues to rise slightly after this point, reaching a maximum of 1550 K at \( z = 30 \text{ mm} \). The agreement between measured and computed species profiles is within the limits of the experimental uncertainty, where the experimental data does not allow for discrimination between the two transport models.

![Figure 5.7 Axial profiles of the major species and temperature (measured and calculated) in the flame with the average fuel-exit velocity 27 cm/s. The symbols indicate the measured data, the solid lines indicate the computations neglecting thermal diffusion and the dotted lines indicate the computations including thermal diffusion effects. “Eqm T” indicates the temperature calculated based on equilibrium using the local element composition.](image)

The calculated temperature profiles are also in excellent agreement with the measurements until just past the maximum temperature; the difference between calculations and measurements increases progressively downstream, up to 120 K at \( z = 100 \text{ mm} \), as the hot gases are further diluted by the coflow air. Since the measured temperatures are more accurate than the species, the discrepancy is more apparent. Comparing the two transport models, we observe that thermal diffusion leads to a
consistently higher temperature, and that this difference is consistent with the higher water and lower oxygen mole fractions, as seen in the radial profiles above.

At this point, it is illustrative to compare the axial temperature profile with that expected at equilibrium. Whereas one would ordinarily characterize the mixture composition using the mixture fraction [19], the substantial differential diffusion in this system renders the definition of the mixture fraction problematical, as discussed above. Instead, as also discussed in Ch. 4, we calculate the equilibrium temperature using the element composition from the computed species profiles. To do so, the temperature of the species composition at a given axial position is varied until the enthalpy of the mixture is the same as the room-temperature mixture of H\(_2\), N\(_2\) and O\(_2\) having the same element composition. Since the mole fractions of all the computed species are used, including the flame radicals (whose concentrations were not known in Ch. 4), the temperature obtained includes any effects of “superequilibrium” radical concentrations; residual differences arise only from radiative losses and radial heat conduction. The equilibrium temperature calculated in this manner is also shown in Fig. 5.7, derived from the results without thermal diffusion. Whereas the peak equilibrium temperature is some 200 K higher than the detailed calculations, indicating significant heat transfer, the equilibrium temperature merges with the detailed curve downstream, where they remain within 10 K of each other until the end of the computational domain (z = 200 mm). As we observed in Ch. 4, since no energy has been lost from the system, we argue that the radiative losses in this system are negligible (see below). The maximum equilibrium temperature is ~ 1695 K, far below the 2023 K for the adiabatic stoichiometric temperature of the initial mixture burning in air. As suggested above, rapid radial diffusion substantially decreases the hydrogen mole fraction in the unreacted fuel; the equivalent H\(_2\)/N\(_2\)/air mixture having the same stoichiometric equilibrium temperature consists of slightly more than 31% H\(_2\).

Similar results are obtained at 50 cm/s, shown in Fig. 5.8. Here, too, the major species are well predicted, with only the suggestion of higher measured oxygen mole fractions at larger axial positions, at the limits of the experimental uncertainty. The temperature profile excluding thermal diffusion describes the rising edge of the profile well, but the disparity downstream between the measured and computed temperatures is larger than at 27 cm/s, up to nearly 200 K by z = 100 mm. Although clearly measurable, the maximum relative discrepancy in temperature at 100 mm is only 20%. Also interesting are the much smaller differences in predictions arising from the two models. As in Fig. 5.7, the calculated equilibrium temperature merges with the computations downstream of the peak, also indicating negligible radiative losses.
Computed results excluding radiation for this flame show indeed no contribution from radiative transfer to the temperature profile. Consequently, the difference between the computed temperature profiles and the equilibrium curves in Figs. 5.7 and 5.8 arises from heat conduction away from the centerline, since the local element composition would have a higher temperature under adiabatic conditions.

Figure 5.8 Axial profiles of the major species and temperature (measured and calculated) in the flame with the average fuel-exit velocity 50 cm/s. The symbols indicate the measured data, the solid lines indicate the computations neglecting thermal diffusion and the dotted lines indicate the computations including thermal diffusion effects.

The question arises as to the origin of the quantitative discrepancy between the computed and measured temperature profiles. Since one expects the hydrogen-air flame to be dominated by transport effects, and given the discussion concerning the challenges in calculating transport coefficients (for example [20, 21]), the sensitivity of the computed solution to transport properties, as well as to chemical mechanism and flow velocities was examined. The solutions obtained by: increasing the diffusion coefficients for H$_2$ and H by 50% with and without thermal transport, decreasing these coefficients by 25%, using another chemical mechanism (see above), varying the coflow by 20% and reducing the fuel exit velocity by 10% were compared. Only the changes associated with the incorporation of thermal diffusion altered the axial temperature profiles by more than 20 K, and these were to enhance the discrepancy with the measurements. The modest changes seen suggest more subtle differences in
the conditions used are responsible for the disparity with the measurements. Although the inlet conditions have been chosen to match the boundary conditions of the computation, we can anticipate that flow field variations between the experiment and the model can lead to the differences observed.

Similar to the study of the pure H\textsubscript{2} diffusion flames as described in Ch. 4, we scale the axial profiles by the average exit velocity, resulting in a residence time. The results are shown in Fig. 5.9, for temperature and mole fractions of H\textsubscript{2}O and O\textsubscript{2}. First, we note that the computed temperature and major species mole fractions (data without thermal diffusion), including those not shown, at the different velocities collapse to essentially a single curve. The largest differences are seen in the scaled temperature profiles, where the differences between the profiles are less than 30 K. In spite of complex differential transport coupled to chemical reaction, the development of the centerline profiles can still be characterized by the convective time scale. As also seen in Ch. 4, it is interesting to observe that the velocity scaling, used by Burke and Schumann [22] to estimate flame heights, scales the details of the axial profile here as well. This similarity appears to obviate the necessity of computing the flame structure for more than one velocity, at least regarding the centerline profiles. We also observe that the measured temperatures and major species collapse to one curve, within the

![Figure 5.9 Scaled axial centerline profiles. Points (□, ○ and *) denote measurements (for 18, 27 and 50 cm/s velocity flames, respectively); the solid lines denote the computed results for 50 cm/s and dashed lines for 27 cm/s.

\[ z/v (\text{m/s}) \]
\[ \text{mole fraction} \]
\[ \text{temperature, K} \]
\[ 0 \quad 50 \quad 100 \quad 150 \quad 200 \quad 250 \quad 300 \quad 350 \quad 400 \]
\[ 0 \quad 0.05 \quad 0.1 \quad 0.15 \quad 0.2 \quad 0.25 \quad 0.3 \quad 0.35 \quad 0.4 \quad 0.45 \quad 0.5 \]
\[ 0 \quad 300 \quad 500 \quad 700 \quad 900 \quad 1100 \quad 1300 \quad 1500 \quad 1700 \]
\[ T \]
\[ H_2O \]
\[ O_2 \]
experimental uncertainty; the scaling of the measured profiles is an important verification of the internal consistency of the results.

That the scaling exercise is of limited value for the details of flame structure can be ascertained by comparing the radial temperature profiles at the same “normalized” axial distance. At \( z/\dot{\varphi} \sim 33 \) ms, corresponding to \( z = 6.7 \) and 16.9 mm, for \( \dot{\varphi} = 18 \) and 50 cm/s, respectively, the radial temperature profile at 50 cm/s is 50% wider than that at 18 cm/s.

5.6 Conclusions

The experimental and computational results for major species and temperature in axisymmetric, laminar H\(_2\)/N\(_2\)/air diffusion flames have been discussed. In the axial and radial cross sections examined, the peak mole fractions and temperatures are predicted quantitatively, and the axial species profiles are predicted to within the experimental uncertainty. The computed radial profiles tend to be broader than those measured, and the predicted axial temperatures downstream of the maximum are significantly higher than the measurements. The relative discrepancies observed are however never more than 25%. Variation of transport properties for H and H\(_2\), and of flow velocities yields modest changes in the base solution, suggesting other causes for the discrepancies seen.

References
