Experimental study of the structure of laminar axisymmetric H2/air diffusion flames

Toro, Vishal Vijay

IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.

Document Version
Publisher's PDF, also known as Version of record

Publication date:
2006

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

Copyright
Other than for strictly personal use, it is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license (like Creative Commons).

The publication may also be distributed here under the terms of Article 25fa of the Dutch Copyright Act, indicated by the “Taverne” license. More information can be found on the University of Groningen website: https://www.rug.nl/library/open-access/self-archiving-pure/taverne-amendment.

Take-down policy
If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Downloaded from the University of Groningen/UMCG research database (Pure): http://www.rug.nl/research/portal. For technical reasons the number of authors shown on this cover page is limited to 10 maximum.
Structure of Laminar Axisymmetric Nonpremixed Coflow H₂/Air Flames
4.1 Introduction

The discussion of the flame structure of pure H\textsubscript{2}/air diffusion flames is presented in this chapter. As described in Ch. 2, the major species (H\textsubscript{2}, O\textsubscript{2}, H\textsubscript{2}O and N\textsubscript{2}) and the temperatures in these flames were measured using spontaneous Raman scattering and coherent anti-Stokes Raman scattering (CARS) techniques, respectively. The data obtained from the OH LIF measurements will be discussed separately in Ch. 6.

4.2 Experimental Approach

The laminar coflow H\textsubscript{2}/air diffusion flames studied in this work were stabilized at average fuel exit velocities (\(\bar{\vartheta}\)) of 9, 18, 27 and 50 cm/s. The major species and temperature data from these flames were collected along the axial centerline (z) and radial (r) directions, as shown in Fig. 4.1. The lowest height above the burner exit at which the Raman scattering and CARS measurements were conducted was \(z = 3\) mm. At \(z < 3\)mm, excessive scattered laser light interfered with the Raman spectra, while the very low concentration of nitrogen in this region also made the CARS spectra difficult to interpret.

![Figure 4.1 Axes of the profiles of the major species and temperature measured in the laminar coflow axisymmetric H\textsubscript{2}/air nonpremixed flames.](image)

In each flame, the axial centerline Raman and CARS profiles were measured in steps of 2 mm, while the radial profiles were measured at \(z = 3\) mm, 70 mm and the
axial distance where the centerline temperature reached its maximum ($T_{max}$). The radial profiles at these three positions provide insight into the flame structure in three distinctive zones: a region dominated by the fuel consumption ($z = 3$ mm), the point at which the stoichiometric surface reaches the centerline ($T_{max}$), and the post-combustion zone ($z = 70$ mm). Obtaining the species and temperature data in radial directions at only these three axial distances considerably reduced the experimental time required for the Raman measurements. (The $T_{max}$ position was determined from the axial centerline temperature profiles).

4.3 Results and discussion
4.3.1 Radial profiles
4.3.1.1 Data reduction

In this section, the results obtained in the radial direction at $z = 3$ mm, 70 mm and at $T_{max}$ will be illustrated using the flame at $\vartheta = 18$ cm/s. As mentioned in Ch. 2,
the maximum distance along the laser line over which the Raman data can be collected in a single experiment is roughly 13 mm. However, this measuring range is insufficient to cover the complete radial distributions of species and temperatures in the diffusion flames. Thus, to measure the entire profiles in the radial direction (covering ~ 25 mm distance), the burner was positioned at 0, +6 and -6 mm from the axis, the Raman images were collected at each position and pasted together. Figure 4.2 shows schematic representation of the burner positions at which the Raman data were collected. Here we see that the Raman CCD image captured at the burner position B shows the species distribution roughly symmetrically around the flame axis. The Raman images acquired at the three locations are shown in Fig. 4.3 and denoted by letters A, B and C, corresponding to the burner locations shown in Fig. 4.2. Figure 4.3

![Figure 4.3 CCD images at different radial positions; A: -6 mm, B: 0 mm and C: 6 mm from the centerline in the flame at 18 cm/s at z = 3 mm.](image)

also shows the complete radial distribution of the species obtained after combining the images A, B and C.

In the current experimental setup, the spatial resolution of the Raman measurements is ~0.21 mm/superpixel (the term “superpixel” denotes the combination of 4 binned pixels). Thus, when the burner is moved 6 mm from the central position (to A or C in Fig. 4.2), a particular point in the flame location shifts by 28 superpixels.
on the CCD chip. This pixel shift was used to paste the data obtained at the three positions.

![Figure 4.4 Radial profiles of water and temperature, obtained from the experimental Raman data shown in Fig. 4.3. Each property is denoted by its name, with the letter indicating the burner position (see Fig. 4.2) in parenthesis.](image)

To obtain the complete radial distribution of species mole fractions and temperature from the Raman data, the Raman intensities at locations A, B and C were fit using the Raman spectral fitting program described in Ch. 3, and the mole fractions and temperatures thus obtained were pasted together as described above. The results of this pasting exercise should be verified before analyzing the flame structure. Figure 4.4 shows the complete radial distribution of the H₂O mole fraction and temperature at \( z = 3 \) mm in the flame at \( \dot{\phi} = 18 \) cm/s, obtained from pasting the results from the three different burner positions. For the sake of clarity, each radial profile in Fig. 4.4 is shown using the different symbols, distinguishing three different burner positions (A, B and C). Each property is denoted by its name, with the letter indicating the burner position in parenthesis. We observe that the data for mole fractions and temperatures in Fig. 4.4 overlap substantially for adjacent burner positions. The smooth profiles indicate that the pasting procedure produces no gross errors. More importantly, in Fig. 4.4 the temperature profile obtained independently by CARS is indistinguishable from that defined by the Raman temperatures. Since the CARS profile was measured in the direction perpendicular to the direction of propagation of the Raman laser beam, this excellent agreement in both distribution and absolute values provides confirmation of the cylindrical symmetry of the flame, as well as additional confidence in the pasting.
procedure for obtaining the full radial profiles. Given the adequacy of the pasting procedure, we no longer distinguish between the different burner positions.

4.3.1.2 Results at $z = 3$ mm: qualitative effects of differential diffusion

Before proceeding to the analysis of flame structure, we briefly illustrate the major effect that transport processes have on the radial profiles. As discussed in the literature (particularly regarding turbulent flame structure) [1-7], given the low molecular weight of the fuel, we expect rapid radial diffusion of hydrogen to play a dominant role in the species and temperature profiles in the flames studied here. The consequences of this effect are illustrated in Fig. 4.5, which show the radial profiles of temperature and $\text{H}_2$ mole fraction at $z = 3$ mm for $\vartheta = 9, 18, 27$ and $50$ cm/s. At $9$ cm/s, the $\text{H}_2$ mole fraction on the centerline is only 0.4 (with mole fractions of water and nitrogen being 0.2 and 0.4, respectively); estimating the convective time scale using the centerline velocity of the cold gas (18 cm/s), allows ~16 ms from the burner exit for the disappearance of 60% of the fuel. Since the temperature here is only ~900 K, far too low for significant combustion, the composition and temperature at this point is solely due to transport processes. Increasing the exit velocity to 18 cm/s (~ 8 ms from the burner exit) increases the $\text{H}_2$ mole fraction on the centerline to ~0.85, while pure fuel is encountered at the centerline upon increasing the velocity to 27 cm/s (~5 ms downstream of the exit).

The rapid diffusion of hydrogen is also manifest in the temperature profiles. For $\vartheta = 18-50$ cm/s, the maximum temperature is at $r = 7$ mm; comparing this location with the inner radius of the fuel tube ($r = 4.5$ mm), we see significant radial flame spread. Transport of the fuel has resulted in placing the high temperature zone far outside the primary flow of fuel. The degree of flame spread at this axial position appears to be significantly larger than that observed methane-air flames [8]. At $\vartheta = 9$ cm/s, the peak temperature is closer to the centerline (near $r = 5.5$ mm); we suggest
Figure 4.5. Temperature and hydrogen profiles at $z = 3$ mm in flames with $\vartheta = 9$ (Fig. 4.5a), 18 (Fig. 4.5b), 27 (Fig. 4.5c) and 50 (Fig. 4.5d) cm/s.
that the combined effects of differential molecular diffusion and substantial radial heat transfer (giving rise to the 900 K temperature on the centerline) result in the broad temperature distribution in this relatively low velocity flame. Increasing the velocity visibly decreases the width of the high temperature zone. Also, we observe that the peak temperature increases progressively from ~ 1900 K to ~ 2200 K between 9 cm/s and 50 cm/s; however, the maximum temperature does not reach the adiabatic stoichiometric temperature of ~ 2380 K for pure H\textsubscript{2}. We shall discuss these observations further, below.

4.3.1.3 Results at \( z = 3 \text{ mm} \): discussion

For a more detailed analysis of the results, we restrict the discussion to the flames at \( \vartheta = 18 \) and 27 cm/s. In these flames, the exit velocity was high enough to minimize possible heat transfer to the burner, and to assure that the inlet conditions were essentially those of pure fuel and air at room temperature, while at the same time being low enough to maintain a stable flame over the entire measurement domain.

Returning to \( \vartheta = 18 \) cm/s, in Fig. 4.6 the data in Fig. 4.4 are repeated for the purpose of analysis. To aid in the visualization of the flame structure in space, the boundary of the fuel tube (\( r = 4.5 \text{ mm} \)) is also indicated in the figure. At this axial distance, the temperature distribution shows a maximum of ~ 2130 K at \( r = 7 \text{ mm} \) (Fig. 4.6a), well into the air flow as indicated above. This is also the location where the mole fractions of H\textsubscript{2} and O\textsubscript{2} are both very low (although not completely consumed), producing the maximum quantity of water. As mentioned above, the significance of radial transport can be observed in the species mole fractions and temperature at the centerline. Here, the hydrogen mole fraction is reduced to ~ 0.85 (Fig. 4.6b), while the temperature has increased to ~700 K. At this axial distance, substantial quantities of water (~ 5 mole%, Fig. 4.6a) and nitrogen (~ 10 mole%, Fig. 4.6b) are also seen on the axis. Since the temperature (700 K) here is too low for combustion to occur, the presence of water can only result from diffusion from the reaction zone. The same is true for N\textsubscript{2}, which is not present in the fuel.
Figure 4.6 Radial profiles of major species, temperatures and local equivalence ratio at \( z = 3 \) mm. Vertical dashed lines indicate the position of the inner wall of the fuel tube \( (r = 4.5 \text{ mm}) \).

To describe the mixing in the system, we can ascribe a local equivalence ratio to each point in flame (analogous to the use of the mixture fraction \([9,10]\)). Implicit in this approach is that we mix fuel and air in specific proportions, neglecting any difference in diffusivity. To do so, we can use \( \phi = \frac{[H]}{2[O]} \), where \([H]\) and \([O]\) are the total mole numbers of hydrogen and oxygen, respectively, in the mixture. Also
implicit is that the enthalpy remains constant. Therefore, to determine the temperature from the composition, we can solve \( \sum_i x_i H_i(T) = \text{constant} \), where \( x_i \) and \( H_i(T) \) are the mole fractions and (temperature dependent) enthalpies of species \( i \). The dependence of the equilibrium temperature and composition on \( \phi \) is shown in Fig. 4.7. As we can see at \( \phi = 1 \), the adiabatic temperature is \( \sim 2380 \) K.

![Figure 4.7 Adiabatic equilibrium composition and temperature as a function of equivalence ratio for H/O/N system at atmospheric pressure](image)

The local equivalence ratio calculated in this fashion is also presented in Fig. 4.6. We see here that the maximum measured temperature at \( z = 3 \) mm occurs near \( \phi = 1 \), and is \( \sim 2130 \) K; this is \( \sim 250 \) K lower than the equivalent adiabatic temperature. Furthermore, in this diffusion flame, where along the measurement path the composition varies from pure air to nearly pure fuel (essentially the full range of \( \phi \)), we see that the temperature nowhere reaches the maximum adiabatic value. This shows that the approach of assuming both thermodynamic equilibrium and equal diffusivity does not describe the system. In section 4.3.1.6 we attempt to relax these assumptions. A number of possible causes have been given in the literature \([1-7,10-13]\), mostly in connection with turbulent non-premixed hydrogen combustion: differential molecular diffusion, conductive heat transport, “fast-chemistry” effects (non-equilibrium radical concentrations) and radiative heat loss.
4.3.1.4 Results at $z = 20$ mm ($T_{\text{max}}$)

Progressing downstream, transport of hydrogen to and reaction in the flame zone ultimately exhausts the fuel [1]. At $z = 20$ mm, where the centerline temperature reaches its maximum, hydrogen is no longer detectable. The major species profiles at this position, given in Fig. 4.8, show only nitrogen, oxygen and water.

Figure 4.8 Radial profiles of major species and temperature at $z = 20$ mm.

The radial temperature distribution at this height is quite broad, with the maximum $\sim 2100$ K (Fig. 4.8a), as also observed at $z = 3$ mm. (Given the scatter in the
Raman data, we also show the CARS measurements at the same axial position (as another confirmation of the reliability of the temperature data). Up to several millimeters away from the centerline, water and nitrogen are the only species, and the equivalence ratio is thus stoichiometric, as indicated in the figure. Here too, the maximum measured temperature is more than 250 K below the stoichiometric adiabatic value assuming equal diffusivity.

4.3.1.5 Results at \( z = 70 \) mm

![Graph showing radial profiles of major species and temperature at \( z = 70 \) mm](image)

Figure 4.9. Radial profiles of major species and temperature at \( z = 70 \) mm
The results for \( z = 70 \) mm are shown in Fig. 4.9. Here we see that through mixing of the hot combustion gases with air the temperature has decreased to slightly above 1300 K (Fig. 4.9a), and the \( \text{O}_2 \) mole fraction (Fig. 4.9b) has increased to \( \sim 0.1 \). The local equivalence ratio (Fig. 4.9b) at the centerline has decreased to \( \sim 0.45 \). The corresponding temperature at adiabatic equilibrium is in excess of 1500 K, still 200 K above the measurements.

### 4.3.1.6 Deviation from the equilibrium temperature

- **Differential molecular diffusion**

As mentioned above, a number of phenomena have been suggested to account for the differences observed between the measured temperatures and those calculated at equilibrium. In [3, 7] differential molecular diffusion was suggested as a significant source of the differences between experimental temperatures and the predicted equilibrium values in turbulent diffusion flames at modest Reynolds numbers. We shall explore this possibility here.

The experimental data obtained provide sufficient information to estimate the degree of differential diffusion occurring in this system. Although it is obvious that differential diffusion of hydrogen is a salient feature determining flame structure, there is no simple method for discounting this effect directly in the analysis of flame structure; the proper consideration of such effects should be approached by detailed numerical calculation, as done in Ch. 5 for \( \text{H}_2/\text{N}_2 \) mixtures. However, we can use the other major species for this purpose. Considering the similar transport properties of nitrogen, oxygen, and to a lesser extent, water molecules, we would naively expect the elemental mole ratio of oxygen and nitrogen to be constant radially through the flame. To test this hypothesis, we calculate \( R \), the elemental ratio of oxygen to nitrogen, relative to that of air:

\[
R = \frac{2[\text{O}_2] + [\text{H}_2\text{O}]}{2[\text{N}_2]} 
\]

In the expression for \( R \), the numerator is the elemental \([\text{O}]/[\text{N}]\) ratio derived from the major species data, while the denominator value of 0.2666 from the \([\text{O}]/[\text{N}]\) ratio of air. The results using the data in Fig. 4.6 are shown in Fig. 4.10, together with the radial profile of equivalence ratio.
Although noisy, it can be seen that the measured oxygen/nitrogen ratio is not conserved across the flame; in the high temperature zone the ratio $R$ shown in Fig. 4.10 decreases to $\sim 0.8$. Given the similar transport properties of nitrogen and oxygen, their non-constant ratio is at first sight rather puzzling. However, we must recall that the species profiles and their various ratios are the consequence of the combination of multicomponent transport and chemical reaction. For example, in the high temperature zone water is the dominant oxygen carrier, which has a significantly different diffusivity than oxygen and nitrogen. Since water diffuses faster than nitrogen, we expect to find a lower concentration of oxygen-containing species (water) in the region in which it was formed, giving $R < 1$, and a disproportionate amount of oxygen-containing species on the centerline, giving $R > 1$. A similar result was observed in computations of turbulent flame structure [3]. Unraveling these effects in a microscopically correct fashion is beyond the scope of the current analysis [1,3]; here we restrict ourselves to providing insight into the consequences of the observed deviation of elemental ratio from the equal diffusivity value for the expected temperatures based on adiabatic equilibrium.

Since the initial condition of this system is a given three-element mixture, we can completely specify the composition of the mixture by using the equivalence ratio (ratio of H and O elements) and $R$. Therefore, at a given equivalence ratio, when $R \neq 1$, the equilibrium state can be considered to be at that equivalence ratio, but
mixed with “air” having a different oxygen/nitrogen ratio (either diluted with nitrogen or enriched in oxygen). When thus translated to the equal diffusivity approach, the mixture at \( R = 0.8 \) can be made by mixing fuel with “air” consisting of roughly 17\% \( \text{O}_2 \) and 83\% \( \text{N}_2 \). The effects of this “dilution” with nitrogen on the equilibrium flame temperature are shown as a function of equivalence ratio in Fig. 4.11 for \( R = 1, 0.8 \) and 0.6. Clearly, this effect can “depress” the temperature by several hundred degrees, similar to that derived in [3].

![Graph showing adiabatic equilibrium temperatures as function of equivalence ratios for different values of \( R \).](image)

Figure 4.11 Adiabatic equilibrium temperatures as function of equivalence ratios for different values of \( R \).

While flame structure is determined by complex interaction of chemistry and transport, the equivalence ratio is the most important parameter (and in many cases the sole parameter) characterizing the properties of the combustion products. We therefore present the experimental results as a function of the local equivalence ratio [9,10]. In Fig. 4.12, we plot both the adiabatic compositions and experimental data for the flame at 18 cm/s (similar results are obtained for radial profiles at other velocities). In this figure, we show the radial data obtained at all three axial positions, and for the purposes of illustration, have also included the axial centerline temperature profiles for both \( \dot{\phi} = 18 \) and 27cm/s. In Fig. 4.12a, the temperature data are given, while the mole fractions of nitrogen and water are shown in Fig. 4.12b. Since we are primarily interested in deviations observed in the reaction zone, we restrict the plots to \( \phi \leq 2 \); in the region around stoichiometric, the mole fractions of \( \text{H}_2 \) and \( \text{O}_2 \) are too low to be
discussed reliably. Also shown in the figures are the equilibrium values at \( R = 1 \) and 0.8.

One interesting observation can be made from plotting the data in this fashion: the temperature and composition data from different axial positions, from different cross sections (axial and radial), and even from (modestly) different exit velocities also appear to collapse (within the scatter) to a single curve. Thus, the temperature and composition of the system can also be described by the equivalence ratio, even though the data may not follow a given equilibrium curve. For \( \phi < 0.5 \), the temperature data follow the equilibrium curves exceptionally well, but in the region close to stoichiometric and above, the data are clustered several hundred degrees lower than at equilibrium. Parenthetically, with an eye towards estimating the local temperature in diffusion flames, the results in Fig. 4.12 suggest a useful correlation.

Examining Fig. 4.12b, we observe that with the exception of at the leanest and richest conditions, the nitrogen mole fraction is consistently above the \( R = 1 \) curve (as observed previously [2,10]) and in reasonable agreement with the predictions at \( R = 0.8 \) for \( 0.6 \leq \phi \leq 1.2 \) (as suggested by Fig. 4.10). The water is consistently below the curve for equal diffusivity, also closer to the curve for \( R = 0.8 \) in the same range of equivalence ratio. This suggests that we can use \( R = 0.8 \) to describe the compositional state of the system for this range of equivalence ratio, and have plotted the equilibrium temperatures using this value of \( R \) in Fig. 4.12a. In this figure, despite the scatter, we see that for \( \phi \geq 0.5 \) a systematically lower experimental temperature than predicted, even for \( R = 0.8 \), while for leaner conditions there is little difference between measurements and equilibrium predictions. These observations are in agreement with the trends predicted by an early laminar flame calculation [1] and discussed in [10]. In particular, the predicted temperatures from these calculations are also several hundred degrees below adiabatic equilibrium (for equal diffusivity), showing the same trend with equivalence ratio as observed here, and the predicted higher nitrogen mole fractions and lower water fractions are also consistent with our measurements. The depressed temperatures, similar to those observed here, were attributed [1] to complex
Figure 4.12. Temperatures, water and nitrogen concentrations as a function of local equivalence ratio.

heat and mass transport arising from the use of a realistic transport model, as well as to deviations from chemical equilibrium. In [10] the emphasis was placed on differential molecular diffusion.
Since the maximum temperatures are several hundred degrees lower than equilibrium, even when accounting differential diffusivity of the main components, we are compelled to conclude that, in this region, there must be other factors involved in determining the temperature.

- **Superequilibrium radical concentrations**

  Another possible cause for the difference between the measured maximum temperature and the maximum stoichiometric adiabatic temperature is the existence of superequilibrium radical concentrations, a “finite-rate chemistry” effect presented in Ch.1. This effect has also been cited as a possible source of subequilibrium temperatures in turbulent diffusion flames burning hydrogen as a fuel (for example in [2,11,12]), and included as a contributing factor in the analysis the numerical laminar diffusion flame structure [1]. Here, if the time scale for radical recombination (to equilibrium) is slow compared to the fluid time scale, the temperature obtained will be below the equilibrium value. To illustrate the effects of the superequilibrium radical concentrations on the maximum temperature, we examine the approach to equilibrium in a premixed stoichiometric H₂/air flame, calculated using the CHEMKIN II package [13], shown in Fig. 4.13. Although the fuel has already been nearly completely

![Figure 4.13 Calculated profiles of temperature and OH mole fraction in stoichiometric H₂/air flame](image_url)
consumed, here we see that the temperature at 3 mm axial distance is \( \sim 2130 \) K, and approaches the equilibrium value at the height of a few centimeters. In this same region, the OH mole fraction (being representative of the flame radicals) decreases from 1.4\% at 3 mm to 0.8\% (nearly the equilibrium value of 0.7\%) at 10 cm. In the premixed flame, this relaxation to the equilibrium value occurs on the order of 10 ms, of the same order as the fluid time scale (\( \sim 16 \) ms, based on the average cold flow velocity) in the diffusion flame, so that it is not unreasonable for finite-rate chemistry to be manifest. Similar magnitudes have been estimated previously in regard to turbulent flames [12]. Thus, excess radical concentrations that are roughly 0.005 mole fraction above equilibrium can also account for several hundred degrees of temperature decrease. We shall return to this aspect in Ch. 6.

### 4.3.2 Major species and temperature data along the axial centerline

In the previous section, we discussed the radial profiles at 3 axial positions. To complete the picture, we consider the development of the centerline profiles as we progress downstream. As we can see from the data presented in Figs. 4.14, the fuel is completely consumed by \( z = 20 \) mm, where the measured temperature (Fig. 4.14a) and water concentration reach their maximum, consistent with the results discussed above. Above \( z = 25 \) mm, we see the increase in O\(_2\) mole fraction and decrease in the H\(_2\)O mole fraction, reflecting the dilution of the hot gases with the coflow air. We note here the decreased scatter in the results compared to the radial profiles, partially obtained through the use of 3 times more pixels (0.6 mm spatial resolution) for these profiles.
Figure 4.14 a) Axial profiles of temperature and water mole fraction for $\vartheta = 18$ cm/s b) axial profiles of other major species and equivalence ratio.

Figure 4.15 plots the equivalence ratio and $R$ derived from the axial data. With substantially less scatter than that obtained in the radial profiles, we see that along the centerline $R$ decreases to $\sim 0.8$ at $z = 20$ mm (maximum temperature) to an apparent minimum of $\sim 0.7$ at $z = 30-40$ mm, after which $R$ increases to $\sim 0.8$ near $z = 70$ mm. We remark in passing that these values for $R$ are in excellent agreement with those
estimated in the radial profiles, despite the scatter in the radial data as shown in Fig. 4.10. The observation of an apparent minimum suggests that the “separation” caused

![Graph](image_url)

**Figure 4.15.** Axial profiles of $R$ (denoted by symbol ▲) and $\phi$ (—) for $\phi = 18$ cm/s flame.

by differential molecular transport coupled to chemical reaction in the flame zone is slowly relaxing to the uniform distribution expected at full thermodynamic equilibrium, as it ultimately should. Referring to the comparison between measurements and equilibrium calculations (Fig. 4.14a), at low axial positions (in the fuel-rich core of the hydrogen jet) the two equilibrium calculations give similar results, with a difference of ~200 K with the measurements. Progressing above $z = 5$ mm, the calculations and measurements diverge rapidly, increasing to a difference of several hundred degrees. We remark that in this region, where both the composition and temperature at the centerline are expected to be solely determined by transport processes, caution must be exercised when relating the composition and temperature of the gas to chemical equilibrium. In spite of this caveat, it is still illustrative to consider a possible degree of non-equilibrium. To do so, we have calculated the structure of the premixed H$_2$/air flame at $\phi = 4$, close to the equivalence ratio at $z = 5$ mm. In Fig. 4.16, we show the profiles of temperature and hydrogen atoms in this flame. At this equivalence ratio, the OH mole fraction is expected to be
very small and we use the H-atom mole fraction as an indication of the size of the radical pool.

![Figure 4.16 Calculated temperature and H atom concentration profiles for premixed H₂/air flame at φ = 4](image)

Here we see that even in this very rich flame the maximum H atom concentration is above 2.5% at flame temperature of ~ 1300 K, decreasing to < 10⁻³ at a temperature of ~ 1560 K (equilibrium). Thus, finite-rate chemistry could be active in the fuel-rich regions of the diffusion flame as well, contributing to the large discrepancy seen at rich equivalence ratios in Fig. 4.12a. Clearly, experimental observation of such high concentrations of hydrogen atoms would help further clarify the contribution of finite chemistry to the temperature depression in these flames. Further, as discussed above, differential diffusion fails to account for the “depressed” temperature around the stoichiometric equivalence ratio, and we must consider other mechanisms, such as superequilibrium OH radicals (see Ch. 6).

Perhaps more importantly, for general ideas about the structure of diffusion flames of pure hydrogen, as we progress downstream we see that, while the equilibrium temperature assuming equal diffusivity remains more than 200 K above the experimental data, the equilibrium temperatures accounting for the local element ratio (local $R$) merges with the measured temperatures near $z = 30$ mm, and remains in
agreement for the rest of the measurement domain, over a region of 600 K. This excellent agreement in the post-flame region excludes radiation as an important heat loss mechanism under these conditions. Furthermore, this agreement demonstrates the importance of differential molecular transport in the thermal structure of these flames.

For completeness, we show the axial profiles of water and temperatures (measured and calculated) at $\vartheta = 27$ cm/s in Fig. 4.17. Regarding the relationship between the measured and calculated temperature profiles, we see the identical behavior to that at $\vartheta = 18$ cm/s (as observed in Fig. 4.12a): the temperatures assuming equal diffusivity are always several hundred degrees above the measurements, while the profile accounting for differential molecular transport merges with the experiments just downstream of the maximum temperature, and remains there for the rest of the profile. Here too, we can only underline the importance of differential diffusion and the absence of radiative losses in these flames.

![Graph showing axial profiles of water and temperature for $\vartheta = 27$ cm/s.](image)

**Figure 4.17. Axial profiles of water and temperature for $\vartheta = 27$ cm/s.**

### 4.3.3 Velocity scaling

It is illustrative to normalize the axial profiles by the average exit velocity, resulting in a residence time, as suggested by the Burke-Schumann analysis [15]. The results are shown in Fig. 4.18, for temperature and mole fractions of H$_2$O and O$_2$. First, we note that the scaled temperature and major species mole fractions at the
Chapter 4

4.18 Scaled axial centerline profiles for 18 (●) and 27 (+) cm/s flames

different velocities yield nearly identical profiles. Further, for this rather limited variation in velocity (factor 1.5), despite complex differential transport coupled to chemical reaction, the development of the centerline profiles can still be characterized by the convective time scale.

4.4 Conclusions

This chapter presents the experimental data of the major species and temperature measured in laminar coflow non-premixed H₂/air flames and the discussion of the observed flame structure. We have obtained good quality radial and axial profiles using Raman scattering and CARS. The excellent agreement between the radial temperature profiles using the CARS and Raman methods provides confidence in the fidelity of the measurements and in the symmetry of the flame.

In the radial profiles at z = 3 mm, significant flame spread, determined from the position of the maximum temperature is the result of the strong radial diffusion of the fuel molecules. At low exit velocities, this diffusion even reduces the centerline fuel concentration substantially below that of the original fuel jet. In all of the experiments, the maximum measured temperature never reaches the maximum value at adiabatic equilibrium. Comparison of the experimental results with those derived from adiabatic equilibrium, using the measurement data to derive the local composition, show the
necessity of including the effects of differential molecular diffusion in the calculation of the equilibrium state. Whereas inclusion of these effects improves the agreement, by lowering the adiabatic temperature, it is insufficient to reconcile the discrepancy at equivalence ratios richer than ~ 0.9. As most clearly evinced in the axial profiles, at leaner equivalence ratios the inclusion of differential diffusion results in excellent agreement with the measurements. The agreement in the axial profiles downstream of the maximum temperature indicates that radiative heat losses are negligible in the flames studied.

The elimination of differential diffusion and radiative heat loss as causes of the large discrepancy between equilibrium and measured temperatures means that the origin of this discrepancy must be sought elsewhere. Instead, significant non-equilibrium radical concentrations, and possible radial heat conduction, may contribute to the reduced peak temperatures observed in these flames. We shall return to these possibilities in Chapter 6.

References