Summary

This thesis presents an experimental study of the structure of laminar axisymmetric coflow hydrogen diffusion flames. The motivation behind studying these flames is the current drive towards sustainable energy and strict pollution norms. In this regard, hydrogen as a fuel is one such candidate, which burns clean and possesses the capability as a future sustainable energy carrier. In addition, hydrogen flames are known to have many interesting properties arising mainly from finite-rate chemistry, thermal diffusion, and large differences in diffusion coefficients between H\text sub{2} and other molecules. Considerable research will be needed to understand H\text sub{2} diffusion flame structure in practical devices, which often possess multidimensional turbulent flow and complex chemical kinetics. Studying flame structure using the model flames discussed in this thesis can provide insight into the behavior of practical systems. The study of steady laminar flames also allows direct comparison of measurements with theoretical predictions.

Understanding flame structure requires the knowledge of the spatial distributions of many parameters, such as species concentrations, temperatures and velocities, which define it. The progress made in the fields of nonintrusive laser diagnostics and numerical simulation helps to provide details of the structure of diffusion flames. In this work (as described in Ch. 2), major species were measured using spontaneous Raman scattering, temperatures were measured using coherent anti-Stokes Raman scattering (CARS) and also derived from the Raman measurements, and hydroxyl (OH) molecules in the flames were measured using laser-induced fluorescence (LIF). The Raman scattering data in the flames were collected along the laser beam and provided a spatial resolution of ~ 0.2 mm, while the CARS setup had typical spatial resolution of ~ 1 mm. The LIF technique was used in the linear regime and the spatial resolution obtained was ~ 0.5 mm.

Chapter 3 of this thesis describes the procedure for obtaining quantitative information from the spontaneous Raman scattering and the LIF measurements. Fitting the measured Raman spectra greatly simplified the experimental procedure; only a single daily calibration measurement of the Raman signal was required. The accuracy of the Raman setup was better than 10% (relative) for major species with mole fraction \( \geq 0.1 \), and 0.01 mole fraction (absolute) for lower concentrations. The temperatures obtained by the Raman measurements in premixed methane/air and hydrogen/air flames were in excellent agreement with those obtained by CARS, with a maximum of 50 K difference between them. The excellent agreement achieved
between the results calculated using equilibrium assumptions and fitting the experimental spectra with different signal-to-noise ratio indicated that the fitting code can even be used to fit moderate quality spectra. The LIF data in the diffusion flames were quantified by direct absorption measurements in a calibration flame, and corrected for the temperature and collisional-dependent factors based on the CARS and Raman measurements. The scatter in the Raman species data used for calculating the correction factors contributed substantially to the scatter in the LIF results.

**Chapter 4** focuses on the experimental study of laminar axisymmetric coflow H$_2$/air flames by discussing the profiles of major species concentrations and temperature obtained by spontaneous Raman scattering and CARS, respectively. The excellent agreement between the temperatures derived from fitting the Raman spectra and those obtained from the CARS measurements provided additional confidence in the experimental procedure for reducing the Raman data, and in the symmetry of the flame. In the radial profiles, significant flame spread due to the strong radial diffusion of the fuel was observed. In all of the experiments, the maximum measured temperature never reached the maximum value at adiabatic equilibrium. Comparison of the experimental results with those derived from equilibrium calculations, using the measurement data to derive the local composition, shows 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 resolve the discrepancy at local equivalence ratios richer than ~ 0.9. As clearly shown in the axial profiles, at leaner equivalence ratios the inclusion of differential diffusion results in excellent agreement with the measurements. This agreement downstream of the position of 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 indicates that the origin of this discrepancy must be sought elsewhere. Instead, significant non-equilibrium radical concentrations and radial heat conduction may contribute to the reduced peak temperatures observed in these flames.

**Chapter 5** presents a combined experimental and numerical study of the major species and temperature distributions of nitrogen-diluted H$_2$ diffusion flames. The addition of N$_2$ in the fuel minimized the gradients of species and temperature at the exit of the fuel tube and allowed matching the inlet boundary conditions in the numerical simulations. Normally, thermal diffusion of light H$_2$ molecules is significant in regions with strong temperature gradients. Inclusion or neglect of
thermal diffusion in these flame structure calculations is seen to have the largest effects on the computed results, and the experimental data have been compared with both models. In the axial and radial profiles 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. In particular, the fuel-rich inner side of the jet is predicted quantitatively; however, the mixing on the fuel-lean side of the jet is underpredicted in both computations. The discrepancy between the measured and the computed results on the fuel-lean side is more pronounced in the model including thermal diffusion. The relative discrepancies observed are, however, never more than 25%. Variation of transport properties for H and H$_2$, and of the flow velocities, yields modest changes in the base solution, suggesting other causes for the discrepancies seen. Significantly, both models are able to reproduce the complex non-monotonic profile of N$_2$, resulting from transport processes and chemical reaction. The predicted axial temperatures downstream of the maximum are significantly higher than the measurements. These differences are ascribed to residual differences in the boundary conditions between the experiments and simulations.

Chapter 6 is devoted to the analyses of hydroxyl (OH) radical formation and consumption in the pure and nitrogen-diluted hydrogen flames. Good agreement is found between the measured OH concentrations and those obtained by numerical solution of the governing equations incorporating full-chemistry and detailed transport in the H$_2$/N$_2$ flames. Similar to the results obtained for the temperature and major species mole fractions in Chapter 5, the OH mole fractions are predicted better in calculations without thermal diffusion than in calculations incorporating thermal diffusion. Analysis of the results from the H$_2$/N$_2$ flames shows that the full equilibrium approach underpredicts the measured OH mole fractions, while the partial-equilibrium assumption overpredicts the mole fractions at all locations. In the flames of pure hydrogen, a substantial impact of differential transport on the properties of these flames is exposed by comparing the measured OH mole fractions with those calculated in the equilibrium approximation. The results show that both finite-rate chemistry and differential molecular diffusion are responsible for “depressing” the temperature below the equilibrium value calculated assuming equal molecular diffusivity.