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.
Introduction
1.1 Introduction

Combustion is one of the oldest technical processes, and is an everyday phenomenon. For instance, we use combustion to cook our food, heat our buildings and power motor vehicles. Combustion is the dominant mode of energy conversion. Today, combustion related research is driven mainly by three objectives: 1) obtaining maximum efficiency, with an eye towards the limited supply of fossil fuels 2) minimizing pollution to preserve the environment and 3) achieving safe and stable operation for the end user. The main challenge for combustion researchers, however, is to accomplish these goals simultaneously. For example, methods to reduce nitric oxides (NO\(_x\)) emissions from natural gas-fired burners, such as lean-premixing and flue gas recirculation (FGR), nearly always lead to reduced stability of the flames. In this case, it becomes important to minimize pollutant emissions yet maintain stable combustion. In order to fulfill all these criteria effectively, it is essential to investigate the underlying physical and chemical processes that are responsible for combustion behavior; i.e. studying flames.

Our understanding of flame structure, the spatial distribution of species and temperature in a flame, has vastly grown; thanks to the improvements in experimental and theoretical methods in the last 30 years. However, gaining insight into the structure of flames in practical combustion systems is often complicated by multidimensional (often turbulent) fluid flow and complex chemical kinetics. Predicting the structure of these flames theoretically is difficult, and measuring species concentrations, temperatures and velocities in these flames is tedious at best, if not impossible. Even if investigations are made in such flames, interpretation of the results is often problematical due to the complexity of the system. To obtain some insight into the behavior of the practical flames, it is thus useful to study simpler model flame systems [1], which are usually used for direct comparison with theoretical predictions.

Given the importance of natural gas as a fuel, many detailed flame structure studies have concentrated on studying model flames using methane as a primary fuel. With the current drive towards sustainable energy and strict pollution norms, it is interesting to study the combustion behavior of sustainable fuels. This thesis describes research into the structure of model flames of one such fuel: hydrogen. In this work, we study the laminar nonpremixed flames of hydrogen, stabilized in an annular flow of air. As a general introduction, this chapter presents a brief description of basic
flame types, the importance of and approach to the study of model flames and research activity on flames using hydrogen as a fuel.

1.2 Flames

Flames are often divided with respect to their premixedness and flow type [2]. In premixed flames, the fuel and the oxidizer are mixed prior to combustion. A flame is said to be stoichiometric when both the fuel and the oxidizer are mixed in proportion corresponding to the balanced chemical equation describing the oxidation of the fuel. The proportion with which the fuel and the oxidizer are mixed is commonly described by the equivalence ratio ($\phi$). It is the ratio of fractions (mass or mole) of the fuel to that of the oxidizer, divided by the stoichiometric ratio of the fractions. If the fuel is in excess compared to the stoichiometric proportion, then the flame is said to be fuel-rich ($\phi>1$), while a flame with excess oxidizer is called fuel-lean ($\phi<1$). In nonpremixed flames, also called diffusion flames, the fuel and the oxidizer are mixed during the combustion process itself. Both premixed and nonpremixed flames can further be classified as either laminar or turbulent depending on the regime of gas flow.

Figure (1.1) sketches typical examples of laboratory flames. The one-dimensional flat premixed flame (Fig. 1.1a) is an example of a model flame system, which is useful for understanding the primary flame front in most domestic gas appliances, and is also often studied to understand the microscopic structure of premixed turbulent flames in gas engines and low-NO$_x$ turbines. Laminar nonpremixed flames are used as models to gain insight into the microscopic flame structure of turbulent nonpremixed flames in large boilers, industrial burners, power plants, etc. These flames are usually used in two geometries: coflow and counterflow. In the coflow geometry (Fig.1.1b), the convective fluxes of the fuel and the oxidizer are parallel to each other. In the counterflow geometry (Fig. 1.1c), the fuel and the
oxidizer flow in opposite directions. The introduction of the so-called Tsuji burner in 1971 [3], schematically shown in Fig. 1.2, accelerated systematic research work on diffusion flame structure.

In the Tsuji burner, a laminar flow of fuel leaves a porous cylinder and stagnates against the laminar flow of oxidizer approaching the cylinder from the opposite direction. The mathematical treatment in this case is simplified if the description is restricted to the flow properties along the stagnation stream line, where the problem reduces to only one spatial coordinate.

1.3 Study of laminar jet diffusion flames

While the Tsuji burner geometry simplifies the nonpremixed flame structure analysis to a one-dimensional problem, a laminar jet nonpremixed flame (such as one
shown in Fig. 1.1b) requires at least two-dimensional treatment. The first formal theoretical interpretation of diffusion flames produced in 1928 [4], is very useful for obtaining a qualitative understanding of the global structure of these flames. In the “Burke-Schumann” model, it is assumed that the reaction occurs in an infinitesimally thin zone around the issuing fuel jet, where air and fuel are in stoichiometric proportions. That is, diffusion alone is the rate-determining process of combustion. Using Burke-Schumann theory, for the burner arrangement with inner tube of radius \( r \) and the outer annular tube of radius \( R \) (Fig. 1.3), the height \( h \) of a jet nonpremixed flame can easily be estimated. In this case, the time \( t \) needed for the fuel to reach the tip of the flame is

\[
t = \frac{h}{\dot{\vartheta}},
\]

where \( \dot{\vartheta} \) is the axial velocity of the jet. The time \( t \) is also equal to that needed for the oxidizer to diffuse to the centerline, which can be estimated by Einstein’s equation for the depth of intrusion by diffusion as

\[
r^2 = 2Dr,
\]
where $D$ is the mean diffusion coefficient. From Eq. 1.1 and 1.2, the expression for the flame height can be obtained as

$$h = r^2 \frac{\dot{\phi}}{2D}.$$  (1.3)

Replacing the velocity $\dot{\phi}$ by the volume flow rate $Q = \pi r^2 \dot{\phi}$, for the cylindrical geometry, Eq.1.3 can be rewritten as

$$h = \frac{Q}{2\pi D}.$$  (1.4)

From Eq. 1.4, we observe that according to the Burke-Schumann flame model, the flame height of a laminar jet nonpremixed flame is proportional to the volume flow and inversely proportional to the diffusion coefficient. According to the Burke-Schumann model, we can expect that the flame height of a hydrogen diffusion flame is smaller than a methane flame at the same flow rate, due to the higher diffusion coefficient of hydrogen. In general, the Burke-Schumann model predicts flame shapes and heights well [4]. However it obviously does not provide insight into many issues that rely on the details of combustion, such as finite-rate chemistry effects, complex transport-chemical kinetics interactions, pollutant formation, etc. in these flames. Investigations on such phenomena demand investigating the microscopic structure of the reaction zone. With the recent progress in numerical methods (see section 1.3.1 below), many phenomena in reactive flows can be predicted. However, experiments must be performed to assess accuracy of those predictions. Today, most of our understanding about diffusion flames comes from computer simulations and non-intrusive laser diagnostic methods.

### 1.3.1 Use of computational methods

Recent advances in computational methods and high-speed computing have created a powerful tool for flame research. Most of these numerical programs solve the governing equations using detailed chemical kinetics and fluid flow properties, with a minimum of approximations. It is now possible to obtain the results of the computer simulations on the coupled fluid dynamic-chemical interactions in flames in a reasonable amount of time. For example, the current computer programs solve coupled conservation equations in a one-dimensional premixed methane-air flame in a few minutes, while a two-dimensional diffusion flame usually requires several hours, even for simple systems. Three-dimensional flame calculations with full chemistry
Introduction

and transport are currently beyond the capacity of today’s computers. In spite of this progress, there are still major uncertainties in the accuracy of the computational results. The most obvious of these is uncertainties in the flame chemistry [1]. An example of the power of the computational methods will be presented later in this thesis.

1.3.2 Use of laser diagnostics in flame studies

Traditionally, flame properties were measured by intrusive means, such as suction probes, thermocouples, etc. The main drawback to these techniques for flame studies is that they usually perturb the flame structure, altering the system under investigation. Further, in flames having steep gradients in temperature and concentration, intrusive techniques usually provide only modest spatial resolution. Use of thermocouples usually requires corrections for conduction and radiation losses, as well as potential catalytic effects on the surface. Accurate results from probe measurements demand proper quenching of the probed sample, which can be difficult for some species, such as CO [5]. Compared to the drawbacks of physical probing, laser diagnostics offers many advantages in flame structure measurements [6]. Besides being non-intrusive in nature, laser-based methods offer potentially excellent spatial and temporal resolution. Laser techniques, such as spontaneous Raman scattering, laser induced fluorescence (LIF), laser Doppler anemometry (LDA), laser induced incandescence (LII), etc. are commonly used to obtain information on flame structure. In this thesis, spontaneous Raman scattering (major species mole fractions and temperature), coherent anti-Stokes Raman scattering (CARS) (temperature) and LIF (OH radicals) have been used. The details of these measurements are provided in subsequent chapters.

1.4 Laminar coflow diffusion flames of H$_2$

The current understanding of the detailed structure of coflow laminar diffusion flames has been obtained mainly from the research work using methane as a fuel [7-16]. Methane flames have been given substantial attention because of the use of natural gas (in which, methane is a predominant component) in industrial and residential appliances. In particular, issues such as detailed chemistry-flow field interactions [8-10], NO$_x$ formation [11-13], soot and polycyclic aromatic hydrocarbons (PAH) formation [14], chemical non-equilibrium effects [15-16], etc. have been discussed.
While considerable progress has been made in understanding the 2-D structure of methane flames, much less attention has been paid towards hydrogen flames. These flames incorporate relatively simple chemistry with pronounced differences in transport properties between fuel and oxidizer, and are also becoming more interesting due to the role that hydrogen can play as a future sustainable energy carrier, as mentioned above. In this regard, the utilization of hydrogen in the wide range of hydrocarbon-fired combustion systems in use today will require understanding the processes governing flame stability and pollutant formation under practical conditions.

Laminar axisymmetric hydrogen-air flames have been used as a test object for developing various diagnostic techniques for flame studies, where diagnostic aspects such as accuracy, selectivity and spatial resolution have been the focus. Appendix 1 [17-51] gives an overview of various experimental studies conducted on laminar H\(_2\) diffusion flames. In these studies, however, no conclusions have been drawn concerning flame structure. Research studies, incorporating numerical simulations of H\(_2\) diffusion flame structure and their validation using appropriate experimental data, are very limited [30, 34, 41]. A brief summary of some interesting properties of H\(_2\)-air diffusion flames studied to date:

- **Finite-rate chemistry effects:** Frequently, temperature measurements in H\(_2\) diffusion flames show experimental temperatures below the stoichiometric adiabatic equilibrium temperatures [20, 24, 52]. One of the reasons for this behavior is the finite-rate chemistry effects. These effects often produce superequilibrium radical concentrations, which only slowly recombine towards equilibrium; excess radicals result in subequilibrium temperatures. The impact of finite-rate chemistry on various flame properties is difficult to predict, *a priori*. Thus, it is important to examine the finite-rate chemistry effects using detailed measurements and accurate calculations.

- **Large differences in diffusion coefficients:** In laminar H\(_2\) diffusion flames, hydrogen molecules diffuse much faster than the heavier species, which can cause several interesting phenomena. For example, the large differences in diffusion coefficients are attributed to “flame-tip opening” of hydrogen diffusion flames when diluted with heavier inert compounds [23]. These effects were also cited as reason for the observed differences in temperatures in convex and concave regions in flame-vortex interactions studies in nonpremixed H\(_2\) jet flames [30].

- **Thermal diffusion effects:** One interesting fact about flames using hydrogen as a fuel component is that it can show thermal diffusion effects. Thermal diffusion,
transport of mass towards regions of high temperature, is more pronounced for light molecules such as H$_2$. Normally inconsequential in most combustion calculations of hydrocarbon fuels, thermal diffusion becomes significant in hydrogen diffusion flames due to the high temperature gradients and disparate molecular weights of the fuel components. Discrepancies between the measured and the numerical temperature profiles observed in [33] were attributed to thermal diffusion in the flames.

The literature cited in this section gives information on the various phenomena appear in laminar coflow H$_2$ diffusion flames. In spite of the studies performed to date, there has yet to be a study of the detailed structure of these flames, which prompted the present investigation.

1.5 Aim and scope of the present investigation

Against the background on the importance of studying laminar 2-D diffusion flame structure, and the interesting aspects of hydrogen as a fuel, a broad objective of this thesis is to gain insight into the structure of laminar diffusion flames of hydrogen. Towards this end, diffusion flames of pure H$_2$ and diluted H$_2$/inert gas mixtures were studied in the axisymmetric coflow geometry. Species concentrations and gas temperatures were measured using non-intrusive laser diagnostic methods. Where possible, the information obtained from the experiments was analyzed using the results of numerical calculations of flame structure in the same geometry. The outline of this thesis is as follows:

- **Chapter 2** introduces the laser diagnostic techniques used in this work. The methods discussed in this chapter are 1) spontaneous Raman scattering 2) coherent anti-Stokes Raman scattering (CARS) 3) laser induced fluorescence (LIF) and 4) direct absorption.
- **Chapter 3** presents the methodology to derive the quantitative information from the spontaneous Raman scattering and the LIF measurements. In particular, the issues, such as the spectral fitting of the experimental Raman spectra and deriving OH mole fractions from the measured LIF signals are discussed.
- **Chapter 4** presents the experimental results of the major species and temperature in axisymmetric laminar diffusion flames of pure H$_2$ in coflowing air.
- **Chapter 5** presents a combined experimental and theoretical study of the flame structure of axisymmetric coflow laminar diffusion flames of N$_2$ diluted H$_2$ in air.
- **Chapter 6** discusses the OH measurements in the coflow H$_2$ diffusion flames.
Chapter 1

References


