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Quantitative atomic hydrogen measurements in premixed hydrogen tubular flames

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ABSTRACT

Quantitative measurements of atomic hydrogen are reported in laminar premixed tubular flames using femtosecond two-photon laser-induced fluorescence. The H-atom fluorescence is corrected for collisional quenching by using local values of major-species concentrations and temperature measured by spontaneous Raman scattering. Lean hydrogen flames are sustained with two different diluents (N_2, CO_2) to investigate low–Lewis number flames under high curvature. When compared to planar stretched flames, the curved tubular flames enhance the H-atom concentration and temperature through increased preferential diffusion. Peak H-atom number densities on the order of 10^{15} per cm^3 are measured, and absolute H-atom profiles show differences up to 40% in peak number density and flame radial position when compared to the pseudo one-dimensional flame model with detailed chemistry and transport. Although the overall agreement in absolute H-atom profiles is “good” considering the relative uncertainties in the model and experiment, the differences suggest the ability for this flame geometry to provide evidence for revision of molecular transport and chemical kinetic modeling in flames with substantial preferential diffusion. Peak absolute H-atom concentrations vary up to 30% depending on the assumed temperature dependency for the collisional quenching factors pointing to the need for high-temperature data for H-atom collisional quenching. Relative H-atom profiles in the flames are minimally affected by the collisional quenching.

1. Introduction

The accuracy of combustion simulation is dominated by chain-carrying reactions in chemical kinetic models (e.g., see [1]). Chemical radicals involved in these reactions strongly define flame structure, and spatially resolved measurements help refine kinetic models. Arguably one of the most important radicals, atomic hydrogen, is the lightest chemical species and therefore the most susceptible to commonly neglected phenomena (e.g., secondary transport processes or non-unity-Lewis number effects). Spatially resolved measurements of this species under specific flame conditions (e.g., artificially lowered Lewis number mixtures) offer the best opportunity to investigate common simplifying assumptions in combustion modeling.

Quantitative measurement of chemical radicals is a difficult task [2]. The most significant hurdle for atomic-radical detection lies with discrimination of the signal from nascent species from photolytic interferences due to the high photon energies required for laser diagnostic techniques [3]. Recent work in a rich $CH_4/O_2/N_2$ Bunsen flame [4] has successfully shown the advantage of using femtosecond laser systems to examine two-photon laser-induced fluorescence (TPLIF) of atomic hydrogen—effectively reducing photolytic interferences to negligible levels. Because of the long timescale of photolytic dissociation compared to the sub-picosecond excitation pulse length, any photolytically produced H atoms are not excited and do not produce fluorescence signal. Additionally, the short high-intensity laser pulse selectively enhances desirable two-photon processes over the parasitic one-photon interferences. In the rich Bunsen flame [4], the H-atom fluorescence signals were corrected for collisional quenching using the collider-species concentrations from a numerical simulation. The relative shapes of the measured and simulated H-atom profiles showed excellent agreement.

A next step in this measurement technology is to make absolute H-atom concentration measurements by calibrating the H-atom TPLIF system in a reference flame and correcting the H-atom fluorescence for collisional quenching using measured values of

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the major-species concentrations and temperature at the same flame location. Additionally, we wish to characterize flame geometries that more closely represent practical combustion. The flame prototype studied here is the tubular flame [5,6], where the premixed flame experiences uniform stretch and curvature. Previous work investigating this geometry has focused on major-species and temperature measurements by spontaneous Raman scattering [7,8], finding strong enhancements to the local mixture due to the thermo-diffusive nature of dilute lean hydrogen mixtures. Comparisons of the major-species and temperature measurements to a reduced numerical model have shown overall good agreement, but the ability of the model to predict important minor-species profiles needs to be assessed. Because of its light mass and active chemistry, H-atom is a very important minor species in tubular flames. In this paper, we make absolute H-atom measurements with TPLIF to characterize the thermo-diffusive effects in tubular flames.

2. Tubular flame geometry

Fundamental flame studies using laminar burners are commonly performed with opposed jets, which impose an approximately constant scalar flame stretch upstream of the flame surface [9]. To study the effects of curvature on stretched flame structure, the tubular burner was developed as an extension of this configuration [5,6]. The flow field remains azimuthally symmetric; however, it is characterized by a negative radial velocity gradient and positive axial divergence. For a flame sustained near the center of the domain, the local flow field is characterized by approximately constant curvature and stretch [10]. This burner geometry then augments the study of uniformly stretched, flat flames with uniformly stretched, uniformly curved flames.

Similar to opposed jets [9], the boundary-layer approximation to the Navier–Stokes equations may be used to reduce the dimensionality of the axisymmetric tubular flame to a function of only the radial coordinate [11,12]. This model has been implemented using a modified form [7] of OPPDIF [13] and has been compared to experimental measurements of major species and temperature in N_2-diluted H_2 flames with good success [7,8]. Simulations were performed using comprehensive H_2/O_2 chemistry [14] assuming a non-participatory diluent (N_2 or CO_2). Full multi-component transport with thermal diffusion is included. It is noted that the kinetic mechanism is configured for N_2 as the main diluent (see [14] for more detail).

The tubular burner was designed [15] specifically to investigate high-stretch and high-curvature cases in the laminar flow regime. The burner is a Radial Flow Nozzle design [16] with three optical windows to permit laser-diagnostic techniques, and is shown in Fig. 1. For a non-reacting flow field, the stretch rate converges to a constant value near the burner center [10],

$$k = \pi\frac{V}{R}\cos\left[\frac{\pi}{2}\left(\frac{r}{R}\right)^2\right] \approx \pi\frac{V}{R}$$

where V is the inlet radial velocity, R is the burner radius, and r is the radial coordinate. Flames at short radii minimally perturb the flow field [10], and the stretch rate ahead of the flame is well approximated by the asymptotic value. Stretch rates reported here will refer to the asymptotic value, as an expression of the boundary conditions.

Four flame cases were investigated that formed an abbreviated parametric study of flame stretch (either 200 or 400 s^{-1}) and Lewis number (0.31 or 0.23). The gas mixtures were each composed of three components: H_2, O_2, and either N_2 or CO_2. The two diluents were chosen to investigate different sub-unity Lewis numbers; the relevant experimental conditions are shown in Table 1. To assess the degree of non-equilibrium, the extinction stretch rates were numerically estimated and are in excess of 1000 s^{-1} for both mixtures. The form of the numerical solution algorithm (without arc-length continuation) is known to not converge near an extinction limit (e.g., [9,12]) and the numerical value provides an underestimate of the extinction stretch rate. Experimentally determining this limit is not possible with the current burner design as the high boundary velocities will cause turbulent flow. Gas flow rates were controlled with Teledyne Hastings mass-flow controllers (HFC-202/203), which are accurate to 1% full scale.

3. Measurement method

Atomic hydrogen is detected using TPLIF through $1 \rightarrow\rightarrow 3$ excitation (205.1 nm) followed by detection of the $2 \leftarrow 3$ fluorescence (656.5 nm). Two-photon absorption scales quadratically with laser fluence, hence using a short-pulse high-intensity scheme significantly reduces the relative probability of single-photon processes (e.g., major photolytic effects [3]) that scale linearly with laser fluence. It was found that using sub-picosecond pulse lengths decreased the relative effects of parasitic single-photon processes to negligible levels [4] for a rich premixed Bunsen flame. The laser system used here was a Coherent Legend Elite Duo (100-fs pulses at 10 kHz), which was directly quadrupled using a fourth-harmonic generation unit built in-house [17]. The spectral profile was tuned to maximize fluorescence signal, resulting in a bandwidth of approximately 1 nm. The laser beam was aligned along the diameter of the tubular flame, providing a measurement volume perpendicular to the H-atom concentration gradients. Pulse spectral content and energy delivered to the measurement volume (4.30 ± 0.16 μJ) varied little over the course of the measurements and were considered constant during the data-reduction procedure. The beam diameter in the measurement volume was estimated at 400 μm, leading to a laser fluence of 3.4 mJ/cm^2; this laser fluence is below the laser-fluence threshold for photolytic production of H atoms or stimulated-emission interference for H-atom LIF measurement with femtosecond lasers [4] and picosecond lasers [18].

Fluorescence was collected with a Nikon f/1.2 lens coupled to an intensified camera system, resulting in projected pixel size of 67 μm. A bandpass filter (Semrock FF01-655/40-50) combined with a 300-ns intensifier gate was used to reduce background flame emission. The detection system was configured to maximize the signal-to-noise ratio (nominally 50–60), and consisted of a LaVision High-Speed Intensified Relay Optic (IRO) system and Andor Newton EMCCD detector. The IRO was optimized for UV wavelengths but had sufficient sensitivity at 656 nm (~3% QE). The EMCCD was operated with no electron-multiplying amplification to function as a conventional CCD detector. The instrument spatial response was estimated as a convolution [19] and was approximated from an acquired image of a knife edge. It was found to be well modeled as a Gaussian profile (full width at half maximum of 400 μm) within experimental error.

To facilitate comparison of the experimental and simulated number-density profiles, the instrument response was removed from the experimental data. Deconvolution was performed using two algorithms simultaneously to increase confidence in the processed results. The algorithms used were *Wiener* and *Lucy-Richardson* [20], implemented natively within the software package MathWorks MATLAB R2012a. The differences between these two algorithms are expected to be small [20]; however, because of the presence of sharp peaks, the Wiener algorithm suffers from artificial oscillations (characteristic of Fourier Transform signal-processing methods). Both algorithms were found to be relatively insensitive to the deconvolution parameters, as long as the