Experimental and numerical study of H₂-air non-premixed cellular tubular flames

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Abstract

Experimental and numerical simulation results are presented for the first time of H₂ fueled non-premixed cellular tubular flames. Non-intrusive measurements of temperature and major species were obtained with Raman scattering. Numerical simulations were performed with a 2D fully-implicit primitive variable finite difference code that includes multicomponent transport and detailed chemical kinetics. Good agreement is found for all flame cases. The experimental cell temperatures and the shapes of the flame cells are well predicted. Less significant discrepancies are observed in the character of the extinction zones and are discussed. Overall, non-premixed cellular tubular flames are found to be valuable experimental and numerical targets for fundamental combustion research.

Keywords: Laminar flames, Raman scattering, diffusion flame, cellular flames

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1. Introduction

Studies of non-premixed laminar flames have long been a part of combustion research and continue to be relevant today [1]. More recently, the cellular instability has been sustained in laboratory burners and allows the investigation of a flame geometry that exhibits local extinction zones and diffusive focusing of reactants [2]. This flame type has been investigated in the axial jet geometry both experimentally with chemiluminescence imaging [3, 4], and numerically with a detailed simulation approach [5].

The next step would be to perform detailed chemical species measurements for comparison to the simulation results. However, the axial jet flow field is 3D and time dependent, greatly complicating numerical and experimental characterizations. A lower dimensionality flow field alleviates this difficulty: cellular flames in the *opposed tubular burner* [6] exhibit structure that is time independent, spatially 2D, and planar [6, 7, 8].

Reported here are 2D spatially resolved quantitative measurements and detailed simulation results for the major species and temperatures found in select non-premixed cellular tubular flame cases. The measurements were performed with Raman scattering, which is a non-intrusive laser diagnostic technique. The simulations were performed with a 2D fully-implicit primitive variable finite difference code [9] that includes multicomponent transport (with Soret/Dufour effects) and detailed chemical kinetics.

2. Tubular Flowfield

Stretched non-premixed flames are commonly examined in the axially-opposed jet orientation, where the stagnating jets create a uniform and flat flame structure [10]. The tubular flow field, which can also be described as the *radially-opposed jet* orientation, allows for a uniform and curved non-cellular flame structure [11]. The opposed tubular burner used here [12] was developed specifically to examine the effects of curvature on stretched non-premixed flame structure. A schematic of the burner is shown in Figure 1. The oxidizer (air) enters from the contoured outer nozzle (8 mm height, 12 mm radius) and the fuel mixture (diluted H_2) enters from the porous metal inner nozzle (8 mm height, 3.175 mm radius). Both nozzles are surrounded by nitrogen co-flows to minimize the effect of the room conditions on the flame and to provide cooling on the inner nozzle.

The governing equations for the inviscid non-reacting flow field can be solved analytically [13] for the stretch rate k on the fuel side of the stagnation zone,

$$k = \pi \frac{|u_i|}{R_i} \left(\frac{\left| \frac{u_o}{u_i} \right| \sqrt{\frac{\rho_o}{\rho_i}} + \frac{R_i}{R_o}}{\frac{R_o}{R_i} - \frac{R_i}{R_o}} \right) \quad (1)$$

Figure 1: Simplified schematic of the cylindrically symmetric tubular burner. Three optical access ports are mounted at mutual right angles to allow laser diagnostic measurements.

where u is the radial velocity, R is the boundary radius, ρ is the mixture mass density, and the subscripts i , o refer to the inner nozzle (fuel) and outer nozzle (oxidizer) streams, respectively. The absolute value signs are used to emphasize the experimental operating conditions: positive u_i , negative u_o , and positive stretch k .

3. Experimental Method

The flame chemiluminescence was imaged with an ICCD camera coupled to a multi-element UV lens (Nikon UV-Nikkor, f/4.5, 105 mm). A colored-glass filter (UG11) was used to discriminate the flame chemiluminescence from ambient and laser light. The camera was focused at the center of the burner with a mirror oriented at 45° , to provide an axial image with the camera safely mounted out of the flame exhaust stream. The mirror has a slot cutout to accommodate the inner nozzle, similar to previous investigations of non-premixed tubular flames [8, 11].

Major species and temperature measurements were performed over 2D flame cross-sections using Raman scattering. This technique is a non-intrusive laser based method for 1D measurements, and was scanned across flame cross-sections to yield fully 2D spatially resolved structure. The experimental setup is similar to that used successfully in prior investigations of non-cellular non-premixed flames [11], and for a detailed description of the setup used here see reference [14].

An uncertainty analysis was performed to estimate experimental error in the Raman measurements. Characteristic values of temperature uncertainty are approximately ± 70 K in the hot products and ± 3 K in the room temperature reactants. Characteristic chemical species uncertainties are approximately ± 2 % (mole fraction) in the hot products and ± 0.5 % in the room temperature reactants.

3.1. Background Subtraction

Strong interferences are observed when measurements are performed near the inner nozzle. This was also found in previous investigations of non-cellular tubular flames [11]. The scattering appears as broadband interference that is spectrally diffuse, and is well subtracted in post-processing. The method implemented here is similar to that reported in literature [15], where the background is fit to a low-order polynomial with an iteratively-weighted least squares algorithm. The order of the polynomial (2-4) was chosen in situ during data reduction to satisfactorily handle the background. The same background subtraction parameters used for the cellular data sets were used on non-cellular validation data cases (not shown here for brevity) to ensure confidence in the accuracy of the reported results.

4. Numerical Method

The flame geometry was simulated using a fully-implicit 2D detailed low-Mach number code [9] based on PETSc [16]. The governing equations are written in 2D (r, θ) utilizing the boundary-layer assumption for stagnating jets [17] that removes the axial dependence for tubular flames [18]. This expresses the velocity field in terms of a linear axial velocity gradient W ,

$$\mathbf{u} = u(r, \theta) \hat{\mathbf{r}} + v(r, \theta) \hat{\boldsymbol{\theta}} + zW(r, \theta) \hat{\mathbf{z}} \quad (2)$$

and when combined with the low-Mach number assumption, the dynamic pressure field $\tilde{p}_1$ [19] is expressed as,

$$\tilde{p}_1 = \frac{z^2}{2} H + I(r, \theta) \quad (3)$$

where H is the scalar pressure eigenvalue and I is the radial and tangential pressure dependence. This type of fundamental assumption is also used to express axially-opposed jet flames as 1D [10].

The problem is formulated in differential form in terms of primitive variables, and the governing equations are shown in reference [9]. Radiation heat loss is included in the optically-thin form [20], and detailed H_2/O_2

chemical kinetics are included [21]. A carbon-containing sub-mechanism ($\text{CO}_2, \text{CO}, \text{HCO}$) [22] is added when simulating CO_2 dilution flame cases. EGLIB [23] is used to calculate the multicomponent forms of diffusive transport, including both the Soret and Dufour effects (thermal diffusion).

4.1. Boundary Conditions

The computational domain consists of a $\frac{2\pi}{N}$ angular section of the 2D polar domain to enforce N-fold cellular symmetry. The azimuthal boundary conditions are symmetry conditions except for the azimuthal velocity (v) at the bottom of the domain ($\theta = 0$), where it is set to zero to keep the flame from rotating in the domain during the solution procedure.

The radial boundary conditions for the outer nozzle ($R_o = 12.0$ mm) are,

$$\rho u = (\rho u)_{BC}, v = 0, W = W_{BC}, \quad (4)$$

$$T = T_{BC}, Y_j(u + u_j^d) = (Y_j u)_{BC} \quad (5)$$

where Y_j is the mass fraction of species j , u_j^d is the radial diffusion velocity of species j , and the subscript BC refers to the boundary condition value. The radial boundary conditions for the inner nozzle ($R_i = 3.175$ mm) are the same as for the outer nozzle, except that adiabatic heat transfer to the reactants is included at the porous metal inner nozzle [7],

$$\lambda \frac{dT}{dr} = \sum_j \left(h_j|_T - h_j|_{T_{BC}} \right) (\rho u Y_j)_{BC} \quad (6)$$

where λ is the heat conductivity of the gas mixture and h_j is the specific enthalpy of species j evaluated at either the heated inlet temperature (T) or the inlet gas supply temperature (T_{BC}). Additionally, the dynamic pressure field is set to zero at a single point ($I|_{r=R_i, \theta=0} = 0$), which is arbitrary as only spatial gradients appear in the momentum conservation equations.

The axial velocity gradient boundary conditions were not measured experimentally; the value used for the porous inner nozzle condition (W_i) is zero, and for the contoured outer nozzle condition (W_o) is a positive value determined with numerical experiments. It was found that increasing W_o shifts the stagnation radius and flame to a longer radial position, but the flame structure does not change significantly. The values for W_o are determined for each simulation case to match the flame positions observed experimentally through

Figure 2: Chemiluminescence images showing the cellular non-premixed cases listed in Table 1. Note that the chemiluminescence imaging method (slot-cut mirror) obscures the left side of the flame.

the temperature profiles.

5. Results

Tubular flame cases were examined with a short parametric study to broadly characterize the cellular structure. Chemiluminescence images of the cellular cases are shown in Figure 2, and values for the boundary
 95 conditions are included in Table 1. The overall symmetry of the four flame cases is good at low velocities (cases **a** and **c**), but noticeably degrades at higher inner nozzle flow velocities (u_i). The slight onset of this effect is observed in cases **b** and **d** as unequal flame cell lengths, and is attributed to varying local velocities (jetting) over the surface of the porous inner nozzle. The measurement location for all flames is on the right portion of the images shown in Figure 2.

100 The case study covers two parameters: stretch rate and dilution gas type. Two values of the stretch rate were chosen (low and high) for two fuel diluents (N_2 or CO_2). The low stretch value was chosen as 100 s^{-1} , and the high stretch value was intended to be 200 s^{-1} for both cases. This was not possible for the CO_2 dilution case, where for stretch rates higher than $\sim 150\text{ s}^{-1}$ the flame was strongly non-symmetric. The initial mixture strength Φ (also referred to as the non-premixed equivalence ratio) was kept consistent
 105 between cases with the same dilution, and for all cases the velocities were chosen to obtain approximately equal cold flow stagnation radii (5.0 mm).

The size of the computational domain is chosen to enforce an observed number of cells, and only a single flame cell is simulated. For most cases in Figure 2 (except case **b**) the number of cells is clear (either 4 or

6 cells). The size of the computation domain for case **b** is chosen as 6 cells, as the size of the cells on the
110 right of the image (i.e. where the measurement takes place) are better approximated with 6-cell symmetry
as opposed to 7 cell symmetry.

Shown in Figure 3 are the simulation results for the 6-cell high stretch N₂ diluted flame case **b** in Table
1. The simulation results are shown mirrored over $\theta \in [0, \frac{\pi}{2}]$ for clarity. Local extinction is clearly seen in
the temperature and product (H₂O) plots, as well as many of the minor species plots (O, OH, H).

115 The character of the combustion varies between the radial and azimuthal edges of the flame cell. The
azimuthal edges (facing the extinction zone) show substantially more low temperature radicals (HO₂, H₂O₂)
as compared to the radial edges. All of the higher temperature radicals (H, O, OH), as well as the predicted
chemiluminescence, show an approximately uniform cell structure. However, examination of the heat release
shows higher release at the edges of the cell, similar to the simulation results reported for the cellular axial
120 jet [5]. This is due to the diffusional focusing at the sides of the reaction cell [24] (due to the low Lewis
number mixture), enriching the local mixture and increasing the flux of H₂ into the azimuthal sides of the
flame cell.

Further analysis of the numerical results, combined with numerical experiments, permits discovery of
the broader impacts available through cellular flame research (e.g. with premixed tubular flames [9]). Non-
125 premixed diffusion flames are expected to be significantly more sensitive to diffusive transport modeling
approximations, but what is first required is to compare the experimental characterization to the numerical
predictions. Following that, the sensitivity of the non-premixed cellular flame to specific phenomena may
be characterized.

5.1. Comparisons

130 The experimental and numerical results are shown in Figure 4 for the high stretch CO₂ case **d** in Table 1.
The numerical simulations were performed over a single cell only, and are rotated and mirrored to reproduce
the experimental conditions. Shown in black is the contour of the stoichiometric mixture fraction (0.690)

Table 1: Boundary conditions for non-premixed cellular tubular flames, along with fuel diluent gas. Dimensions are: $\frac{cm}{s}$ for
radial velocity u and $\frac{1}{s}$ for stretch k and the axial velocity gradient W .

	u_i	u_o	W_o	Φ	k	dil.
a	15.4	29.1	40	0.32	101	N ₂
b	30.5	58.3	10	0.34	201	N ₂
c	15.2	35.3	20	0.45	100	CO ₂
d	22.5	53.1	40	0.45	151	CO ₂

Figure 3: Select numerical results for the 6-cell high stretch N_2 diluted flame case **b** in Table 1. The simulation is performed over a single flame cell ($\theta \in [0, \frac{\pi}{3}]$), and is shown mirrored over $\theta \in [0, \frac{\pi}{2}]$ for clarity.

and shown in white is the radial stagnation surface, which is only shown on the simulated flame structure due to the lack of experimental velocity measurements.

Qualitatively, excellent agreement is found between the experiment and simulation. The location of the peak cell temperature is on the lean side (longer radius) of the stoichiometric mixture fraction, and is observed in the simulation to also be slightly on the fuel side of the stagnation surface. Quantitatively the agreement is good for the peak temperature found in the reaction cell, but the character of the extinction zones and the widths of the flame cells are predicted less well. This trend is observed in all 4 of the cases.

The widths of the reaction cells is expected to be very sensitive to the accuracy of the inner nozzle boundary condition. As seen in Figure 2 the individual cells for each case vary somewhat in length, suggesting that slight nonuniformities in the inner nozzle construction may be responsible for this behavior. This hypothesis agrees with previous findings where the flame was perturbed with a probe—individual cells can be extinguished or reignite without affecting others [7], suggesting that there are preferred locations for cells around the inner nozzle.

For a clearer comparison, radial profiles are interpolated along the centers of the reaction cell and the extinction zones and shown in Figures 5, 6, 7, and 8. For all cases it is observed that the temperature and species profiles through the reaction cell are better predicted than for profiles through the extinction zone. The peak temperatures for all cases are approximately 1200 K, and the locations of the gradients in O_2 and

Figure 4: Comparison between numerical and experimental results for the 6-cell high stretch CO_2 diluted flame case **d** in Table 1. The black contour is the stoichiometric mixture fraction (0.690), and the white contour is the radial stagnation surface. The simulation is performed over a single flame cell ($\theta \in [0, \frac{\pi}{3}]$), and is shown mirrored and rotated to reconstruct the experimental conditions.

150 H_2 are well predicted through the reaction cells.

The N_2 diluted cases shown in Figures 5 and 6 are very similar despite doubling the stretch rate from 100 to 200 s^{-1} . The main observable difference for the profiles through the reaction cell is that the cell width narrows with increasing stretch, agreeing with previous observations of chemiluminescence [8]. The character of the extinction zone varies considerably; the temperature is over predicted and O_2 is consumed at longer radii for the low stretch simulation, while the opposite is true for the high stretch case. This inconsistency is not present in the H_2 profile, which is well predicted for both flame cases.

Figure 5: Radially interpolated profiles along the flame cell center (blue) and extinction zone (red) for the 6-cell low stretch N_2 diluted flame case **a** in Table 1. The simulation is shown as solid lines, and the experiment is shown as data points.

Figure 6: Radially interpolated profiles along the flame cell center (blue) and extinction zone (red) for the 6-cell high stretch N_2 diluted flame case **b** in Table 1. The simulation is shown as solid lines, and the experiment is shown as data points.

Figure 7: Radially interpolated profiles along the flame cell center (blue) and extinction zone (red) for the 4-cell low stretch CO_2 diluted flame case **c** in Table 1. The simulation is shown as solid lines, and the experiment is shown as data points.

The CO_2 diluted cases shown in Figures 7 and 8 are also similar, despite increasing the stretch rate from 100 to 150 s^{-1} and changing the number of cells from 4 to 6. The main discrepancy is the under-prediction of temperature and H_2O through the extinction zone. Additionally, the profile of O_2 is found experimentally to be similar between the extinction zone and reaction cell—a trend not predicted with the simulation results.

6. Conclusions

Overall, the 4 cases show strong similarities despite changing the dilution gas type and the stretch rate. The local extinction is found to be thermal-diffusive in nature, as observed in the minor species results from the detailed numerical simulation that were not experimentally measured. The numerical simulation predicts the flame structure well, with the number of cells and the peak temperature values. Discrepancies are observed; however, the sensitivity of the discrepancies to the non-ideal boundary conditions of the inner nozzle is not known.

The non-premixed tubular flame geometry is a powerful investigative tool for investigating local extinction and curvature effects in non-premixed flames. The time steady and 2D structure of tubular flames

Figure 8: Radially interpolated profiles along the flame cell center (blue) and extinction zone (red) for the 6-cell high stretch CO_2 diluted flame case **d** in Table 1. The simulation is shown as solid lines, and the experiment is shown as data points.

simplifies the experimental and numerical requirements for characterization. Further work has been performed with these flame cases, and minor species measurements (H atoms and OH) have been obtained. This data will be analyzed and presented in a future manuscript, allowing for a clearer interpretation of the ability for the detailed numerical simulation to predict the flame structure observed in reality.

With this characterization of the accuracy of the numerical simulation approach, numerical experiments may be performed to aid the development of computationally inexpensive approximations for problem physics (e.g. reduced chemical mechanisms or mixture-averaged transport) that are required for large scale simulations of combustion at engine-relevant conditions.

Acknowledgments

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Captions

Figure 1. Simplified schematic of the cylindrically symmetric tubular burner. Three optical access ports are mounted at mutual right angles to allow laser diagnostic measurements.

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