Laminar flame speeds of H$_2$/CO mixtures: Effect of CO$_2$ dilution, preheat temperature, and pressure

J. Natarajan *, T. Lieuwen, J. Seitzman

School of Aerospace Engineering, Georgia Institute of Technology, Atlanta, GA 30332-0150, USA

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Abstract

Laminar flame speeds of lean H$_2$/CO/CO$_2$ (syngas) fuel mixtures have been measured over a range of fuel compositions (5–95% for H$_2$ and CO and up to 40% for CO$_2$ by volume), reactant preheat temperatures (up to 700 K), and pressures (1–5 atm). Two measurement approaches were employed: one using flame area images of a conical Bunsen flame and the other based on velocity profile measurements in a one-dimensional stagnation flame. The Bunsen flame approach, based on imaging measurements of the reaction zone area, is shown to be quite accurate for a wide range of H$_2$/CO compositions. These data were compared to numerical flame speed predictions based on two established chemical mechanisms: GRI Mech 3.0 and the Davis H$_2$/CO mechanism with detailed transport properties. For room temperature reactants, the Davis mechanism predicts the measured flame speeds for the H$_2$/CO mixtures with and without CO$_2$ dilution more accurately than the GRI mechanism, especially for high H$_2$ content compositions. The stagnation flame measurements for medium levels of H$_2$ at both 1 and 5 atm, however, show lower than predicted strain sensitivities, by almost a factor of two at lean conditions ($\Phi = 0.6–0.8$). At preheat temperatures comparable to those found in gas turbine combustors, the accuracy of the flame speed predictions worsens. For example in fuels with low levels of H$_2$, both models underpredict the measurements. In contrast, for medium H$_2$ content fuels, both measurement techniques show that the models tend to overpredict flame speed, with the discrepancy increasing as $\Phi$ decreases and temperature increases. In general, the Davis mechanism predictions are in good agreement with the measurements for medium and high H$_2$ fuels for preheat temperatures up to 500 K but overpredict the measurements at higher temperatures.

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Keywords: Syngas; Laminar flame speed; CO$_2$ dilution; Reactant preheat; Pressure

1. Introduction

Technologies such as integrated gasification combined cycle plants enable combustion of coal, biomass, and other solid or liquid fuels while still maintaining high conversion efficiencies and low pollution emissions. Synthetic gas (syngas) fuels derived from coal are particularly promising in this regard. Syngas fuels are typically composed primarily of H$_2$ and CO and may contain N$_2$, CO$_2$, H$_2$O, CH$_4$, and other higher-order hydrocarbons [1,2]. The specific composition depends upon the fuel source and processing technique. These substantial variations in composition and heating value are among the largest barriers toward their usage. Elucidating the impact of this vari-
ability on combustor performance or emissions requires an elucidation of the fundamental combustion properties of these mixtures.

Therefore, the purpose of this study was to better characterize laminar flame speeds of syngas fuels. Laminar flame speed is an important parameter of a combustible mixture as it contains fundamental information on reactivity, diffusivity, and exothermicity. The value of the flame speed has important impacts upon the propensity of a flame to flashback and blowoff and controls other key combustion characteristics, such as the flame’s spatial distribution.

Several prior studies have initiated measurements of the flame speeds of syngas-type mixtures. Laminar burning velocities of syngas mixtures have been measured with Mach Hebra nozzle burners [3] and with Bunsen burners [4]. Laminar flame speeds of CO/H2 mixtures have also been measured with spherically expanding flames [5] and flat flames [6]. Most of these flame speed measurements are for stoichiometric and fuel-rich mixtures, while many modern low-emissions combustion approaches, especially in gas turbines, emphasize lean premixed combustion. There is also substantial scatter in the reported data that is not explained solely by measurement uncertainties [7].

Stretch-corrected measurements of laminar flame speed in H2/CO counter flow flames [8] and spherically expanding flames [9–13] have been obtained more recently and are in fair agreement with each other. However, they also cover a limited range of equivalence ratios and relative H2/CO concentrations and, most significantly, are restricted to room temperature reactants. Furthermore, most of the measurements are for atmospheric pressures; an exception is the work of Hassan et al. [12], who measured flame speeds at pressures up to 5 atm. Similarly, limited measurements for fuels with CO2 dilution are available. Some measurements and computational studies of CH4 diluted with CO2 (to simulate landfill gas) have been reported [14,15]. Few data, however, for H2/CO mixtures diluted with CO2 are available.

Clearly, there is a need to extend the range of available flame speed data for syngas mixtures, particularly at realistic engine conditions. Obtaining such measurements is the primary objective of our study. In addition, the measured flame speeds are compared to model predictions based on two detailed mechanisms: GRI-Mech 3.0 [16], which includes reactions relevant to the combustion of H2, CO, and light hydrocarbons (e.g., CH4 and C3H8), and a simpler chemical mechanism optimized for H2/CO mixtures [17]. This paper describes laminar flame speed results for H2/CO mixtures over wide ranges of fuel composition (i.e., H2:CO ratio), equivalence ratio, CO2 dilution, reactant preheating (unburned temperatures up to 700 K), and pressure (up to 5 atm). Two flame speed measurement approaches are employed: an indirect technique that measures the luminous flame area of a conical flame and a more time-consuming, direct velocity measurement in a stagnation flame. The indirect method is used to determine flame speeds over a wide range of conditions, while the stagnation technique is used at a limited set of conditions identified from the first approach. The stagnation flame approach is more accurate than the conical flame approach; moreover it allows us to obtain the strain sensitivity of the unburned flame speed.

2. Experimental facility

2.1. Bunsen flame

Fig. 1 is a schematic of the experiment used for the laminar flame speed measurements. The desired fuel composition is first prepared using a bank of calibrated rotameters, one for each gas. After mixing thoroughly, the fuel is split into two flows: the desired flow rate of fuel passes through another rotameter (calibrated for the particular fuel composition), while the remainder is flared in a diffusion flame. Finally, the required quantity of air is added, and the mixture goes to the burner. This arrangement allows simple control over the equivalence ratio (Φ) and average velocity through the burner while maintaining the desired fuel composition. All the rotameters are calibrated with a bubble flow meter and wet test meter to ±1% accuracy, with fuel and air flows in the range of 0.1 to 50 slpm.

Various burners are employed; each is a straight cylindrical stainless steel tube, with inner diameter (D) ranging from 4.5 to 18 mm. The length of each tube is at least 50D to ensure that the flow is laminar and that the exit velocity profile is fully developed. The burner diameter is chosen to ensure that the flow remains laminar (Reynolds number, ReD < 2000) and that the average velocity is at least five times greater than the estimated laminar flame speed. The reactants are preheated by electrical resistance tape wrapped around the burner. Once the desired reactant temperature is achieved (as determined by a type-K thermocouple, TC2, temporarily placed at the center of the burner exit), the surface temperature of the burner is monitored by a second thermocouple, TC1, and held constant by a temperature controller. The mixture temperature at the exit of the burner has a nearly uniform radial profile (∆T ≈ 3–5 K).

Digital images of the flame emission are captured with a 12-bit intensified charge-coupled device camera (576 × 384 pixels) and a 105-mm, f/4.5 UV camera lens. The camera system is sensitive in the ultra-
Fig. 1. Schematic of the experimental setup (TC = thermocouple). Mixing is achieved through long flow lines.

Fig. 2. Images of flame emission for various fuels and conditions: (a) H₂:CO = 95:5, Φ = 0.61 without knife edge; (b) same as (a) but with knife edge; (c) H₂:CO = 95:5 and 20% CO₂, Φ = 0.62 without knife edge; (d) same as (c) with knife edge. The color scale is shown to the right.

violet and visible regions (∼220–650 nm) and hence is capable of capturing both OH* and CO₂* chemiluminescence from the flame reaction zone.

Fig. 2 shows some typical images of the flame radiation. The majority of the flame emission comes from the flame edge, i.e., chemiluminescence from the reaction zone. The less intense region in the central portion of the image is due primarily to chemiluminescence from the front and back edges of the flame. The intensity of the flame edge varies along the flame height, mainly due to two causes. First, the integrated flame area decreases along the flame height, which causes the measured flame radiation intensity to decrease. Second, the reactant mixtures studied are lean and contain a considerable amount of hydrogen. Thus, the Lewis number (Le) of these mixtures is expected to be below one due to the high diffusivity of hydrogen. Since negative strain on the flame surface increases downstream along the conical Bunsen flames, the burning intensity for Le < 1 flames is reduced [18]. This reduction in burning intensity can reduce the radiation intensity along the flame height. Moreover for very lean mixtures, a high negative strain at the flame tip can extinguish the flame locally, leading to tip opening [18].
As described below, our flame speed calculation depends on locating the flame reaction zone to determine the reaction zone area. Thus large variations in intensity with height can be problematic. The imaging system includes an unusual feature, a horizontal knife edge (see Fig. 1) placed in front of the lens to vary the collection solid angle along the flame height. Figs. 2a and 2c show the flame emission from high hydrogen content, lean flames acquired without the knife edge, and with a 3-ms exposure time. Locating the flame reaction zone in these images is clearly difficult. The vertical location of this knife edge can be adjusted so as to reduce the amount of light coming from the flame base while the amount of light coming from the flame tip remains unchanged. Then by increasing the exposure time (∼25 ms), the tip of the flame is made visible, without saturating the image at the flame base. The result is seen in Figs. 2b and 2d. The flame tip is clearly more visible, and thus the flame area can be calculated more accurately. These images also show that if flame extinction happens, due to high negative strain, it occurs only at very small flame radius (high curvature). Thus the reaction zone area is only weakly affected.

2.2. Wall stagnation flame

Wall stagnation flames are a well established environment for flame speed measurements [19]. A schematic of the stagnation burner employed here is shown in Fig. 1. Fuel and air flows are monitored and mixed in a fashion similar to that of the Bunsen flame experiments. The desired flow rate of the premixed fuel mixture is sent to the burner while the remainder is bypassed. With this arrangement, the average velocity of the mixture at the exit of the burner is easily adjusted without altering the equivalence ratio. The mixture, after passing through flow straighteners, expands through a smoothly contoured nozzle with high contraction ratio to create a steady, laminar flow with top hat velocity profile, even at high Reynolds number. The exiting fuel/air mixture is surrounded by a N₂ coflow. Various nozzle exit diameters (D = 6.25, 9, and 12.5 mm) are employed to produce a stable flame, with high flame speed mixtures requiring the smaller diameters.

Flow stagnation is achieved with a plug produced from a stainless steel rod (38 mm diameter). The end of the rod is formed into a hemisphere and then removed and machined to produce a flat surface with 12.5 mm diameter. The rounded plug, compared to a flat plate, greatly improves flame stability especially at high pressure and high flame speed conditions (e.g., high preheating). The distance (L) between the burner exit and the stagnation plug is adjusted according to the burning velocity. For high burning velocities, a lower L/D leads to a stable stagnation flame. In the current measurements, L/D ranges from 0.5 to 1 (it should be noted that this corresponds to L/D = 1–2 for the commonly employed counterflow flame configuration). These L/D values are sufficiently large that the effect of finite domain on the measured flame speed can be considered small [20]. The use of a solid wall as a stagnation plane, as opposed to the counterflow configuration with adiabatic twin flames, has an insignificant effect on the measured unburned flame speed, provided that the flame is stabilized sufficiently away from the stagnation plane [19]. The effects of the solid wall are mainly downstream heat loss from the flame products to the wall and zero radial velocity gradient at the wall. In all our experiments, the flame is located at least two flame thicknesses away from the plate (and generally more than five flame thicknesses).

Axial velocity is measured using a laser Doppler velocimetry system. The fuel mixture is seeded with alumina (Al₂O₃) particles. The nominal size of these particles is chosen to be 1–2 µm to minimize thermophoretic effects [21]. The radial profile of axial velocity and the centerline axial velocity gradient were measured close to the nozzle exit to establish the boundary conditions at the nozzle exit for the simulations. The measurements show less than 15% variation of the axial velocity along the radial direction. Also, the axial velocity gradient along the centerline approaches zero at the nozzle exit. This confirms that the outflow from the high contraction ratio nozzle is nearly a plug flow, as expected.

The burner is wrapped in electrical resistance tape to preheat the reactants. Once the desired reactant temperature is achieved (as determined by a type-K thermocouple, TC₄, placed at the center of the burner 25 mm below the exit), the surface temperature of the burner is monitored by a second thermocouple, TC₃, and held constant by a temperature controller. The mixture temperature at the exit of the burner, like the axial velocity, has a nearly uniform radial profile (∆T ≈ 3–5 K).

3. Flame speed measurements

3.1. Bunsen flame approach

The laminar flame speed is defined as the velocity that a planer flame front travels relative to the unburned gas in a direction normal to the flame surface [18]. Though the laminar flame speed is straightforward in definition, in practice it is difficult to measure. Hence some assumptions have to be made in its measurement. A flame stabilized on the rim of a
cylindrical burner is conical in shape and not one-dimensional (1d). This conical flame is affected by strain and curvature; their influence on local flame speed depends on the Markstein length of the mixture [18]. As such, our flame speed measurement is an area-weighted average over the entire flame surface. The average flame speed is calculated by dividing the volume flow rate of the mixture by the luminous cone surface area (flame reaction zone area). It is clear from the definition of the unburned laminar flame speed that the true flame area should be the unburned flame area, just upstream of the preheat zone of the flame. Though the unburned flame area can be measured with the schlieren technique, this does not address the problem that the conical flame is strongly affected by curvature and stretch. Thus the measured flame speed would still not be the 1d flame speed. However, as outlined below, use of the reaction zone area to calculate the flame speed can provide a result that more closely matches the unstretched flame speed.

Sun et al. [22] derived the sensitivity of the unburned and reaction zone flame speeds (\(S_u\) and \(S_b\), respectively) for a curved flame traveling in a nonuniform flow field with a generalized integral analysis that includes thermal expansion in the preheat zone and neglects higher-order terms. Generally the flame speed is affected by flame movement (\(R\)), strain (\(\kappa\)), and pure curvature (\(\gamma\)). For a stationary flame, they showed that the burned flame speed at the reaction zone is affected only by strain, while the unburned flame speed is affected both by strain and by pure curvature effects. Their analysis produces the expressions for the unburned (\(S_u\)) and reaction zone (\(S_b\)) flame speeds relative to their 1d values (\(S_u^0\) and \(S_b^0\))

\[
\frac{S_u}{S_u^0} = 1 + \frac{Ze}{2} \left( \frac{1}{Le} - 1 \right) \frac{\alpha^0 \delta_T^0}{S_u^0} + \gamma \delta_T^0 \quad \text{and}
\]

\[
\frac{S_b}{S_b^0} = 1 + \left[ \frac{Ze}{2} \left( \frac{1}{Le} - 1 \right) - \frac{1}{Le} \right] \frac{\alpha^0 \kappa \delta_T^0}{S_u^0},
\]

where \(Ze\) is the Zeldovich number, \(\alpha\) is a factor that accounts for thermal expansion, \(\gamma = \nabla T \cdot n\) is the curvature of the flame front,

\[
\kappa = \nabla T \cdot \left[ \frac{\mu_T}{\mu} \right]
\]

is the strain rate, and \(\delta_T\) is the flame thickness. Since \(S_b\) is affected only by flame strain, the effect of strong azimuthal curvature in our conical flame case should not influence the flame speed at the reaction zone. Considering the effect of flame strain on \(S_b\), Choi and Puri [23] have shown that the magnitude of the strain rate measured at the reaction zone in the shoulder region of the conical flame is much less than that at the tip, and its effect on the reaction zone speed is minimal. All the flames reported here were stabilized with the highest possible velocity, such that the heights of the flames are large compared to the burner diameter. This reduces the ratio of strain-affected flame tip area to the flame shoulder area. Hence considering both curvature and strain effects, it can be concluded that the measured flame speeds at the reaction zone for the conical flame should be very close to the 1d reaction zone flame speed. Hence the mass balance for the conical flame can be shown as

\[
S_b^0 \approx S_b = \frac{\dot{m}}{\rho_b A_b},
\]

where \(\rho_b\) is the density and \(A_b\) is the surface area at the end of the heat release zone. From a 1d flame mass balance,

\[
S_b^0 = \frac{\rho u}{\rho_b S_u^0},
\]

and this expression for \(S_b^0\) can be substituted into the former expression to produce

\[
S_u^0 \approx \frac{\dot{m}}{\rho_u A_b} = \frac{\dot{Q}}{A_b},
\]

where \(\dot{Q}\) is the volumetric flow rate of the unburned mixture. Since chemiluminescence is primarily produced in the thin heat release zone of the flame, the surface area measured from a chemiluminescence image can approximate \(A_b\). Hence it can be seen that for a conical flame, the flame speed calculated by dividing the volumetric flow rate of the mixture by the luminous cone surface area should closely approximate the unstretched (1d) unburned laminar flame speed.

An edge detection program was developed to determine \(A_b\) from the chemiluminescence images. Since these Bunsen flames are essentially axisymmetric, each flame image is split in half along the burner axis. The edge detection program detects the reaction zone edge by locating the maximum derivative of the flame intensity along the radius of the flame. The flame area is then found by revolving the detected edge along the axis of the burner. The same procedure is repeated for the other half of the flame image (the change between these two areas is always below 1%). For each experimental condition, 25 images are typically recorded, and the reported flames speeds are based on the average of the 50\(A_b\) values (25 images × 2 half-flames). The maximum deviation of the measured flame area (for a single image) from the average of all the images is always less than 2% and this can be represented as the uncertainty in the measured flame speed.

To validate this approach, experiments were conducted for two H\(_2\):CO fuel compositions (50:50 and 5:95 by volume) previously measured with a spherically expanding flame method [9,12,13]. The burner diameters used for these two compositions were 4.5
Fig. 3. Measured laminar flame speeds in Bunsen flame for H$_2$:CO 50:50 and 5:95 compositions at $p = 1$ atm and $T_u \approx 300$ K, including previous spherical flame experiments [9,12,13].

Fig. 4. Measured axial velocity along the stagnation streamline for H$_2$:CO 50:50 fuel mixture with equivalence ratio $\Phi = 0.58$. Figure inset shows layout of nozzle-generated wall stagnation flame.

3.2. Wall stagnation flame approach

To illustrate this method, the measured axial velocity along the stagnation streamline for a H$_2$:CO 50:50 fuel mixture with equivalence ratio of 0.58 is shown in Fig. 4. The axial velocity decreases from the exit of the nozzle and reaches a minimum where the preheat zone starts. After reaching a local minimum, the axial velocity increases sharply inside the flame and then decreases to zero at the wall. Based on a common approach [24], the minimum velocity before the preheat zone is considered the reference strained unburned flame speed ($S_u$). Also, the maximum gradient of the axial velocity in front of the flame, which occurs just before the minimum velocity location, is taken as the imposed strain rate ($K$) (see Fig. 4). The

and 13.6 mm, and the equivalence ratio was varied from 0.6 to 1. As seen in Fig. 3, the measured flame speeds for the 50:50 H$_2$:CO fuel mixture are in good agreement with values obtained from all stretch-corrected spherically expanding flames throughout the lean equivalence ratio range tested. For example, the reported stoichiometric flame speed [9] for the 50:50 mixture is 115 cm/s; the present measurement is 112 cm/s (a 2.6% difference). The 5:95 results are also in close agreement, though with slightly greater differences (<10%) near stoichiometric mixtures. Overall, these comparisons indicate that the errors in flame speed measurement associated with the reaction-zone-area-based Bunsen approach are small, and the technique is reasonably accurate for a wide range of lean H$_2$/CO fuel mixtures.
imposed strain rate is controlled by changing the nozzle exit velocity. As the nozzle exit velocity increases, the flame moves closer to the stagnation surface. For each fuel mixture, the strain rates and corresponding strained flame speeds are measured for a range of nozzle exit velocities. Effort has been taken to measure the strained flame speeds at low strain rates, limited either by flashback or flame stability (unsteadiness). The uncertainty in the strained flame speed measurement can be estimated from the rms fluctuation of the axial velocity at the location where the average velocity is a minimum. At each location along the stagnation stream line 10,000 measurements have been taken and the rms fluctuation is less than 3% for all the conditions reported here.

4. Flame speed modeling

To identify potential regions for improvement of flame speed models, the experimental results are compared to predictions of standard models. Unstrained 1d laminar flame speeds are calculated with the PREMIX algorithm of the Chemkin package, while premixed, 1d strained stagnation flames are simulated with the Chemkin OPPDIFF code. To simulate the stagnation wall flame, the distance between the nozzle and the stagnation plane (L) was matched to the experimental value, since it can have a significant effect on the predicted strained flame speed [20]. The flame speed and strain rate are determined from the stagnation simulation using the same definitions and methods applied to the experimental data. In all the flame simulations, the converged solution was obtained for a large number of grid points by considering the gradient and curvature to be 0.1.

The plug flow boundary condition, which is a close representation of the measured nozzle data, is used in the OPPDIFF simulation. While there is evidence [25] that the inflow boundary condition can have a significant effect on predicted extinction strain rates, its influence on flame speed determination is less [26]. To determine the influence of nozzle exit boundary conditions on the flame speeds in this study (defined as noted previously as the minimum velocity ahead of the flame), detailed numerical simulations were performed with both plug flow and potential flow boundary condition in the manner described in [26] for conditions (fuel mixture, Φ, L, and strain rate) representative of the current experiments. For a fixed strain ahead of the flame, the results show that the minimum velocity is only weakly affected (less than 1%) even by the complete change in nozzle boundary conditions. Therefore, the small difference between the measured nozzle exit conditions and the plug flow boundary used in the simulations can be considered negligible in comparisons of the experiments and modeling results. This conclusion would likely change if the flame conditions were close to the extinction limit, which they are not in the current study.

Two reaction mechanisms are employed: GRI Mech 3.0 [16] and the H2/CO mechanism of Davis et al. [17]. The GRI mechanism has been tested and validated extensively for methane and natural gas combustion over a wide range of pressure and temperature conditions. It consists of 325 elementary chemical reactions with associated rate coefficients and thermochemical properties for the 53 species involved. The second, recent mechanism was developed specifically for H2/CO combustion. It consists of 14 species and 30 reactions and incorporates recent updates for rate parameters and third-body efficiencies of a few key reactions. It also includes modifications of thermodynamic and transport properties for species relevant to high-temperature H2 and CO oxidation. In all the simulations, multicomponent diffusion and Soret effects have been included, as they have significant influence on the calculated flame speeds, especially for high H2 content flames. Generally, multicomponent and thermal diffusion effects increase the predicted flame speeds for lean mixtures and decrease values for near-stoichiometric mixtures compared to predictions employing a mixture average diffusion model.

5. Results and discussion

One of the prime objectives of the present work is to measure the flame speed for syngas compositions with varying levels of CO2 dilution and preheating under lean conditions. The effects of CO2 dilution and unburned temperature (Tub) are examined for three different H2:CO compositions: 95:5, 50:50, and 5:95 ratios by volume. These compositions were chosen to cover a broad range of syngas mixture variations and to aid in validation (or improvement) of syngas flame speed models.

5.1. Effect of CO2 dilution

The equally weighted, 50:50 H2:CO, fuel mixture was tested with 0 and 20% CO2 dilution. The burner diameter used for these mixtures was 4.5 mm. For very lean mixtures with 20% dilution, the tip of the flame becomes less intense, and hence the knife edge was used to make the tip more visible. Fig. 5 shows the measured flame speeds of these two mixtures, with horizontal error bars indicating the uncertainty in the measured equivalence ratios associated with the flow metering uncertainties. The flame speed increases with equivalence ratio and decreases with
CO₂ dilution. This is not surprising, as dilution with excess air or CO₂ decreases the flame temperature, which reduces the rate of CO and H₂ oxidation reactions and hence flame speed. Fig. 5 also shows the flame speeds for these mixtures computed with the two reaction mechanisms. Generally the GRI predictions are lower than the measurements for both diluted and undiluted mixtures. The calculated speeds are in good agreement (∼5%) with the measured values over the whole equivalence ratio range tested for the undiluted fuel mixture. For 20% CO₂ dilution, however, the discrepancies between the measurements and the GRI results increase, especially for leaner equivalence ratios (0.6–0.8), where the model underpredicts the measurements by 10 to 15%. On the other hand, the Davis H₂/CO mechanism is able to more accurately predict the data, within ∼5% for the undiluted mixture and ∼7% for 20% CO₂ dilution (see Fig. 5). The Davis H₂/CO mechanism shows better agreement with the diluted data at lean conditions than GRI, while it matches the GRI simulations closer to stoichiometric mixtures. Like the GRI results, the Davis H₂/CO mechanism values consistently underpredict the experimental data for 20% CO₂ dilution. At a minimum, the predicted flames speeds can be characterized as being in reasonable agreement with the data for atmospheric pressure, ambient temperature conditions.

Since the model predictions systematically underpredict the diluted data using both chemical mechanisms, with greater fractional differences for the leanest mixtures, this fuel composition was also studied in the stagnation flame configuration. The burner nozzle diameter \( D \) was 12.5 mm with \( L/D = 1 \). Fig. 6 shows the variation of measured flame speed with strain rate at three equivalence ratios ranging from 0.6 to 0.8. Also shown are the corresponding strained flame speeds predicted with the two kinetic mechanisms. As in the Bunsen flame results, the leanest mixtures still show the greatest fractional difference between experiments and predictions, however, the models now tend to overpredict the flame speeds (except at high \( \Phi \) and low strain).

Both the measurements and the predictions show the flame speeds linearly increasing with strain rate. The mixture strain sensitivity (\( L_M \)), a measure of the sensitivity of the flame speed to strain, can be found from the slope of these lines, i.e.,

\[
S_u = S_u^0 - L_M \kappa.
\]

While the measurements indicate similar values of \( L_M \) for \( \Phi = 0.68 \) and 0.78, the strain sensitivity for the leanest mixture (\( \Phi = 0.59 \)) is nearly twice as large. There is also a significant difference between the observed and the predicted strain sensitivities. Table 1 lists the measured and predicted \( L_M \) values for this medium H₂ level fuel composition at the three equivalence ratios. The predicted strain sensitivity by the two mechanisms are very similar at all \( \Phi \) but roughly twice the measured strain sensitivity at \( \Phi = 0.68 \) and 0.78 and 1.5 times the value at \( \Phi = 0.59 \).

Unstrained flame speeds are commonly determined from strained flame measurements by extrapolating the measured flame speeds to zero strain rate. Table 2 compares the Bunsen flame measurement and the linearly extrapolated strained flame speeds for this fuel composition at all three equivalence ratios.

There is remarkably good agreement between the two distinct measurement approaches; for the experimental data, the difference is less than 3%. At this point it should be considered that there is some uncertainty in the way that the unstrained flame speed is calculated in the stagnation flame technique. Ide-
ally the strained flame speeds (from OPPDIFF) extrapolated to zero strain rate should equal the unstrained flame speeds predicted in the PREMIX simulation. However, significant differences, as much as 8.4%, are seen between these two predictions (see Table 2). For both mechanisms, the differences increase with a reduction in $\phi$, and the linearly extrapolated strained speeds always overpredict the PREMIX results. This disagreement between the unstrained model (PREMIX) and the strained extrapolation may simply be a result of the somewhat arbitrary definition of the unburned strained flame speed as the minimum velocity point in the approaching velocity profile. In addition, the increasing discrepancy between the models for leaner mixtures may be attributable to the corresponding increase in flame thickness. Although large $L/D$ values were employed (in the experiment and computations) finite domain effects may be influencing the strained flame results at the leanest equivalence ratios. Due to this significant uncertainty in extrapolating the stagnation flame experiment technique, this technique is used to verify the measured flame speeds only at conditions where there is a large discrepancy between the PREMIX and the Bunsen flame data. Moreover, because of this finite domain effect, all the remaining strained flame speed measurements are compared only with the corresponding strained flame speed prediction in the same strain rate range.

Table 1
Measured and predicted unburned strain sensitivities $L_M$ for a 40:40:20 H$_2$:CO:CO$_2$ fuel mixture at three lean equivalence ratios

<table>
<thead>
<tr>
<th>$\phi$</th>
<th>Experiment</th>
<th>GRI Mech 3.0</th>
<th>Davis H$_2$/CO Mech</th>
</tr>
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<tbody>
<tr>
<td>0.78</td>
<td>−0.0131</td>
<td>−0.0247</td>
<td>−0.0240</td>
</tr>
<tr>
<td>0.68</td>
<td>−0.0128</td>
<td>−0.0311</td>
<td>−0.0294</td>
</tr>
<tr>
<td>0.59</td>
<td>−0.0250</td>
<td>−0.0399</td>
<td>−0.0371</td>
</tr>
</tbody>
</table>

Table 2
Measured and OPPDIFF predicted unstrained flame speed by linearly extrapolating to zero strain and their comparison with Bunsen flame measurements and PREMIX predictions for 40:40:20 H$_2$:CO:CO$_2$ mixture at three different equivalence ratios

<table>
<thead>
<tr>
<th>$\phi$</th>
<th>Bunsen flame</th>
<th>Stagnation flame</th>
<th>%Δ</th>
<th>PREMIX</th>
<th>OPPDIFF</th>
<th>%Δ</th>
<th>PREMIX</th>
<th>OPPDIFF</th>
<th>%Δ</th>
</tr>
</thead>
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<tr>
<td>0.78</td>
<td>57.4</td>
<td>59.1</td>
<td>3.0</td>
<td>52.5</td>
<td>54.4</td>
<td>3.6</td>
<td>53.5</td>
<td>55.7</td>
<td>4.1</td>
</tr>
<tr>
<td>0.68</td>
<td>43.9</td>
<td>45.1</td>
<td>2.7</td>
<td>39.7</td>
<td>42.0</td>
<td>5.8</td>
<td>42.0</td>
<td>43.8</td>
<td>4.3</td>
</tr>
<tr>
<td>0.59</td>
<td>31.7</td>
<td>31.0</td>
<td>−2.2</td>
<td>28.6</td>
<td>31.0</td>
<td>8.4</td>
<td>30.9</td>
<td>33.1</td>
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</tbody>
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Fig. 7. Laminar flame speed for fuels with 5:95 H₂:CO composition and 0 and 10% CO₂ dilution at $p = 1$ atm and $T_u \approx 300$ K; Bunsen flame measurements (symbols) and PREMIX predictions (lines).

Fig. 8. Laminar flame speed for fuels with 95:5 H₂:CO composition and 0 and 20% CO₂ dilution at $p = 1$ atm and $T_u \approx 300$ K; Bunsen flame measurements (symbols) and PREMIX predictions (lines).

The high CO content fuel mixture, 5:95 H₂:CO, was tested with 0 and 10% CO₂ dilution. The burner diameter used for these mixtures was 13.6 mm. Fig. 7 shows the flame speeds for these mixtures over a range of lean equivalence ratios. Again, the flame speed increases with equivalence ratio and decreases with CO₂ dilution. For this low H₂ mixture, with and without CO₂ dilution, both mechanisms predictions are essentially the same for all lean conditions, and they are in reasonable agreement with the measurements (within 5–7% over most of the range).

The high H₂ content fuel composition, 95:5 H₂:CO, was evaluated for 0 and 20% CO₂ dilution. Due to the very high flame speeds of these mixtures, the burner diameter was reduced to 4.5 mm. For lean conditions with this high H₂ content fuel, the tip of the flame becomes less intense, and hence the knife edge was used to make the tip more visible for accurate flame area calculations. Fig. 8 shows the measured and computed flame speeds for a range of lean equivalence ratios. The GRI Mech 3.0 predictions are consistently lower than the measured flame speeds. For the undiluted mixture, the discrepancy between the GRI predictions and the measurements is about 15% near stoichiometric conditions and increases to 20%...
as the equivalence ratio decreases to 0.6. The Davis H₂/CO mechanism predictions are similar to the GRI mechanism productions near stoichiometric conditions but, unlike GRI, the deviation from the measurements decreases as the equivalence ratio is reduced. At the leanest conditions studied (Φ = 0.6–0.75), the agreement between the Davis H₂/CO mechanism predictions and the measurements is within 3–4% for the undiluted mixture. Similar behavior is observed for 20% CO₂ dilution. However, as the dilution increases, the agreement between the GRI Mech 3.0 predictions and the measurements improves near stoichiometric conditions but worsens at lean conditions. For 20% CO₂ dilution, GRI underpredicts the measurements by as much as 25–30% at very lean equivalence ratios. On the other hand, the Davis H₂/CO mechanism predicts the measured flame speed within 5% at the very lean equivalence ratio for 20% CO₂ dilution. Moreover, the Davis H₂/CO mechanism predictions are much better for the diluted mixtures near stoichiometric conditions compared to those for the undiluted mixture. Overall, the laminar flame speed predictions with the Davis H₂/CO mechanism are considerably more accurate than those with GRI Mech 3.0 for this high H₂ content fuel mixture, especially at lean equivalence ratios. Since the Davis H₂/CO mechanism, which was nominally optimized for hydrogen mixtures, shows good agreement with measurements for both diluted and undiluted cases, stagnation flame experiments were not conducted for this mixture.

5.2. Effect of preheating

Experiments for lean mixtures with the same three H₂:CO ratios over a range of reactant preheat temperatures from 300 to 700 K were conducted. As the unburned reactant temperature increases, the flame speed should increase due to increased chemical rates and thermal and mass diffusivities. This increase in flame speed requires the burners to be operated at higher average flow velocities (compared to the room temperature case). Fortunately, the increase in the unburned reactant temperature also increases the viscosity of the unburned mixture, which allows the flow to remain laminar even at the higher operating velocities. Hence, the same diameter burners used for the room temperature cases were used for the preheated cases.

The influence of preheat temperature for the 50:50 H₂:CO composition is shown in Fig. 9. The measured flame speeds increase rapidly with the unburned temperature for any given equivalence ratio and are in good agreement (within ~5%) with the GRI Mech 3.0 predictions, up to a preheat temperature of about 500 K. For further increases in preheat temperature, the discrepancy between the measurements and the GRI Mech 3.0 predictions increases. For the 600 K preheat temperature, the simulations with GRI Mech 3.0 overpredict the measured flame speeds by as much as 15% near stoichiometric and 30% at the leanest conditions tested. The flame speeds predicted with the Davis H₂/CO mechanism are also shown in Fig. 9. The Davis H₂/CO mechanism predictions are very similar to the GRI Mech 3.0 predictions except at the leanest Φ, where the Davis H₂/CO mechanism predictions are higher than the GRI predictions by 5%. Thus the Davis H₂/CO mechanism has an even larger overprediction for the undiluted, 50:50 H₂:CO mixture at 700 K preheat temperature.

For comparison, the stagnation flame technique was also used to determine (strained) flame speeds for
the 50:50 H\textsubscript{2}:CO mixture at high preheat (700 K) and at two equivalence ratios (0.6 and 0.8) where large discrepancies between the Bunsen measurements and the model predictions were observed. Due to the very high flame speeds for these mixtures and the need for high hydrodynamic strain rate to produce a stable flame, a small nozzle diameter (6.25 mm) with \( L/D = 0.8 \) was used.

As seen in Fig. 10, the measured strained flame speeds increase linearly with imposed strain rate for both equivalence ratio cases. It is important to note that the flame at \( \Phi = 0.6 \) is more strain sensitive than that at \( \Phi = 0.8 \). This is qualitatively similar to the room temperature results (Fig. 6). While the predicted and measured strain sensitivities are quite similar, the strained flame speeds predicted with both mechanisms are consistently higher than the measurements for both \( \Phi \) (see Fig. 10). This trend is similar to that found for the conical flame results (Fig. 9). The GRI Mech 3.0 calculated strained flame speeds overpredict the measurements by 12\% while the Davis H\textsubscript{2}/CO mechanism overpredicts the measurements by 17\% at \( \Phi = 0.6 \). The predictions improve at \( \Phi = 0.8 \), with the GRI results overpredicting the measurements by 7\% and the Davis H\textsubscript{2}/CO mechanism by 9\%. As with the Bunsen flame results, the discrepancies between the measurements and the model predictions increase as the equivalence ratio decreases for the 50:50 H\textsubscript{2}:CO mixture at high preheat temperature. Yet the amount of overprediction with GRI Mech 3.0 decreases from approximately 30\% in the Bunsen flame case to about 12\% for the stagnation flame measurements at \( \Phi = 0.6 \).

In addition, for the preheat case, unlike the room temperature data, the unstrained flame speeds from the linearly extrapolated stagnation flame data are noticeably higher than the flame speeds measured with the Bunsen flame method (see Figs. 9 and 10). For example at \( \Phi = 0.6 \), the unstrained flame speed obtained from the stagnation data is 301 cm/s, whereas the flame speed determined from the Bunsen flame approach is 250 cm/s. As noted previously, linearly extrapolating the strained flame speeds is known to overpredict the true unstrained flame speed. However, the high preheat temperature also tends to increase the flame thickness, which could lead to a bias in determining the reaction zone flame area for the Bunsen approach that would cause a measurement lower than the true flame speed.

The effect of CO\textsubscript{2} dilution at high preheat temperature (700 K) was studied for this 50:50 H\textsubscript{2}:CO composition with 40\% CO\textsubscript{2} dilution at lean equivalence ratios using the stagnation flame technique. Fig. 11 shows the measured strained flame speeds for this composition at 700 K preheat temperature for \( \Phi = 0.6 \) and 0.8. The leaner case has a higher strain sensitivity than the \( \Phi = 0.8 \) mixture, which is consistent with both the room temperature and the high preheat results for the undiluted 50:50 H\textsubscript{2}:CO mixture. Fig. 11 also shows the predictions for both equivalence ratios. The predictions with both mechanisms are consistently higher than the measurements and the difference decreases with increasing \( \Phi \). In fact, the deviations from the measurements are about the same levels seen in the undiluted, high preheat case; the GRI predictions are 10\% (\( \Phi = 0.6 \)) and 9\% (\( \Phi = 0.8 \)) above the measurements, while the Davis H\textsubscript{2}/CO mechanism results are 14\% (\( \Phi = 0.6 \)) and 12\% (\( \Phi = 0.8 \)) too high. This suggests that the radiation absorption/emission effect of CO\textsubscript{2} addition is not
important for this mixture even at these high preheat, lean conditions (at atmospheric pressure). This agrees with previous literature [27,28] showing the radiation has little influence on measured flame speeds as long as the fuel mixture is not near the flammability limits or at elevated pressure.

Fig. 12 shows the influence of preheat temperature on flame speed for the low hydrogen content fuel (5:95 H₂:CO). As was the case for the no preheat case, the predictions from the GRI and Davis H₂/CO mechanisms are essentially the same at each preheat temperature, and the measured flame speeds are higher than the predictions. The agreement between the measurements and the predictions improve as the equivalence ratio drops. For example, the discrepancies are within 5% for the leanest mixtures (Φ = 0.6–0.7) for preheat temperature up to 400 K. Beyond 500 K, the discrepancy between the measurements and the predictions increases to 10–15% in this lean equivalence ratio range. Because preheating improves flame stability, measurements were possible for even leaner mixtures at high reactant temperatures (for the same burner diameter). Measurements at these very lean conditions are in good agreement with the prediction by both mechanisms.

Similar results for the high hydrogen content fuel (95:5 H₂:CO) are shown in Fig. 13. As the flame speeds are extremely high (>8 m/s) for this mix-
ture with preheating, the velocities needed to stabilize the flames are also high. To reduce the exit velocities and maintain laminar conditions, the fuel stream was diluted with 20% CO\textsubscript{2}, thereby reducing the flame speeds. Hence the results include the effects of both dilution and reactant temperature. The computed flame speeds from the two mechanisms are nearly the same at near-stoichiometric conditions for all the preheat temperatures. However, for lean conditions, the GRI Mech predictions are lower than those from the Davis H\textsubscript{2}/CO mechanism, by as much as 30% at very lean equivalence ratios. As in the room temperature case, the predictions with the H\textsubscript{2}/CO mechanism are in good agreement with the experiments (within 10%) up to 500 K throughout the tested equivalence ratio range. For the 600 K preheat case, the H\textsubscript{2}/CO mechanism overpredicts the measurements across the complete \( \Phi \) range, while the GRI results now better reproduce the measurements. Given the poorer prediction with the GRI mechanism at lower temperatures, the improved agreement at high preheat temperatures may simply be fortuitous.

5.3. Effect of pressure

The effect of higher operating pressure was studied for three H\textsubscript{2}/CO compositions: 5:95, 10:90, and 20:80 at 5 atm and \( \Phi = 0.6 \) using the stagnation flame technique (see Fig. 14). As in the earlier measurements, the flame speed increases linearly with strain.
rate, indicating a negative (unburned) strain sensitivity. As the amount of H₂ increases in the mixture, the strain sensitivity increases. For the 5:95 H₂:CO fuel, predictions produced by both the GRI and the Davis H₂/CO mechanisms are in excellent agreement (less than 5% discrepancy) with the measurements; this is similar to the finding from the atmospheric pressure tests. Similar agreement between the measurements and the predictions is observed for the 10:90 fuel mixture with the Davis H₂/CO mechanism, while the GRI mechanism results slightly underpredict the measurements. For both these low H₂ content fuels, the predicted strain sensitivities also are in good agreement with the measurements. Thus the good agreement observed at atmospheric pressure between the model predictions and the measurements is maintained at this higher operating pressure.

As the amount of H₂ is raised to 20%, the discrepancy between the measurements and the predictions increase. The GRI mechanism now underpredicts the measurements by about 10%. More importantly both mechanisms fail to predict the higher strain sensitivity for this mixture. Recall that the model predictions also failed to accurately capture the strain sensitivities for the atmospheric pressure cases with intermediate H₂ levels (Fig. 6).

6. Conclusions

Laminar flame speeds for lean syngas (H₂/CO/CO₂) fuel mixtures were measured over a wide range of fuel composition and reactant preheat temperature. Most results were obtained at atmospheric pressure; some measurements were carried out at 5 atm. The data, especially the high reactant temperature results, significantly extend the existing database. The measured flame speeds were also compared to laminar simulations based on two leading kinetic mechanisms to verify their validity at gas turbine operating conditions.

Two flame speed measurement approaches were employed: a Bunsen flame method based on images of reaction zone area and a stagnation flame technique employing velocity measurements along the flame axis. Laminar flame speeds measured with the Bunsen technique agreed well with (limited) previous results from expanding spherical flames and with the current stagnation flame results (extrapolated to zero strain) over a wide range of compositions (e.g., 5 and 50% H₂ and 0 and 20% CO₂ dilution). Thus it can be concluded that the easily implemented Bunsen flame technique, based on reaction zone area, is reasonably accurate for a range of H₂/CO fuel mixtures.

For room temperature reactants, the model predictions are generally in good agreement with the experimental results. This is not surprising, as both mechanisms were optimized with validation sets that included room temperature data. For both low and medium H₂ content fuels, the Davis H₂/CO and GRI mechanisms provide similar predictions of laminar flame speeds, though the Davis H₂/CO mechanism predictions are slightly higher at lean conditions for the medium H₂ composition fuels. Both models also predict the measured strained flame speed and strain sensitivity well for low H₂ content compositions. For medium H₂ levels (e.g., 20–50%), however, both the atmospheric and the 5 atm data reveal a significant discrepancy between the measured and the predicted strain sensitivities at very lean conditions (e.g., Φ ≲ 0.6). For high H₂ fuels, the experimental flame speeds are generally higher than the predictions, though the Davis H₂/CO mechanism is quite accurate (within a few percent) for equivalence ratios below ~0.8. For comparison, the GRI results differ from the experiments by as much as 25–30% for very lean mixtures. Overall, the laminar flame speeds for lean mixtures of syngas fuels at room temperature are well predicted (within ~10%) with the optimized Davis H₂/CO mechanism.

With preheated reactants, however, there are noticeable differences between the experimental data and the predictions. For example, the medium H₂ content fuel mixture data are in good agreement with the predictions (from both models) up to 500 K. As the preheat temperature increases to 700 K, larger discrepancies are observed, especially at lean equivalence ratios. For instance, the models overpredict the experimental data by ~25–30% near Φ = 0.6. The strained flame speed measurements at this condition confirm this trend but with smaller discrepancies. For the high H₂ content fuels, the predicted flame speeds again increase with temperature faster than the measured values. Thus while the Davis H₂/CO mechanism shows good agreement with the measurements for preheat temperatures below 500 K, the GRI mechanism’s predictions are fortuitously closer at higher preheat temperatures. For low H₂ content fuels, both models also produce similar results, but now they tend to underpredict the highly preheated flame speeds. The underprediction is not severe, as the model results are in reasonable agreement with the measurements over most of the lean conditions, though the differences are as much as 10–15% at near-stoichiometric conditions.

In summary, the current models tend to overpredict the temperature dependence of the flame speed for medium and high H₂ content fuels but underpredict it for low H₂ (high CO) content fuels. Improving the accuracy of the models would enhance their use in detailed design of gas turbine combustors for syngas fuels. The failure of the current models is most
likely caused by deficiencies in the chemical mechanisms, e.g., errors in the temperature dependence of the rate coefficients for one or more reactions. However, since the two models employed here also use different transport property databases, effects due to errors in the diffusion rates cannot be neglected.

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References