Laminar Burning Velocities of Spherically Expanding Hydrogen/Air Mixtures for Temperatures up to 423 K at Ambient Pressure

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Introduction

Over the last few years, a number of publications have evaluated the performance of chemical mechanisms for hydrogen combustion against experimental data found in the literature for ignition delay times, species profile measurements, and burning velocities [1,2]. Usually, the performance of a kinetic mechanism for individual test systems is quantified according to overall performance indicators. In practice, it could happen that overall performance indicators are mistakenly used instead of specific ones in order to find a well suiting mechanism, e.g. for the calculation of burning velocities. Thus, it is important to demonstrate and discuss differences in the predictions and their origin with respect to the chemical mechanism formulation.

In this study, new experimental laminar burning velocity data for hydrogen / air flames at temperatures of 298 K, 373 K, and 423 K under ambient pressure are presented with equivalence ratios of 0.5, 1.0, 1.5, and 2.0. The experimental data are compared to data found in the literature under ambient conditions. The chemical mechanisms by Burke et al. [3] and Hong et al. [4], typical representative mechanisms utilised for flame speed predictions, are chosen for simulation. According to Olm et al. [1], the Burke mechanism performs very well and the one by Hong et al. is less convincing with respect to flame speed simulations. Reasons for the difference in flame speed simulations are briefly addressed.

Experimental Setup

Experiments were performed using the closed-vessel method combined with an optical Schlieren cinematography setup in order to acquire burning velocities at elevated temperatures and ambient pressure. The experimental setup is shown schematically in Figure 1. The internal shape of the pressure vessel is a sphere with an inner diameter of 100 mm; quartz windows with a diameter of 50 mm are positioned on opposite sides. The edge of the propagating flame is imaged using a dual-field-lens Schlieren arrangement together with a pulsed high power LED (ix) emitting green light, which allows very short exposure times of the high speed CMOS camera (i). This is particularly beneficial for fast burning fuels like hydrogen / air mixtures. Images were taken at 25000 frames per

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Figure 1: Schematic of image acquisition setup using a dual-lens Schlieren arrangement.

second (fps), with 448x448 pixels, and a resolution of 10.33 pixel/mm as well as at 16000 fps, with 576x576 pixels, and a resolution of 14.23 pixel/mm in an earlier measurement campaign. Optical lenses are an aspheric condenser lens (viii) and three spherical lenses (ii, v, vi). Two pinholes (iv, vii) with a diameter of 0.5 mm are used, and overexposure of the camera from flame radiation is prevented by an optical filter (iii). [5]

An external mixing vessel, directly connected to the combustion chamber via pipes, is employed for an external preparation of the air / fuel mixture. The amount of fuel needed can be calculated as a function of ϕ , T, and p. The partial pressure method can be used for accurately measuring and controlling the filling process. In addition, a real gas correction has been applied by using the Soave modification of the Redlich–Kwong equation of state. Before sparking, the heaters are turned off and the mixture is allowed to settle. A two-step ignition system is used for igniting the mixture at the midpoint of the vessel with extended spark plug electrodes of 1 mm diameter. Experimental conditions were set up with dehumidified compressed air, which consists of 20.94 % oxygen, 78.13 % nitrogen, and 0.93 % argon. Fuel was hydrogen of grade 6.0.

Chemical Kinetic Mechanisms and Flame Speed Calculation

The mechanism by Hong et al. [4] consists of 8 species and 20 reactions. The reaction scheme is based on the H_2/O_2 sub-mechanism of the GRI-Mech 3.0 [6]. The Burke et al. [3] mechanism is also based on the same sub-mechanism scheme, except that the reaction $H + HO_2 = HO + O$ is not included. The laminar flame speed calculations were performed using the 1D premixed, freely propagating flame module of the FlameMaster software package [7].

Determination of Flame Speeds

The evaluation of the observed experimental data was restricted to smooth spherical flame fronts with a radius above 7 mm in order to avoid the spark's influence as a result of the ignition process. The flame front was extracted from the captured Schlieren images. The post-processing of the propagating flame images yields information on the expanding flame radius r_f over time t. The stretched propagation speed with respect to the burnt mixture S_b can then be determined by the derivative dr_f/dt . The stretch rate κ is defined as the temporal change of a flame surface area A. In the case of a spherical, outward expanding flame front, κ can be expressed as $\kappa = (1/A)(dA/dt) = (2/r_f)S_b$. A non-linear model is utilized to extract the unstretched flame speed S_b^0 and the burnt gas Markstein length L_b , which is used in this work. It is given by:

$$\left(\frac{S_b}{S_b^0}\right)^2 \ln\left(\frac{S_b}{S_b^0}\right)^2 = -\frac{2L_b\kappa}{S_b^0}.$$
(1)

A least-squares fit is applied to obtain the unknowns, s_b^0 and L_b [8]. The laminar burning velocity S_L is defined as the unstretched flame displacement speed with respect to the unburnt mixture, S_u^0 . It is determined from mass continuity through a planar unstretched flame $S_L = S_u^0 = S_b^0(\rho_b/\rho_u)$. ρ_b and ρ_u are the densities of the burnt and unburnt mixture. ρ_b is evaluated at adiabatic flame temperature conditions.

Figure 2 shows for illustration a typical post-processing graph of a stoichiometric hydrogen/air flame at ambient conditions. The temporal evolution of the flame goes from high to low stretch values. Symbols indicate the results of the post-processed flame images. The dashed line represents the non-linear extrapolation according to Eq. 1 and the solid line the linear extrapolation technique $S_b^0 - S_b = L_b \kappa$ [9]. The extrapolation of the data must be performed from the biggest usable flame radius in the set of measurements towards smaller, higher curvature flames. The solid blue lines represent varying flame isoradii r_f . For the experimental conditions considered in this study, both extrapolation methods yield nearly same results.



Figure 2: Measured flame propagation speed over stretch for a stoichiometric hydrogen / air flame; $p_i = 1$ atm and $T_i = 298$ K; linear and non-linear extrapolation method; flame isoradii at $r_f = 2.1$ cm, 1.5 cm, 1 cm, and 0.7 cm.

Results and Discussion

Images of the spherical flames of stoichiometric hydrogen / air mixtures at 298 K are shown in Figure 3. They are taken at three different flame radii of 0.8 cm, 1.5 cm, and 1.8 cm. In addition, the stretch rate κ and the corresponding flame propagation speed S_b are given. The flame contour remains smooth throughout the entire visible flame radius evolution. In the case of leaner mixtures, first small artefacts are visible on the flame surface resulting from the ignition process. But these artefacts do not influence the propagation speed. This is followed by an increase in cellular structures

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Figure 3: Captured images of spherical hydrogen flames $\phi = 1.0$; $p_i = 1$ atm, and $T_i = 298$ K.

and significant wrinkling of the flame, the leaner the mixture becomes. Flame wrinkling increases the flame surface area, thereby increasing the resulting flame propagation speed, which must be avoided. The constraint of quasi isobaric conditions for post processing must be also taken into account. This is indicated in Figure 2 by a maximum isoradius of 2.1 cm. Experimental data for 1 atm and the corresponding flame speed predictions are shown in Figure 4. The new experimental data increase from $\phi = 0.5$ to a maximum around $\phi = 1.5$ and decrease again at richer mixtures. The simulations with the Hong mechanism slightly overpredict the new ITV experimental data, but



Figure 4: Laminar burning velocities of hydrogen / air flames; p = 1 atm and T = 298 K, 373 K, and 423 K; Simulations: Hong [4]—, Burke [3]---; Measurements: ITV •, Huang [10]•, Aung [11]•, Tse [12]•, Pareja (p = 0.96 bar) [13]•, Dayma [14]•, Kwon [15]•, Hu [16]+, Qin [17]×.

both closely follow the same trend. The data from Tse et al. [12] also follows the simulations nicely for rich mixtures. The predictions with the Burke mechanism significantly overpredict the data from this work. The data from Dayma et al. [14], however, follow a different trend. In general, for lean Beeckmann, J.

to stoichiometric mixtures, the data by Dayma et al. [14], Hu et al. [16], and this study yield results within a range of less than 5%. Also, the predictions are within that range. The same trend holds true for higher temperatures. Experimental data by Qin et al. [17] and by Pareja et al. [13] are always significantly faster compared with other experimental data as well as with the predictions. One reason might be the uncertainty of the experimental method used for the data acquisition. Qin et al. and Pareja et al. use a bunsen type and a small slot burner setup, respectively. The others use spherical vessel setups. The differences in experimental data comparing the spherical vessel setups on the rich side are not understood yet and need further investigation. When comparing the two mechanisms with each other, more than half of the reactions incorporated are identical. The dissociation of the molecular hydrogen into hydrogen atoms, the reaction of a hydrogen radical with molecular oxygen, the hydrogen peroxide dissociation into hydroxyl, and the hydro peroxide reaction with a hydrogen or an oxygen atom are the main differences. Again, further work needs to be conducted to evaluate each reaction rate's validity with respect to laminar burning velocity.

Concluding Remarks

Laminar burning velocities were measured for hydrogen / air mixtures in a spherical combustion vessel at 1 atm and temperatures of 298 K, 373 K, and 423 K. The experimental results were presented in comparison with computed flame speeds performed with two detailed chemical mechanisms. The laminar burning velocity predictions using the Hong mechanism agree very well with the new experimental data, whereas those computed using the Burke mechanism overpedict significantly for rich mixtures. Future work needs to focus on the validity of experimental data with respect to the definition of performance parameters and agreement of predictions with the validated experimental data.

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