



New insights into the peculiar behavior of laminar burning velocities of hydrogen–air flames according to pressure and equivalence ratio



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ABSTRACT

Hydrogen is a clean and energetic fuel, and its oxidation mechanism is a subset of the oxidation mechanisms of all hydrocarbons. Therefore, the validation of the available kinetic schemes is of great importance. In the current study, experimental measurements of laminar flame speeds and modeling studies were performed for H₂–air premixed flames over a wide range of equivalence ratios (0.5–4.0) and pressures (0.2–3 bar). The large scale in mixture and thermodynamic conditions allows a better understanding of the peculiar behavior of hydrogen flame speeds with pressure. Two very recent detailed chemical kinetic mechanisms for hydrogen combustion were selected. Excellent agreement was observed between calculations and experimental results, confirming the validity of the kinetic schemes selected. The kinetic analyses performed allow proposing an explanation for the nonmonotonic variation of hydrogen/air flame speed with pressure observed in the experiments.

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1. Introduction

Description of hydrogen oxidation is important for at least two reasons: hydrogen is a renewable fuel, biologically or chemically derived, which burns and cleanly, and its oxidation mechanism is a basic building block required in oxidation mechanisms of more complex fuels. Extensive research has been conducted experimentally [1–5] and numerically [6–11] on the reactivity of hydrogen/air and hydrogen/oxygen mixtures, including diluted mixtures and high-pressure conditions. Although the combustion of hydrogen under standard conditions is globally well simulated, the effects of pressure and equivalence ratio variations are less accurately captured [12]. Furthermore, whereas recent efforts concerned high-pressure flames, only a few data have been obtained under reduced pressure, as presented in Table 1, where nonmonotonic variation of flame speed with pressure has been reported [13]. The subatmospheric conditions, specifically around 200 mbar, are of particular importance to guarantee safe operation of the International Thermonuclear Experimental Reactor (ITER) [14]. Updates of hydrogen kinetic reaction mechanisms have been presented recently by Hong et al. [15], Burke et al. [12], and Keromnès et al. [16]. The model of Hong shows significant improvements regarding the prediction of ignition delay but brings no further

resolution to discrepancies observed for flame speeds [15]. Though mainly focused on high pressures, the mechanisms of Burke et al. and Keromnès et al. were considered in the current study. Experimental measurements of laminar flame speed, analysis of the key reactions and kinetic pathways, and modeling studies were performed for H₂–air premixed flames over a wide range of conditions: equivalence ratios ranged from 0.5 to 4.0 and pressures from 0.2 to 3 bar. The experimental matrix was selected to check the validity of the two mechanisms over a wide range of pressures and equivalence ratios and to better understand the peculiar behavior of hydrogen flame speeds with pressure.

2. Experimental setup and data processing

Laminar flame burning velocity measurements were performed using a stainless steel spherical combustion chamber with an inside volume of 4.2 L, as used previously [24]. Four windows provided optical access into the chamber. Before being filled, the combustion chamber was evacuated. After each combustion event, the combustion chamber was vented with compressed air for 2 min to decrease the chamber temperature and to flush the residual gases. Then the chamber was filled with synthetic air and pure hydrogen. The composition of synthetic air was (in volume) 20.9% O₂ and 79.1% N₂. The concentrations of other species (H₂O, CO₂, CO, NO) were lower than 100 ppb. Compressed hydrogen had a purity of 99.95%, with less than 20 ppm of O₂ and H₂O. Air and hydrogen

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were injected using thermal mass flow meters (2 NL/min for air and 0.5 NL/min for hydrogen). These flow meters were used in their full operation scales. The global precision was 1% (0.7 in the measure and 0.3 in the repeatability). This leads to a maximal error in equivalence ratio of 2%.

The temperature of the chamber was regulated at a desired value of $T_u = 303$ K. Two different temperatures are monitored with K-type thermocouples: one on the inner surface of the chamber and the other inside the chamber (3 cm away from the inner chamber surface). The chamber temperature regulation uses the first thermocouple. The temperature measured by the second thermocouple is assumed to be representative of the initial fresh gas temperature. Moreover, the temperature homogeneity inside the chamber was validated using three thermocouples located all around the inner volume. During the experiments, temperature variations were lower than 1%. The compressed air flow flushing the chamber after each ignition was very helpful in keeping temperature constant.

Inside the chamber, an electric fan mixed the reactants. To avoid any disturbances from the homogenization system (fan) and achieve a perfect homogenization of the mixture (both in temperature and in composition), a delay of 2 min was imposed prior to ignition. Two tungsten electrodes separated by 1 mm, connected to a capacitive discharge ignition system, were used for spark ignition at the chamber center. To avoid confinement effects, the maximal flame radius considered in the flame velocity determination was fixed at 25 mm. This value was selected based on previous work [25], where a criterion of one-third of the chamber diameter was proposed. Moreover, the chamber was equipped with a rapid pressure sensor (Kistler 7001). In all experiments, the pressure increase in the chamber during the flame front visualization was less than 2%. Thus, the inside chamber total pressure can be considered constant.

For small flame radii, propagation is strongly affected by the initial energy deposit. The propagation speed then decreases and starts to evolve autonomously. The critical radius corresponding to this transition was found here to be less than 6.5 mm. One can assume that beyond this critical radius, the effect of the initial energy deposit is negligible. Certainly this critical value may be different as a function of the air–fuel mixture, but of the same order of magnitude. The value determined here was corroborated by a critical value previously suggested [26]. It was reported that the propagation speed becomes independent of igniting energy for radii greater than 6 mm. In all our postprocessing, an initial radius of 8 mm was assumed. For each condition, the measurements were repeated three times. The standard deviation corresponding to the scattering in the experiments (± 2 cm/s) is contained in the symbol size.

Shadowgraphy was used to record flame images. Parallel light was obtained from an Ar-ion laser source with two plano-convex lenses of focal length 25 and 1000 mm, respectively. Shadowgraphic images recorded using a high-speed video CMOS camera (Photron APX) operating at 20,000 frames per second with an exposure time of 20 μ s were used to analyze the temporal evolution of the expanding spherical flames. After the spark, the flame front propagates spherically and the temporal flame radius evolution is postprocessed using a nonlinear relation between the flame propagation speed and the stretch, as described in [27]. Finally, the laminar speed is evaluated from the propagation speed, using the expansion ratio. The densities of the burned gases were evaluated using the chemical composition at the constant pressure adiabatic conditions.

The experimental conditions of the present paper are summarized in Table 2. Experiments were performed under varying equivalence ratio (ER) and pressure (P) conditions, respectively from 0.5 to 4.0 and from 0.2 to 3 bar. For fuel-lean conditions and low

Table 1

Previous studies on laminar burning velocities of H₂–air flames under subatmospheric pressures.

Pressure (atm)	Temperature (K)	Equivalence ratio	Refs.
0.025–0.987	300	0.26–5.56	[13]
0.25/0.5	300	0.4–1.0	[17]
0.25/0.5	373	2.6–5.0	[18]
0.35/0.5	298	0.45–4.0	[19]
0.5	298	1	[20]
0.5	298	0.4	[21]
0.6	300	0.8–1.2	[22]
0.767	298	0.6–4.0	[23]

pressure, the flame propagation was relatively difficult to track because of the low signal received by the high-speed video camera. A minimal pressure of 0.2 bar was imposed in the current study because of luminosity considerations. We report in Fig. 1 some events of the flame propagation for different pressures and equivalence ratios. The limitation in flame detection described previously is also explicitly illustrated in Fig. 1. Another experimental limitation encountered can also be observed in Fig. 1. For an equivalence ratio of 0.5 (i.e., low-Lewis-number mixtures), the flame structure presents cellularities when pressure is increased. To avoid overestimation of the propagation speed and thus of the laminar burning velocity, the maximal radius considered in the nonlinear extrapolation was adjusted before the occurrence of flame instabilities. In Table 3, the maximal flame radius considered for the velocity evaluation is indicated for the cases impacted by instabilities. This radius corresponds to a moderate wrinkling of the flame front. For all the other cases, a maximal radius of 25 mm was considered.

3. Computational methods

Two very recent and extensively validated kinetic reaction mechanisms were selected to model our experimental results. The first mechanism, proposed by Burke et al. [12], is an updated version of that proposed by Li et al. [6]. This chemical kinetic mechanism consists of 19 reactions involving 9 species and was specifically developed for high-pressure combustion. The second mechanism, proposed by Keromnès et al. [16], is an updated version of the mechanism presented earlier by Ó Conaire et al. [10]. This chemical kinetic mechanism also consists of 19 reactions and 9 species. It has been extensively validated over a large number of experimental conditions but also mainly focused on high pressure. Both of these mechanisms were used with their own thermodynamic and transport data.

Computations were performed using the CHEMKIN package and the PREMIX code [28]. All the laminar burning velocities were calculated using the multicomponent option to formulate transport properties and taking thermal diffusion into account. The same conditions of gradient and curvature (GRAD = 0.01 and CURV = 0.01) leading to a similar number of meshes (ca. 1000) for each equivalence ratio or pressure were also considered. These conditions allow the computation time to be very short (ca. 30 s) for each condition and the calculated laminar burning velocity to

Table 2

Experimental conditions of the present study.

Mixture	T (K)	P (bar)	ER (–)
H ₂ /air	303	1	0.5–4
		0.2–2	0.5
		0.2–1.3	1
		0.3–1.7	1.7
		0.3–3	3

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