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Laminar premixed flat non-stretched lean flames of hydrogen in air

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ABSTRACT

Laminar burning velocity of lean hydrogen + air flames at standard conditions is still a debated topic in combustion. The existing burning velocity measurements possess a large spread due to the use of different measurement techniques and data processing approaches. The biggest uncertainty factor in these measurements comes from the necessity to perform extrapolation to the flat flame conditions, since all of the previously obtained data were recorded in stretched flames. In the present study, laminar burning velocity of lean hydrogen + air flames and its temperature dependence were for the first time studied in stretch-free flat flames on a heat flux burner. The equivalence ratio was varied from 0.375 to 0.5 and the range of the unburned gas temperatures was 278–358 K. The flat flames tended to form cells at adiabatic conditions, therefore special attention was paid to the issue of their appearance. The shape of the flames was monitored by taking OH* images with an EM-CCD camera. In most cases, the burning velocity had to be extrapolated from flat sub-adiabatic conditions, and the impact of this procedure was quantified by performing measurements in H_2 + air mixtures diluted by N_2. The effect of extrapolation was estimated to be of negligible importance for the flames at standard conditions. The measured burning velocities at 298 K showed an important difference to the previously obtained literature values. The temperature dependence of the burning velocity was extracted from the measured results. It was found to be in agreement with the trends predicted by the detailed kinetic modeling, as opposed to a vast majority of the available literature data.

1. Introduction

Since the early studies comparing measurements of the flame propagation in hydrogen + air mixtures and model predictions, e.g., [1,2], it was observed that even when a model agrees well with experimental results over a wide range of equivalence ratios, important disagreement remains for very lean near-limiting flames. Significant spread in the experimentally determined burning velocities obtained before 1980's was attributed to the impact of stretch and curvature, because all popular methods for measuring burning velocities, such as Bunsen burner or spherical flames, explored non-planar flame configurations. Nowadays, the importance of stretch correction in the interpretation of experimental data obtained in spherical or counterflow flames is well recognized, yet alarming disagreement is still not resolved [3,4]. This remaining disagreement in experimental determinations means that evaluation of the kinetic models at these conditions is highly uncertain.

Remaining scattering of the stretch-corrected burning velocities, S_L, obtained in lean hydrogen + air flames is illustrated in Fig. 1, which shows available data [5–23] at standard conditions, i.e., pressure of 1 atm and unburned gas temperature $T_g = 298$ K, and modeling results obtained with two recent hydrogen models [24,25]. The color codes denote the measurement method and stretch-correction model implemented: green – spherical flame, linear model [26]; blue – spherical flame, non-linear model of Kelley and Law [27]; red – counterflow burner, linear model (LM) originated from [28]; orange – counterflow burner, non-linear extrapolation (NLM) based on the study of Tien and Matalon [29] for the data of Das and Sung [22], and on the work of Wang et al. [30] for the data of Park et al. [23].

It is well known that lean hydrogen flames, even subject to stretch, are highly unstable. Bradley et al. [11] go as far as questioning the utility of S_L for these unstable flames, especially at high pressures. Formation of flame balls, an extreme limit of evolution of cellular instability, was suggested [31] to explain flame propagation beyond the theoretical 1D flammability limit, that was substantiated in microgravity experiments [32]. In contemporary experiments designed for measuring burning velocity in closed vessels (bombs), the onset of cellularity can be identified using shadowgraphy [11], Schlieren or tomography [18] techniques, and the propagating cellular flames are not considered in the data processing. The occurrence of cellularity limits the range of available raw data from these experiments to flames with relatively low radii and high stretch. This and other

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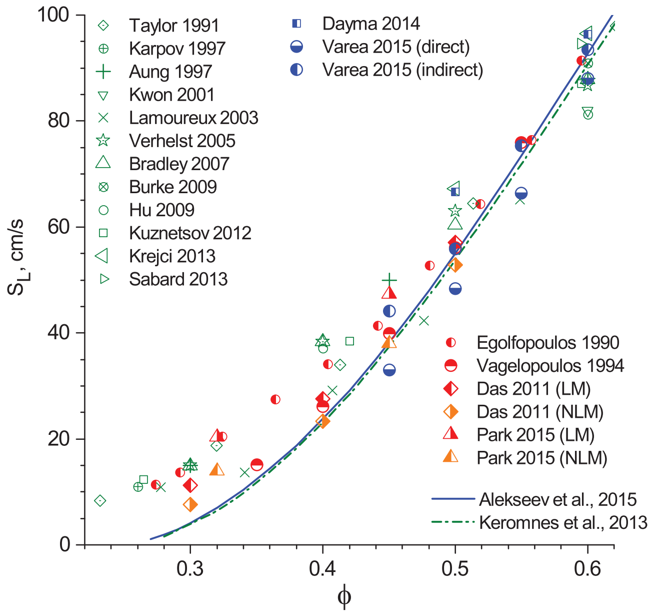

Fig. 1. Laminar burning velocity of H_2 + air flames at standard conditions (T_g = 298 K, p = 1 atm). Symbols: experiments, lines: calculations using models from [24,25]. The source of experimental data: green - Taylor [5], Karpov et al. [6], Aung et al. [7], Kwon and Faeth [8], Lamoureux et al. [9], Verhelst et al. [10], Bradley et al. [11], Burke et al. [12], Hu et al. [13], Kuznetsov et al. [14], Krejci et al. [15], Sabard et al. [16]; blue - Dayma et al. [17], Varea et al. [18]; red - Egolfopoulos and Law [19], Vagelopoulos et al. [20], Das et al. [21], Park et al. [23]; orange – Das et al. [22], Park et al. [23]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

experimental challenges discussed by Egolfopoulos et al. [33] could, at least partly, be responsible for the large data scattering depicted in Fig. 1.

Proper stretch-correction is probably the most important issue affecting the burning velocity derived from counterflow or spherical flames. Direct comparison of the measurements performed in counterflow configuration by Das et al. and processed using linear [21] or non-linear model [22] shows that non-linear extrapolation yields S_L about 32% lower when equivalence ratio, ϕ, equals 0.3. At $\phi = 0.4$, however, these models produced results closer to each other and to the measurements of Vagelopoulos et al. [20], see Fig. 1. In the most recent study of H_2 + air flames using counterflow technique, Park et al. [23] demonstrated that non-linear extrapolation lowers the S_L values significantly in lean flames with $\phi = 0.32$ and 0.45 as compared to the linear model.

The situation in analysis of spherical flame experiments is even more upsetting. As was shown in the recent study of Wu et al. [34], all existing methods for stretch correction in the spherical flame configuration, which was employed to get the vast majority of data in H_2 + air flames, overestimate the laminar burning velocity, and for the classical linear model [26] the difference can reach up to 60%. Other effects related to highly diffusive nature of H_2 and applicability of the calculated density ratio between cold reactants and products to convert flame displacement speed into burning velocity were addressed by Varea et al. [18]. The authors used the technique for direct measurement of the local instantaneous unburned gas velocity [35,36] (denoted "direct" in Fig. 1) and compared the results obtained to S_L determined with a common approach by assuming jump conditions across the flame (denoted "indirect" in Fig. 1). It was clearly demonstrated that these two methods lead to different values of S_L, with the discrepancy increasing when equivalence ratio decreases, even though the numerical simulations predict similar values for the two formulations.

To avoid the impact of stretch correction, independent measurements of the laminar burning velocity of H_2 + air mixtures using flat non-stretched flames could be most useful. This was the first goal of the present study, since no measurements with hydrogen burning in air and stabilized on the heat flux burner were performed so far. There are, however, several investigations of S_L in diluted H_2 flames. The first experiments in $H_2 + O_2 + N_2$ mixtures were done by Hermanns et al. [37]. The authors compared the data obtained in flat flames to the earlier counterflow measurements of Egolfopoulos and Law [19]. The amount of O_2 in the oxidizer, $O_2 + N_2$, was from 7 to 11%, and the equivalence ratio was varied in the range $\phi = 0.65$–3.1. Ratna Kishore et al. [38] also used the heat flux method and studied the effect of dilution by Ar, N_2 and CO_2 on $H_2 + O_2$ flame propagation. They found that lean and stoichiometric CO_2-diluted flames (65% in the total mixture) become cellular, whereas Ar and N_2-diluted flames remained flat. For N_2, the dilution ratios were similar to [37] and direct comparison to 7.7% O_2 flames of [37] showed good agreement in S_L. Voss et al. [39] studied diluted H_2 flames, adding N_2 to the fuel and mixing it with air. If expressed the same way as by Hermanns et al. [37], the studied range covered O_2 content from 6 to 17.7%, and ϕ was varied from 0.45 to 1.5. In the absence of stretch, hydrogen flames stabilized on the heat flux burner are prone to cell formation, especially at higher pressures. Goswami et al. [40] measured burning velocities of syngas and hydrogen up to 10 atm, and had to use O_2 + He oxidizer mixture to suppress cellularity in lean flames. Yu [41] showed numerically that for a typical burner geometry and operating conditions, lean ($\phi = 0.56$) $H_2 + O_2 + N_2$ flame with 18% O_2 in the oxidizer is subject to thermal-diffusive instability. Yu et al. [42] further concluded that in order to increase the stability and obtain an adiabatic flame on a heat flux burner, the flame has to be brought closer to the burner surface below what was called "a critical standoff distance". For a mixture of given composition, this can be achieved by decreasing the unburned gas velocity and moving to sub-adiabatic conditions. In the present work, special attention was paid to the cell appearance in the premixed hydrogen flames due to its possible effect on S_L.

In addition to the spread in S_L, observed in Fig. 1, another serious issue related to lean H_2 + air flames was identified in our recent study [25]. It concerns the temperature dependence of the laminar burning velocity, commonly evaluated in the form of an empirical power law:

$$S_L = S_L^0 (T_g/T_0)^{\alpha}, \qquad (1)$$

where S_L^0 is the burning velocity at a reference temperature T_0, and α is the power exponent. It was found that for very lean H_2 + air mixtures ($\phi < 0.5$), all available data obtained in spherical vessels fail to confirm the rise of the temperature exponent α anticipated close to the flammability limit and predicted by kinetic modeling. Only power exponent α extracted from the measurements of Das et al. [21,22], a single counterflow study performed at elevated temperature, followed the modelling trend, however, with rather high uncertainty, since this study [21] was not aimed at revealing the temperature dependence due to very narrow temperature range (25 K). Thus the second goal of the present work was to study the temperature dependence of S_L in lean H_2 + air flames with the aim to resolve the discrepancy observed in [25], and in addition, to identify whether the temperature exponent α can serve as a criterion of reliability and consistency of the burning velocity measurements.

2. Experimental details

The laminar burning velocity of lean H_2 + air mixtures and its temperature dependence was determined with the heat flux method covering the equivalence ratios of $\phi = 0.375$–0.5 and unburned gas temperatures T_g = 278–358 K. The latter range is conditioned by the use of water as a temperature controlling agent. The range of