Experimental Study of Laminar Burning Velocities of Ammonia/Hydrogen/Air Mixtures at Elevated Temperatures

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INTRODUCTION

As the concerns regarding climate change are gaining high awareness in our societies, a majority of governments agreed on common objectives to try mitigating this phenomenon [1]. These imply an increasing share of fossil-free Renewable Energy Sources (RES) in the global energy mix, reaching up to 65% of the total final energy consumption by the middle of this century in some ambitious scenarii [2]. This transition will rely on diverse, mostly intermittent RES, such as wind or solar PV, and thus require flexibility and grid-balancing strategies.
An important feature will be the ability to efficiently store and transport renewable energy, generally produced primarily as electricity, from the (space-time) point of production to the point of use, where it should be delivered in the desired form. Power-to-X strategies are promising options, where X represents any hydrogen-containing chemical in gaseous, liquid or any other form. Those new energy carriers present a high energy density, are stable in time (seasonal or long-term storage), can be transported over long distances and their production can be carbon-neutral. In spite of being already recognized as an energy carrier, molecular hydrogen (H_2) presents major drawbacks caused by its high volatility and flammability, including the need of a tailored infrastructure and the associated cost and safety issues.
Ammonia (NH_3) has received recent interest as an alternative energy carrier [3]. Its relatively high energy density, with a lower heating value of 18.6 MJ/kg for a density of 0.73 kg/m^3 (300 K, 0.1 MPa), and its carbon-free nature make it a promising competitor to hydrogen and carbon-based energy carriers. Moreover, ammonia has the advantage of being stored in liquid form under 10 bar at standard temperature, and already transported and stored safely at industrial scale. Furthermore, its production from renewable electricity, water and air using electrolysis and air separation is currently subject to several research efforts [4]. Last, ammonia combustion in gas turbines or internal combustion engines could constitute a meaningful way to retrieve the stored energy while minimizing the need for new resources or breakthrough technologies.
Following these considerations, several studies on NH_3 combustion have been initiated [5]. One main drawback of NH_3 as a fuel is its very low Laminar Burning Velocity (LBV), s_u^0, which is one order of magnitude smaller than the one of methane in atmospheric conditions [6–12]. Thus, several experimental studies have considered enhancing the combustion by blending NH_3 with H_2, leading to a significant increase of the LBV [13–16]. Based on experimental data and the detailed reaction mechanism of Mathieu et Petersen [17], Goldmann and Dinkelacker [18] recently proposed correlations for the LBV of NH_3/H_2/air mixtures. However, most of the previous experimental data were measured under standard atmospheric conditions. Therefore, there is a lack of LBV measurement data at higher temperature and pressure, which would allow for improvement of both the reaction mechanisms and the LBV correlations for NH_3/H_2/air combustion. So, the objective of the present study is to provide new experimental LBV data of NH_3/H_2/air spherically expanding flames at several elevated temperatures as a function of the equivalence ratio. The previous correlations [18] are tested in regard of the new data.

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EXPERIMENTAL SET-UP

Experiments are carried out in a spherical stainless steel combustion vessel with an inner diameter of 200 mm, heated with incorporated heating coils. Once the desired temperature up to 473 K is reached, a vacuum pump empties the vessel and the preheated reactants are then introduced in gaseous form thanks to Brooks 5850S mass flowmeters, while being stirred by a fan. After the intake, a quiescence phase of 15s is imposed and a discharge energy of 95 mJ is delivered to ignite the mixture at the center. For a more detailed description of the experimental set-up, please refer to [19]. In some cases, the mixture fails to ignite, in others buoyancy or cellular instability phenomena alter the flame propagation too much to extract meaningful data.
In the cases with successful flame propagation, Schlieren images of the flame are recorded with adjusted acquisition rate up to 12 000 fps, in order to maximize the number of usable images. The flame radii used for the analysis are kept between 6.5 and 25 mm in order to eliminate the spark ignition and wall-pressure effects. The flame propagation speed is then extracted and extrapolated to zero stretch using a non-linear relation by Kelley and Law [20]. The laminar burning velocity is finally calculated by multiplication with the burnt-to-unburnt density ratio obtained from equilibrium computations.
The investigated mixtures range from x_{H_2} = 0 to 60 vol.% in the NH_3/H_2 gas blend and from 0.8 to 1.4 for the global fuel/air equivalence ratio. Unburnt gas temperatures T_u of 298, 323, 423 and 473 K are tested at P_u = 1 bar.

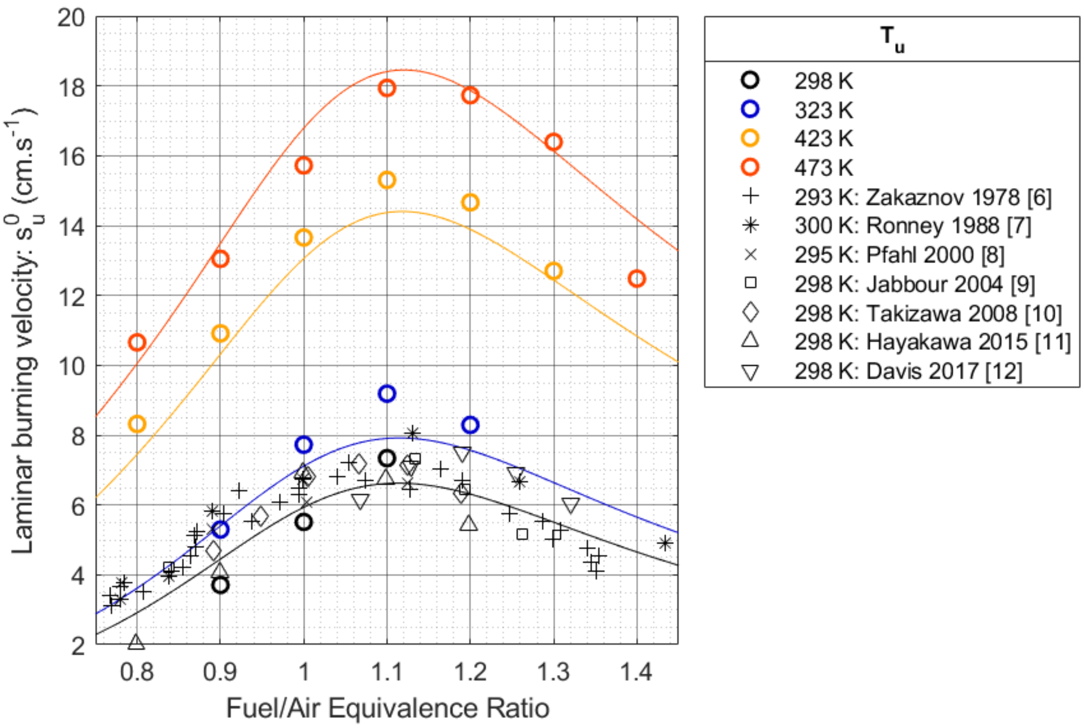

Figure 1: Laminar burning velocities of NH_3/air mixtures as a function of initial ambient temperatures at 1 bar. Circles: present experimental data. Other symbols: experimental data from the literature at Pu ≈ 1 bar and Tu ≈ 298 K. Lines: estimated data by using the NH_3/air correlation as proposed by Goldmann and Dinkelacker [18].

RESULTS AND DISCUSSION

The first part of a new laminar burning velocity dataset is introduced here. Further refinement and extension are intended. Figure 1 shows both experimental LBV measurements and estimated values by using the correlation introduced in [18] for ammonia/air mixtures at 1 bar and various temperatures. The present results agree well with the literature data around 298 K and with the NH_3/air LBV correlation. But by considering each dataset, some discrepancies remain important: it should be mentioned here that such measurements for pure ammonia as fuel are difficult due to the narrow flammability range and the high sensitivity to buoyancy effects induced by the low LBV of the mixtures. As expected, a significant increase of the LBV is observed with temperature increase. A maximum value around equivalence ratio 1.1 is confirmed.

Present experimental and estimated data from [18] are presented in Figure 2 for NH_3/H_2/air mixtures at T_u = 323 K and P_u = 1 bar. The LBVs of the investigated mixtures exhibit similar features over the whole range of tested temperatures and are therefore not plotted here for readability. The LBV increases significantly with the temperature for all mixtures.

Hydrogen addition leads to higher LBVs for all equivalence ratios in a non-linear fashion. It needs around 50 vol.% H_2 to reach LBV values in the same order than methane ones as noted in [15], thus indicating promising potential of NH_3/H_2 blends as a fuel. The correlation in [18] yields very good predictions of the present data for hydrogen-lean mixtures, despite a slight underestimation for fuel-rich blends with the highest ammonia contents. However, a noticeable overestimation is consistently produced by the correlation in comparison with present data for x_{H_2}= 60 vol.%. This reproduces a mismatch observed at Tu=298 K with the data of Li et al. [14], since that experimental data (Bunsen burner method) were used by [18] to fit the correlation. Generally, experimental LBV data from the literature shows an important scatter at high hydrogen ratios [13–16] (not plotted here). Although the reasons for this scatter are not well understood yet, one could question the significance of fitting the correlation on scarce experimental data, especially in the hydrogen-dominated range. The correlation accuracy at hydrogen ratios higher than 60 vol.% is consequently reduced.

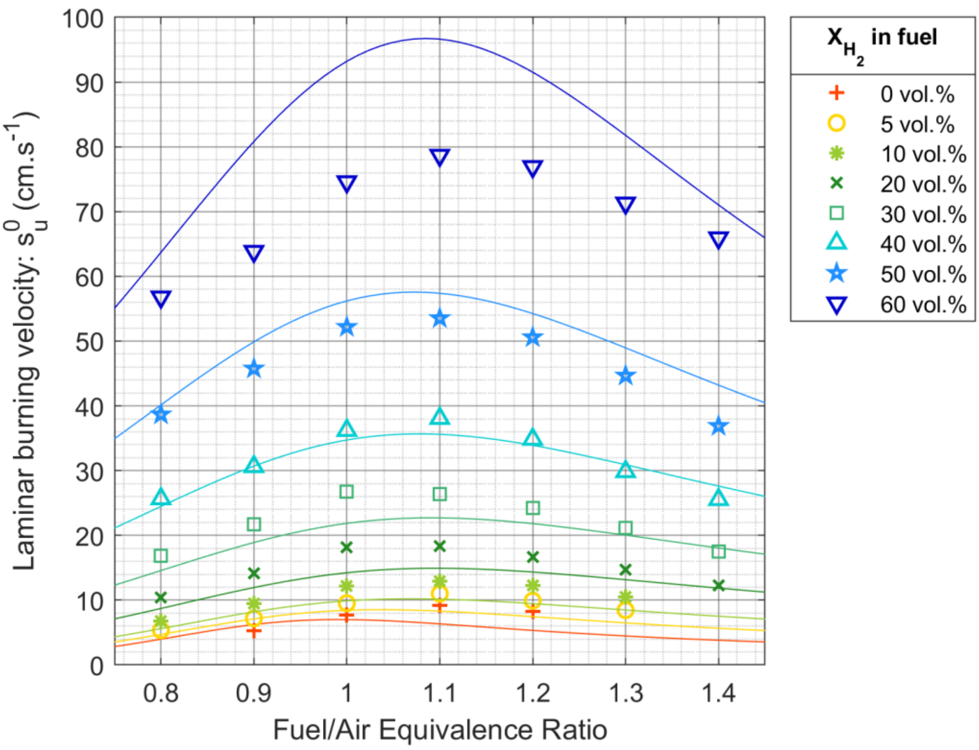

Figure 2: Laminar burning velocities of NH_3/H_2/air mixtures at T_u=323 K and P_u=1 bar. Symbols: present experimental study. Lines: estimated data by using the NH_3/H_2/air correlation as proposed by Goldmann and Dinkelacker [18].

CONCLUSION

A new experimental dataset of NH_3/air and NH_3/H_2/air Laminar Burning Velocities at various temperatures is presented. It confirms the potential of blending NH_3 and H_2 in fuels. A good global agreement is found with other experimental data and a LBV correlation from the literature. However, improvements of this correlation are required especially in the case of high hydrogen contents and will depend on additional and reliable experimental data entries. This will be the focus of the following work, along with LBV measurements at higher pressures.

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