Effect of pressure on the high-temperature flame propagation of ammonia/hydrogen blends

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1 Introduction

Characterization of the combustion behavior of potential fuels of the future is of utmost importance for the design of novel, clean, and efficient energy systems. Among the candidates to replace fossil fuels, carbon-free molecules such as hydrogen and ammonia have gained increased attention, with research efforts being made globally towards their prompt implementation in practical combustion devices. As there are several drawbacks in the direct utilization of either of these alternative fuels, their blends are deemed promising for exhibiting good reactivity and stable flame propagation [1].

High-quality experimental data is necessary for developing and optimizing chemical kinetic models that predict the oxidative characteristics of ammonia/hydrogen mixtures at conditions that emulate those found in practical combustion devices. Laminar flame speed, a thermochemical property dependent on pressure, temperature, and chemical composition, is frequently employed as a global target to validate kinetic models and is conventionally measured using setups such as tube burners [2,3] or constant-volume chambers [4–14]. Given the challenges posed by externally heating these devices and the timescale of this process being comparable or larger than the chemical timescales (i.e. auto-ignition delay time of the combustible mixture), experiments are rarely performed at elevated temperatures and pressures. Using a shock tube as an impulse-heating reactor coupled with non-intrusive laser-induced spark ignition has been proven as a successful methodology for obtaining challenging data at extreme conditions. The shock tube flame speed method, first introduced by Ferris et al. [15], has been employed for the measurements of atmospheric-pressure, high-temperature flame speeds of carbon-containing fuels such as methane [16–18], ethanol [19], iso-octane [19], and propane [20], and carbon-free fuels including neat ammonia [21] and ammonia/hydrogen blends [22]. Recent works have explored extending the capabilities of this methodology to higher pressures, with methanol flame speeds being measured at up to 2.6 atm [23].

In this work, ultra-lean, lean and stoichiometric flames of ammonia/hydrogen mixtures with hydrogen fuel fractions varying from 0.2 to 0.6 were studied in the temperature range of 680-780 K at pressures up to 5 atm. Here, fuel fraction is defined as the ratio of hydrogen concentration to the total concentration of fuel. Some of the most recent and widely used chemical kinetic models available in the literature were employed to simulate flame speeds at the conditions of interest, and their predictions are compared against new experimental measurements.

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2 Experimental methods

Laminar flame speed measurements of ultra-lean (ϕ = 0.5), lean (ϕ = 0.8) and stoichiometric (ϕ = 1) mixtures of ammonia/hydrogen diluted in “airgon” (79% Ar, 21% O_2) were performed in the Imaging Shock Tube (IST) facility at Stanford University. Flame propagation was started by a laser-induced spark ignition system consisting of a Nd:YAG pulsed laser emitting at 532 nm and focusing optics. Beam energy was modulated using a variable power attenuator and the flame ignition was set to occur 0.45 ms after the reflected shock wave’s passage. Tracking of the outwardly expanding flames was performed using a Z-type schlieren imaging setup with a Phantom v2012 high-speed camera and two off-axis parabolic mirrors [24]. Further information about the ignition and imaging systems, and specifications of the optical section of the IST facility utilized in this work can be found in [20].

Flame speeds were extracted by processing the obtained images using an area-averaged linear-curvature model [25]. The total experimental uncertainty of the unburned-gas flame speeds (δS_u^0), or laminar flame speed, was calculated considering the contributions from the uncertainty on the temperature behind the reflected shock and the errors related to both linear regression and the use of a linear curvature model.

3 Kinetic modeling

Simulated laminar flame speeds were computed using the 1-D freely-propagating-flame model implemented in ChemkinPro using multicomponent transport properties and including Soret effect. Gradient and curvature parameters were chosen to be 0.02 and 0.1, respectively, to guarantee a minimum of 500 grid points during the computation. The kinetic models by Okafor et al. [26], Zhou et al. [6], Chen et al. [27] and Jian et al. [28] were selected as representative mechanisms from literature.

Model-predicted unburned-gas flame speeds (S_u^0) of ultra-lean mixtures (ϕ = 0.5) with hydrogen fuel fractions, x_{H2} = 0, 0.2, 0.4, and 0.6 at P = 3 and 5 atm are presented in Fig. 1. For all models and compositions, computed flame speeds increase monotonically with temperature while being inversely proportional to pressure. Discrepancies between the models are observed at all conditions, with larger deviation observed at higher temperatures and hydrogen fuel fractions. Remarkably, the model by Okafor et al., found to be a good predictor of neat ammonia high temperature flame speeds in a previous work [21], consistently showcases a significantly lower value compared to the rest of the models. These observations confirm the existence of high model uncertainty at conditions of high temperature and elevated pressure, and thus, experimental and modeling efforts are still necessary for the improvement of ammonia/hydrogen oxidation models.

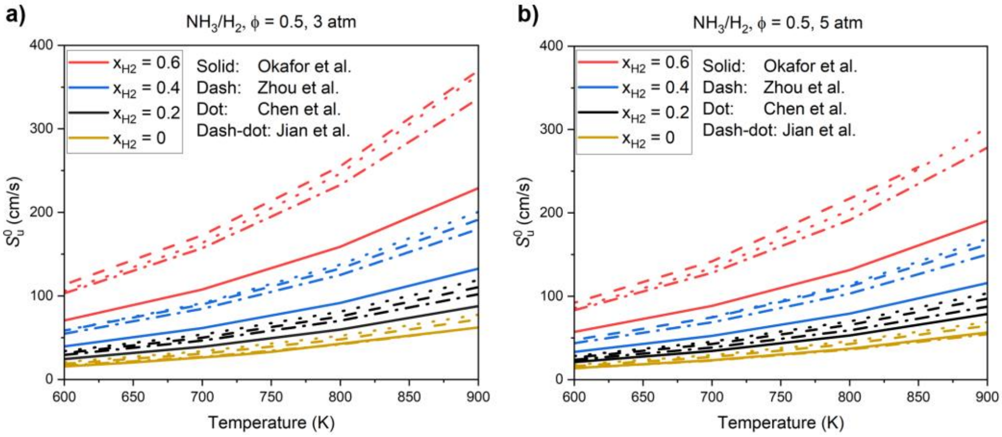

Figure 1: Simulated unburned-gas flame speeds of ultra-lean (ϕ = 0.5) mixtures of ammonia/hydrogen blends, with x_{H2} = 0, 0.2, 0.4, 0.6 at a) P = 3 atm and b) P = 5 atm. Computations were performed using the models by Okafor et al. [26], Zhou et al.[6], Chen et al. [27] and Jian et al. [28]

4 Results

Figure 2 includes the high-temperature measurements obtained employing the shock tube flame speed method at P = 3, 5 atm for ultra-lean, lean and stoichiometric blends of ammonia/hydrogen diluted in "airgon". Ignition energy was minimized at each condition to mitigate the impact of the initial ignition kernel on flame morphology and reduce experimental uncertainty. Notably, the energy input necessary for the onset of a stable flame was inversely proportional to pressure, equivalence ratio and the hydrogen fuel fraction. An average uncertainty of ~6.1% was found across all the conditions studied in this work.

Simulations performed using the model by Zhou et al. [6] are also included in Fig. 2 for prediction evaluation. As projected by the model, experimentally determined flame speeds increase monotonically with temperature and the rate of change is observed to be dependent on hydrogen concentration. The measurements at the highest hydrogen fuel fraction, x_{H2} = 0.6, showcase the largest temperature sensitivity. Additionally, at constant temperatures, a non-linear flame speed enhancement can be observed with increasing hydrogen content in stoichiometric cases, as identified in previous atmospheric-pressure measurements [22]. Moreover, the ultra-lean cases portray significantly lower flame speeds than the data obtained at ϕ = 0.8 and 1. Overall, flame speeds simulated using the model by Zhou et al. [6] have good agreement with experimental data at P = 3 atm, with a maximum deviation of 14% found for the ϕ = 0.5 and x_{H2} = 0.6 case. Discrepancies of 20% and 19% are observed between simulations and nominal experimental measurements at P = 5 atm and x_{H2} = 0.6 for the ultra-lean and lean cases, respectively. These findings emphasize the importance of providing low-uncertainty data which facilitate accurate modeling near flammability limits. As several data points from this work are underpredicted by the model even when considering experimental uncertainty bounds, these data should prove useful for constraining and optimizing kinetic models.

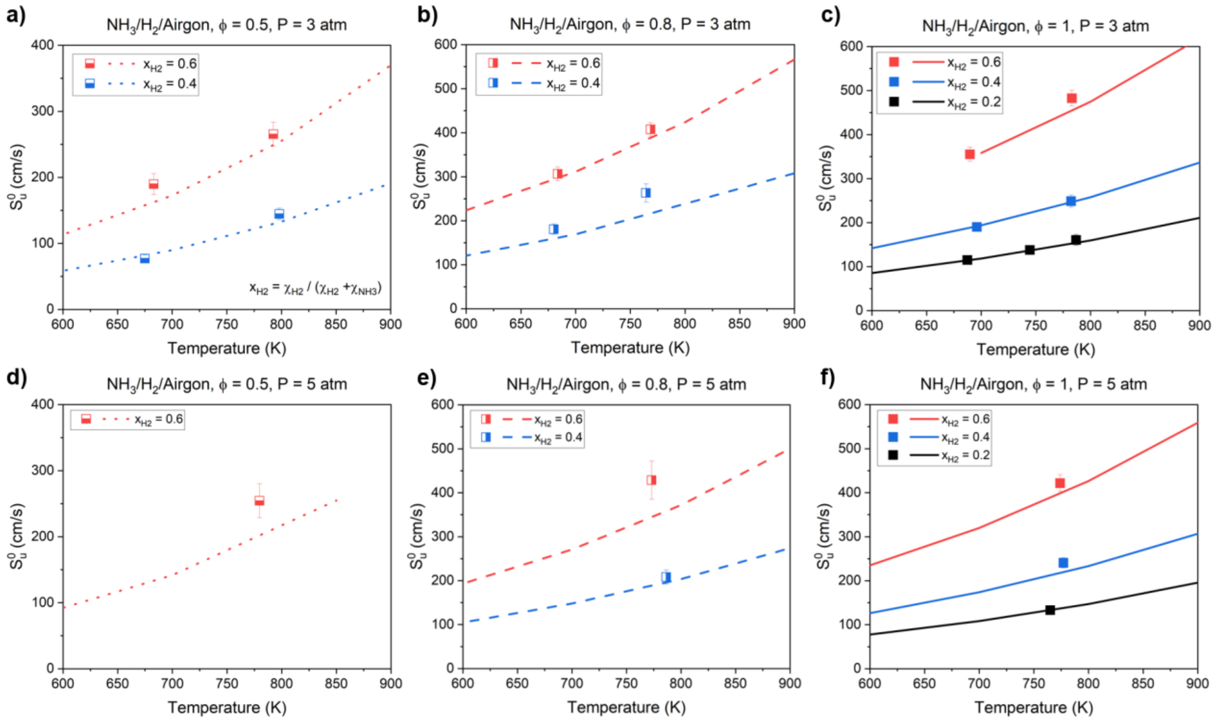

Figure 2: Laminar flame speed measurements of ammonia/hydrogen mixtures at P = 3 atm with a) ϕ = 0.5, b) ϕ = 0.8 and c) ϕ = 0.1; and at P = 5 atm with d) ϕ = 0.5, e) ϕ = 0.8 and f) ϕ = 1. Lines are simulations computed using the model by Zhou et al. [6].

5 Conclusions

This work presented the first study on the propagation of outwardly expanding ammonia/hydrogen flames at temperatures above 650 K and elevated pressures up to 5 atm. The shock tube flame speed method was employed in the measurement of flame speeds of ultra-lean, lean, and stoichiometric mixtures containing ammonia fuel blends with up to 60% hydrogen content. A monotonic relationship was observed between flame speed and temperature, while increasing pressure promoted slower flame propagation. Ultra-lean flames ($\phi = 0.5$) showcased significantly lower flame speeds when compared to mixtures closer to stoichiometric composition. Simulated flame speeds were computed using four different chemical kinetic models and notable differences were identified in their predictions. The model by Zhou et al. [6] provides the best agreement with experimental data at all conditions studied. Both experimental and computational research efforts are still necessary for further uncertainty reduction and optimization of kinetic models. Future work will focus on the identification and quantification of the most impactful reactions in flame speed computation at various blend compositions through sensitivity analysis.

6 Acknowledgements

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