

# Laminar Burning Velocity Measurement of NH<sub>3</sub>/H<sub>2</sub>/air Mixtures at Elevated Temperatures

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## 1 Introduction

The world has reached a point where immediate measures to limit carbon emissions are imperative due to the alarming increase in global warming and the catastrophic change in the climate. Having exhausted a substantial amount of fossil fuels, the focus shifted towards alternative, clean fuels over the past few decades. Hydrogen has been deemed the most efficient alternative fuel to address the current global energy crisis, owing to its high energy density (i.e.,  $120\text{MJ/kg}$ ) [1] on a mass basis compared to other conventional fuels like propane or gasoline (i.e.,  $44\text{MJ/kg}$ ), absence of a carbon footprint, and abundance.

Despite its high gravimetric energy density, hydrogen has a very low energy content by volume [2]. Liquid hydrogen has a volumetric energy density of  $8\text{MJ/L}$ , which is significantly lower than gasoline's volumetric energy density of  $32\text{MJ/L}$ . Pure hydrogen as a fuel is debatable since onboard vehicular hydrogen storage systems need extremely high pressures and low temperatures to store hydrogen compactly. Furthermore, the high reactivity and broad flammability limits of hydrogen present a major storage challenge and, thus, a transportation difficulty. Numerous hydrogen carriers have been investigated to address this issue, with ammonia being one of the most promising fuel because it is carbon-free and contains 17.8% [3] hydrogen by mass. Additionally, as ammonia has been used in the fertiliser industry for more than 100 years, it has a well-established transportation and storage infrastructure, requiring little initial investment when utilized as the primary fuel. Pure ammonia combustion faces a number of difficulties that restrict its usage as a pure fuel, such as low laminar burning velocity (LBV) and  $\text{NO}_x$  emissions at high temperatures. Numerous researchers have made various attempts to improve the burning velocity of ammonia. According to Okafor et al. [4], a 50 – 50 mix of  $\text{NH}_3 / \text{CH}_4$  results in an increase in laminar burning velocity of about 114% when compared to pure ammonia fuel, which has a burning velocity of  $7\text{cm/s}$  at stoichiometric condition. In 2023, Berwal et al. [5] investigated laminar burning velocity of premixed  $\text{NH}_3/\text{H}_2/\text{CH}_4/\text{air}$  mixture and noted that blending ammonia with hydrogen and methane improved fuel performance in addition to producing low carbon emissions.

Blends of hydrogen and ammonia have also been investigated in an effort to totally eliminate carbon emissions in gas turbines at engine relevant conditions [6] [7]. According to Rocha et al. [8], the addition of hydrogen prompts the flame speed to increase exponentially, but  $\text{NO}_x$  emissions were also seen to increase due to the production of  $\text{OH}$  and  $\text{O}$  radicals that result from the addition of hydrogen. Osipova et al. [9] performed ammonia oxidation experiments using various hydrogen fractions and found

laminar burning velocity for  $NH_3/H_2$  in 50/50 volume fraction under stoichiometric condition at standard temperature and pressure to be  $24.7\text{cm/s}$ , which is comparable to the commonly used fuels. By varying the hydrogen fraction from 0 to 30% in the  $NH_3/H_2$  fuel blend, Wang et al. [10] examined the engine performance. They found that adding just 5% hydrogen to the mixture significantly improved the ignition performance of  $NH_3$ . Laminar burning velocity of the mixture rose as hydrogen was added, culminating at about  $\phi = 1.1$ , and the trend was steeper for large hydrogen fractions. Additionally, it was found that flame temperature rose with the addition of hydrogen, peaking at about  $2200\text{K}$  for 40% hydrogen [10].

Mei et al. [11] has suggested that partial cracking of ammonia into hydrogen and nitrogen is a feasible option to enhance the performance of ammonia based fuel blends. To imitate the partial cracking of ammonia,  $H_2/N_2$  were used in 3 : 1 ratio. LBV was found to peak at equivalence ratio 1.1 with a value of  $38.1\text{cm/s}$ , which is close to the LBV of a  $CH_4/air$  mixture, at standard temperature and pressure. Because nitrogen causes lower flame temperature and have a detrimental impact on laminar flame propagation, LBV of the employed blend were found to be lower than the burning velocities of pure  $NH_3/H_2$  mixture. In the similar studies conducted by Ji et al. [12], partial dissociation of ammonia was studied in which nitrogen is added to simulate the cracking of ammonia in ratio  $H_2 : N_2 :: 1 : 1/3$ . At  $303\text{K}$  and  $0.1\text{MPa}$  pressure, the laminar burning velocity of completely dissociated ammonia was found to be  $228\text{cm/s}$ . LBV was found to be higher at ammonia dissociation degree of 0.6 than it was for  $CH_4/air$  under the same initial conditions. The Lewis number changes as a result of the change in the mixture's diffusion characteristics due to the increased hydrogen content in the fuel blend with increment in  $NH_3$  cracking. In opposition to lean fuel conditions, laminar burning velocity was seen to significantly increase under rich fuel conditions because Lewis number was more than unity.

A number of mechanisms have been developed to investigate the ammonia/hydrogen mixture's combustion properties. Shrestha et al. [13] developed a mechanism for oxidation of ammonia and ammonia/hydrogen mixture and measured the laminar burning velocity of oxy ammonia when oxygen content in the oxidizer was increased from 21% to 30% and compared it with the LBV of  $NH_3/H_2$  mixture when 30% hydrogen by volume was added to ammonia. According to the study, adding 30% hydrogen increased the LBV of  $NH_3$  by a factor of 2.9 whereas adding 9% more oxygen to the oxidizer increased the LBV of  $NH_3$  by a factor of 2.8, demonstrating that both values were comparable. Otomo et al. [14] proposed an improved mechanism to simulate  $NH_3/H_2/air$  combustion in practical combustor application. Model predicted that under rich fuel conditions if  $NH_3/H_2$  ratio is regulated optimally in accordance with the equivalence ratio, low  $NO$  and  $NH_3$  concentration can be found in flue gases while high  $H_2$  content can be detected [14].

Present study deals with the measurement of laminar burning velocity of  $NH_3/H_2$  mixture using externally heated diverging channel technique for varying  $H_2$  concentration up to 40% by volume in fuel at elevated temperatures up to  $750\text{K}$ . The current experimental data is also compared with kinetic models by Otomo [14] and Shrestha [13] and sensitivity analysis is conducted for better understanding of the major participating reactions and species.

## 2 Experimental Set-up

Experiments were conducted in a mesoscale diverging channel to test the laminar burning velocity of  $NH_3/H_2$  mixture in a premixed configuration [15]. The planar flames produced by the homogeneous flow field and high aspect ratio are used to determine the laminar burning velocity in a variety of operating conditions. A quartz diverging channel with  $25\text{mm} \times 2\text{mm}$  intake dimensions, a  $15^\circ$  divergence angle, and a 12.5 starting aspect ratio was used to carry out the experiments. In order to prevent flame

flashback, the divergence creates a negative velocity gradient in the flow direction of the premixed mixture. An external heater is used to heat the channel at the open end of the premixed mixture flow in order to create a positive temperature gradient. As the aspect ratio of the channel grows, the flame velocity drops, but this is balanced by the positive temperature gradient in the flow direction, creating a stable planar flame. The flame becomes practically adiabatic because the external heater compensates for heat losses from the flame. To produce a premixed mixture with a particular equivalence ratio, a precise volumetric flow rate of fuel and air is supplied using mass flow controllers. This mixture is permitted to pass through the diverging channel and ignited at open end. Following ignition, the flame starts to travel towards the unburnt gaseous mixture and stabilizes inside the channel when the flow velocity approaches the local flame propagation velocity. K-type thermocouple is used to measure the temperature of unburned mixture. Laminar burning velocity is calculated using mass continuity equation:

$$S_L = U_{inlet} \left( \frac{T_f}{T_{u0}} \right) \left( \frac{A_{inlet}}{A_f} \right) \quad (1)$$

Where  $U_{inlet}$  and  $T_{u0}$  is inlet velocity and inlet temperature of the fuel mixture respectively.  $A_{inlet}$ ,  $A_f$  and  $T_f$  is inlet area, area at flame location and temperature at flame location respectively.

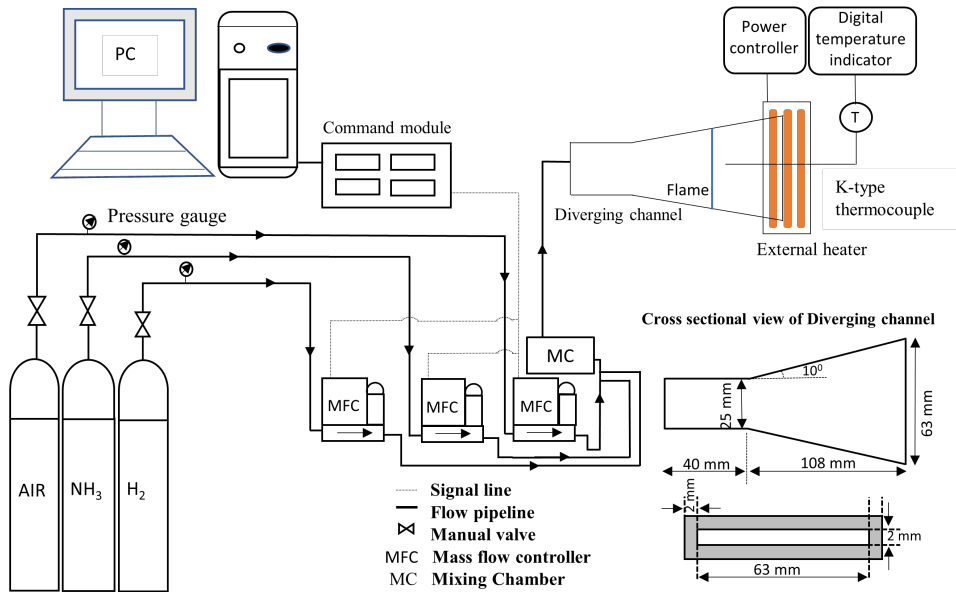


Figure 1: Schematic of the experimental set-up

### 3 Results and Discussions

The laminar burning velocities of  $NH_3/H_2/air$  flames were measured as shown in Figure 2 for equivalence ratios 0.9, 1, and 1.1 at elevated temperatures up to  $750K$ . Pressure and temperature at the inlet were kept at  $1atm$  and  $300K$  respectively. Mole fraction of hydrogen was varied up to  $X_{H_2} = 0.4$  and comparisons between the LBVs of  $X_{H_2} = 0.3$  and  $X_{H_2} = 0.4$  are shown to understand the variation in burning velocities. Experiments for  $X_{H_2} > 0.4$  were not conducted due to the occurrence of cellular instabilities [11] with the increment in hydrogen addition as above  $X_{H_2} = 0.4$  mixture. LBV was observed to increase with the increasing temperature ratios at all mixture conditions and the results are plotted in comparison with the results obtained using the mechanism developed by Shrestha et al. [13]

and Otomo et al. [14]. Mechanism developed by Shrestha has shown good consistency with the experimental values with the accuracy of  $\pm 5\%$  for both the fuel compositions. As the hydrogen fraction was increased from  $X_{H_2} = 0.3$  to  $X_{H_2} = 0.4$ , approximately 54% increment was observed at standard inlet condition.

At lean fuel conditions, temperature exponent value was high for  $X_{H_2} = 0.4$  which suggests the higher dependency of the LBVs on the temperature as compared to  $X_{H_2} = 0.3$ . This phenomena reverses for stoichiometric and rich fuel conditions. Current study suggest the viability of using an ammonia reformation system in conjunction with a practical engine to reduce carbon emissions.

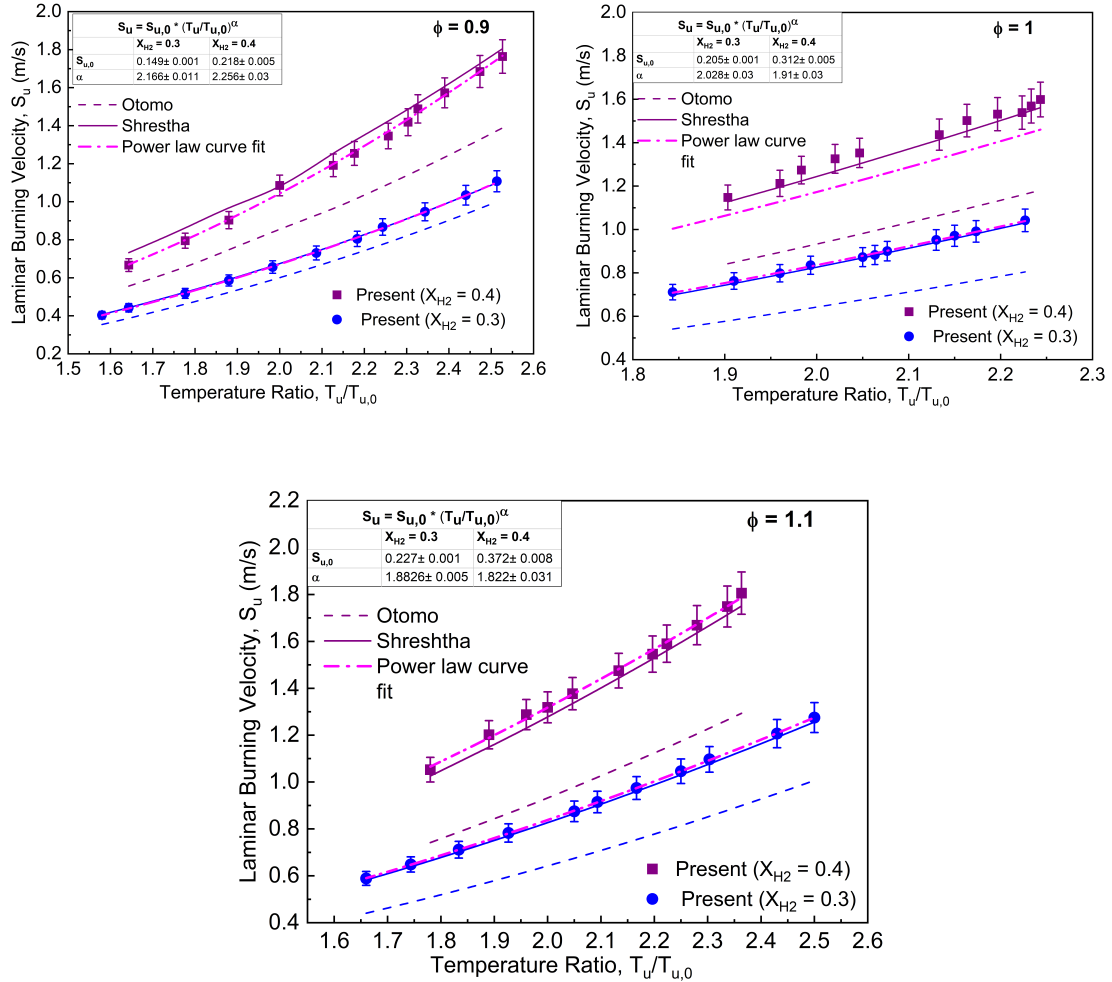


Figure 2: Laminar Burning Velocity vs Temperature ratio at equivalence ratios 0.9, 1 and 1.1 for  $X_{H_2} = 0.3$  and  $X_{H_2} = 0.4$

To comprehend the effects of major participating reactions on the laminar burning velocity of  $NH_3/H_2/air$  mixture, sensitivity analysis has been performed using the mechanism given by Shrestha et al. [13]. The normalized sensitivity of the laminar burning velocity ( $S_u$ ) with respect to the rate constant ( $k$ ) is defined as:

$$Sensitivity(S_u, k) = \left( \frac{\partial S_u}{\partial k} \right) \left( \frac{k}{S_u} \right) \quad (2)$$

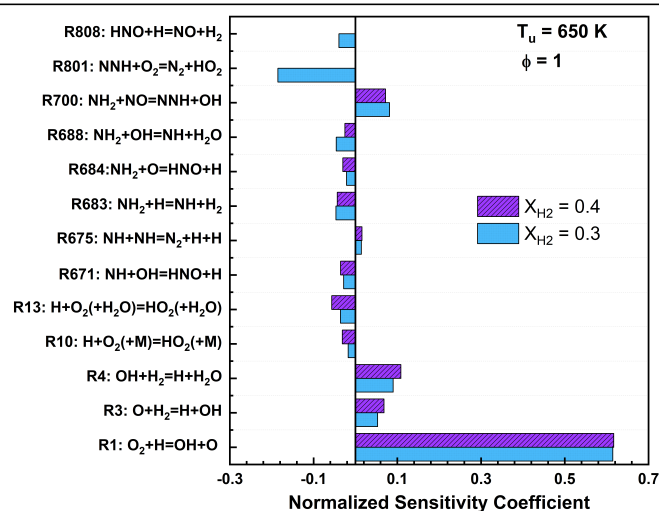


Figure 3: Normalized sensitivity coefficient variation of  $NH_3/H_2/air$  at 650K for  $X_{H_2} = 0.3$ ,  $X_{H_2} = 0.4$  and  $\phi = 1$

It can be deduced from the analysis that the major burning velocity enhancing reactions are  $R1$  ( $O_2 + H = OH + O$ ) and  $R4$  ( $O + H_2 = H + OH$ ). For  $X_{H_2} = 0.4$ , H and OH radical pool is high by 2% for  $R1$  and by 40% for  $R3$  which can be deemed as the reason for high burning velocities at high hydrogen fractions.  $R801$  ( $NNH + O_2 = N_2 + HO_2$ ) and  $R808$  ( $HNO + H = NO + H_2$ ) are major inhibiting reactions in case of  $X_{H_2} = 0.3$  and are missing for the case of  $X_{H_2} = 0.4$ . Hence, lower LBV for lower molar fractions of hydrogen. In case of  $R808$  and  $R801$ , there is a formation of  $NO$  and  $N_2$  by  $HNO$  and  $NNH$  radicals respectively for  $X_{H_2} = 0.4$ . Radical  $HNO$  utilizes  $H$  radicals and act as a competing species which in turn reduces burning velocity for low hydrogen fractions in  $NH_3$ . When hydrogen fraction was increased from  $X_{H_2} = 0.3$  to  $X_{H_2} = 0.4$ , due to the dominance of hydrogen chemistry,  $H$  and  $O_2$  radicals are readily consumed by  $R1$  and  $R3$  hence diminishing the formation of  $N_2$  and  $NO$  radicals.

## 4 Conclusions

Present study measure the LBV using Externally heated diverging channel method for varied hydrogen fraction in ammonia by volume. The increased amount of hydrogen atoms causes a significant increase in laminar burning velocities with hydrogen substitution, especially under fuel-rich scenarios. Major conclusion from the study are as follows:

- Increment of approximately 52% was observed when hydrogen fraction was increased from  $X_{H_2} = 0.3$  to  $X_{H_2} = 0.4$  at equivalence ratio 1.
- Current experimental data is in good consistency with the model developed by Shrestha et al. [13] while model proposed by Otomo et al. [14] needs improvements
- $HNO$  and  $NNH$  radical are major species responsible for lower burning velocity at low hydrogen fraction in ammonia.
- This study suggests that an ammonia reformation system, which breaks ammonia and produces 30% to 40% hydrogen at the exhaust stream, could be used in conjunction with a practical engine to reduce carbon emissions.

**References**

- [1] Jain, I. P. "Hydrogen the fuel for 21st century." *International journal of hydrogen energy* 34.17 (2009): 7368-7378.
- [2] Zamfirescu, C., and Ibrahim Dincer. "Ammonia as a green fuel and hydrogen source for vehicular applications." *Fuel processing technology* 90.5 (2009): 729-737.
- [3] Mukherjee, Shreya, et al. "Low-temperature ammonia decomposition catalysts for hydrogen generation." *Applied Catalysis B: Environmental* 226 (2018): 162-181.
- [4] Okafor, Ekenechukwu C., et al. "Experimental and numerical study of the laminar burning velocity of CH<sub>4</sub>-NH<sub>3</sub>-air premixed flames." *Combustion and flame* 187 (2018): 185-198.
- [5] Berwal, Pragya, and Sudarshan Kumar. "Laminar burning velocity measurement of CH<sub>4</sub>/H<sub>2</sub>/NH<sub>3</sub>-air premixed flames at high mixture temperatures." *Fuel* 331 (2023): 125809.
- [6] Pugh, Daniel, et al. "An investigation of ammonia primary flame combustor concepts for emissions reduction with OH\*, NH<sub>2</sub>\* and NH\* chemiluminescence at elevated conditions." *Proceedings of the Combustion Institute* 38.4 (2021): 6451-6459.
- [7] Lhuillier, Charles, et al. "Experimental investigation on ammonia combustion behavior in a spark-ignition engine by means of laminar and turbulent expanding flames." *Proceedings of the Combustion Institute* 38.4 (2021): 5859-5868.
- [8] da Rocha, Rodolfo Cavaliere, Mário Costa, and Xue-Song Bai. "Chemical kinetic modelling of ammonia/hydrogen/air ignition, premixed flame propagation and NO emission." *Fuel* 246 (2019): 24-33.
- [9] Osipova, Ksenia N., Oleg P. Korobeinichev, and Andrey G. Shmakov. "Chemical structure and laminar burning velocity of atmospheric pressure premixed ammonia/hydrogen flames." *International Journal of Hydrogen Energy* 46.80 (2021): 39942-39954.
- [10] Wang, Yang, Xiaohu Zhou, and Long Liu. "Theoretical investigation of the combustion performance of ammonia/hydrogen mixtures on a marine diesel engine." *International Journal of Hydrogen Energy* 46.27 (2021): 14805-14812.
- [11] Mei, Bowen, et al. "Enhancement of ammonia combustion with partial fuel cracking strategy: laminar flame propagation and kinetic modeling investigation of NH<sub>3</sub>/H<sub>2</sub>/N<sub>2</sub>/air mixtures up to 10 atm." *Combustion and Flame* 231 (2021): 111472.
- [12] Ji, Changwei, et al. "Experimental and numerical study on premixed partially dissociated ammonia mixtures. Part I: Laminar burning velocity of NH<sub>3</sub>/H<sub>2</sub>/N<sub>2</sub>/air mixtures." *International Journal of Hydrogen Energy* 47.6 (2022): 4171-4184.
- [13] Shrestha, Krishna Prasad, et al. "An experimental and modeling study of ammonia with enriched oxygen content and ammonia/hydrogen laminar flame speed at elevated pressure and temperature." *Proceedings of the Combustion Institute* 38.2 (2021): 2163-2174.
- [14] Otomo, Junichiro, et al. "Chemical kinetic modeling of ammonia oxidation with improved reaction mechanism for ammonia/air and ammonia/hydrogen/air combustion." *International Journal of Hydrogen Energy* 43.5 (2018): 3004-3014.
- [15] Akram, Mohammad, V. Ratna Kishore, and Sudarshan Kumar. "Laminar burning velocity of propane/CO<sub>2</sub>/N<sub>2</sub> -air mixtures at elevated temperatures." *Energy fuels* 26.9 (2012): 5509-5518.