Laminar Burning Velocity Measurement of Ammonia Fuel Blends at Elevated Temperature and Pressures

Pragya Berwal and Sudarshan Kumar
Indian Institute of Technology Bombay, Powai
Mumbai, Maharashtra, India

1 Introduction

Ammonia, a carbon-free chemical, offers to be a clean alternative fuel to widely used hydrocarbon-based transportation fuels secondary to hydrogen. The focal point of Sustainable Development Summit 2015 was to encourage the easy access to clean energy globally by 2030 to mitigate the negative impact of hydrocarbon-based fuels. Currently, both hydrogen (H\textsubscript{2}) and ammonia (NH\textsubscript{3}) are promising carbon-free, green fuels. Low values of boiling point and volumetric energy density (8.4 MJ/l) with wide flammability limits, and high combustion instabilities poses a serious concern in the storage and transportation of H\textsubscript{2} [1]. NH\textsubscript{3} on the other hand has a high boiling point of 240 K with a volumetric energy density of 11.2 MJ/l, and is being commercially used for fertilization, and refrigeration due to the ease in storing and transporting. Hence, making NH\textsubscript{3} a potential candidate as a carbon-free fuel for different practical combustion systems.

Laminar Burning velocity (LBV), a fundamental combustion property is used in the development/validation of a reaction mechanism and determining heat release rate [1]. It also plays a key role in the investigation of flame characteristics, such as flame stabilization, flashback, blow off and flame extinction. For any fuel, commonly used LBV measurement techniques are spherically expanding combustion chamber, heat-flux method, and counterflow stagnation method [1]. With a low LBV (0.07 m/s) of NH\textsubscript{3} at atmospheric conditions [2], various approaches have been suggested such as fuel blending, preheating air, and ammonia cracking. Hayakawa et al. [3] experimentally demonstrated a decrement in the LBV of pure NH\textsubscript{3}/air mixtures as the constant volume combustion chamber pressurized. NH\textsubscript{3} flame propagation was observed to be clearer at high pressures. However, for pressures up to 0.5 MPa, LBV was found to be less than 7 cm/s at 298 K mixture temperature. Furthermore, NO formation/reduction species participated in pure NH\textsubscript{3} flames and reported a decrease in NO mole fraction as the mixture turned richer (\(\phi \geq 1.1\)) at high pressures (up to 0.3 MPa) due to excess NH\textsubscript{3} (i=0,1,2) radicals [4]. In the context of enhancing NH\textsubscript{3} LBV, blending with methane (CH\textsubscript{4}) and H\textsubscript{2} have been conducted separately. Henshaw et al. [5] observed a LBV reduction of 17% - 23% for CH\textsubscript{4}-air mixture (with 4% NH\textsubscript{3} addition) from lean to rich mixture conditions. A non-linear decreasing trend of CH\textsubscript{4} LBV was reported by Okafor et al. [6] with NH\textsubscript{3} addition (up to 50%) and developed a reaction mechanism for CH\textsubscript{4}/NH\textsubscript{3} flames. For CH\textsubscript{4}/NH\textsubscript{3}-air flames, presence of OH radical resulted in the formation of NH\textsubscript{2} molecules with an increase in NOx emissions. Okafor et al. [6] reported high pressure (up to 0.5 MPa) LBV measurements of blends (NH\textsubscript{3}/CH\textsubscript{4}) using constant volume combustion chamber at 298 K mixture temperature. The NH\textsubscript{3} mole fraction ranged between 0 and 0.52 in the CH\textsubscript{4}/NH\textsubscript{3} fuel blend for an equivalence ratio of 0.7-1.3. The obtained unstretched LBV tend to reduce with the rise in both pressure and NH\textsubscript{3} concentration. For the H\textsubscript{2}/NH\textsubscript{3} blends, Lee et al. [8] reported 84% increment in the NH\textsubscript{3} LBV as the H\textsubscript{2} volume fraction increased from 30% to 50% in the spherical flame propagation

Correspondence to: pragyaberwal_93@iitb.ac.in
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method. The reason being high diffusivity of H₂ [9]. Similar behavior was observed in the study of Lhuillier et al. [10] for the H₂ addition (X_{H₂}) up to 60% in the premixed mixture of NH₃/H₂. However, unstable flames were evident for X_{H₂} < 20% and X_{H₂} > 50%. Recently, investigations of Ji et al. [11] and Mei et al. [12] focused on the effect of partial cracking of NH₃ to H₂ and N₂ on the LBV of NH₃-air mixtures. The partial cracking (≥ 40%) of NH₃ contributed in higher LBV of the mixture. Further, high H₂ concentration gave rise to cellular instabilities. In the study of Ichikawa et al. [13] at elevated pressures (up to 0.5 MPa) in the constant volume combustion chamber, the LBV of NH₃/H₂ flame with 40% H₂ (by vol.) was found to be 31.2 cm/s (similar to other hydrocarbons). Interestingly, wrinkled flames were formed at 0.5 MPa with 40% H₂ (by vol.) in fuel-air mixture due to thermal-diffusive instability. Xiao et al. [14] assessed the efficacy of 12 kinetic models using the available experimental data of NH₃/H₂/air mixtures on the basis of flame speed, NOx emissions and ignition delay.

This work investigates the effect of NH₃ concentration (0-30%) on the LBV of NH₃/H₂/CH₄-air mixtures. The measurements are performed using the externally heated diverging channel technique (EHDCM) for temperatures and pressures up to 750 K and 0.3 MPa respectively. The outcomes of this study will be crucial in advancing the expedition in proposing alternate fuel blends of NH₃ with a comparable laminar burning velocity and low carbon emissions compared to methane. For an equivalence ratio (ϕ = 0.7-1.4), current measurements are compared with the detailed kinetic models of Li et al. [15], Shrestha et al. [16], and Okafor et al. [6].

2 Experimental set up

For this high-pressure study, the externally heated diverging-channel method is appropriately adapted to measure LBV inside a cylindrical pressure chamber (40 L) as shown in Fig. 1 [17]. A quartz made diverging channel with the 10° divergence and 50 mm² (25 mm × 2 mm) inlet area is used primarily because of its high heat capacity, low thermal conductivity, and optical access for flame visualization. The top flange of the chamber is carefully designed to have a provision for toughened glass (visual access) along with a pressure relief valve and a pressure gauge. The divergence assists in establishing a negative velocity gradient along the mixture flow direction. An ignition system is stationed near the channel exit for igniting the premixed fuel-air mixture at high-pressure conditions. The high aspect ratio (12.5) at the channel inlet creates a uniform flow field and subsequently stabilizes planar flames (Fig.2) followed with LBV determination at distinct operating conditions. A positive temperature gradient is formed in the direction of mixture flow using an external heater distanced at 2 cm below the quartz channel. This heater also helps achieving adiabaticity. The channel and heater are aligned parallel to each other with an overlap distance of 4 cm. The position of a stabilized flame is fixed by the balance between the negative and positive velocity gradient. The flow rates of fuel and air are monitored using the mass flow controllers (MFC) for different experimental conditions (equivalence ratio and mixture velocity).

Air is used as an oxidizer for the different mixtures of methane, hydrogen, and ammonia for the present measurements. The MFC’s for methane (0-2 slpm), hydrogen (0-0.5 slpm), ammonia (0-0.2 slpm), and air (0-5 slpm) have an accuracy of ±1% of the full-scale flow and are controlled via computer using a command module. A K-type thermocouple is used for temperature measurements. All the LBV measurements were repeated thrice, and the final values have an uncertainty of ±5%. The LBV is calculated using Eq. (1) (obtained from the continuity equation and equation of state):

\[ S_u = U_{inlet} \times \frac{A}{A_f} \times \frac{T_u}{T_{u,o}} \quad (1) \]

Following correlation (Eq. (2)) was obtained between LBV and temperature ratio using the power-law curve fitting to calculate the temperature exponent:

\[ \frac{S_u}{S_{u,o}} = \left( \frac{T_u}{T_{u,o}} \right)^{\alpha} \quad (2) \]
The current measurements are compared with the reaction mechanisms of Li et al. [15], Okafor et al. [6], and Shrestha et al. [16]. The predictions of the kinetic models are evaluated using Chemkin-Pro software tool.

Figure 1: Experimental set up schematics

Figure 2: Stabilized planar flame of NH$_3$/H$_2$/CH$_4$-air mixture under 0.3 MPa at stoichiometric condition

3 Results and Discussion

In this study, laminar burning velocity (LBV) measurements of NH$_3$/H$_2$/CH$_4$-air premixed mixtures are carried out using the externally heated diverging channel method. For an equivalence ratio ($\phi = 0.7$-1.4) at mixture temperature (300 – 750 K) and elevated pressures (0.1-0.3 MPa), the mole fraction of NH$_3$ ($X_{NH3}$) is varied between 0.1 and 0.3 in CH$_4$/H$_2$ (4:1) fuel mixture. The influence of varying experimental conditions on the LBV of the proposed ternary blend have been discussed and compared with pure CH$_4$-air mixtures in the following subsections. The measurements being novel in terms of the blend.
composition, no literature data is available for comparison. Therefore, mechanism predictions of Li et al. [15], Okafor et al. [6], and Shrestha et al. [16] has been used in this investigation.

Figure 3 illustrates the LBV variation of NH₃/H₂/CH₄-air mixtures with X₉₃ = 0.1, 0.3 and pure CH₄ with temperature ratio (T₀/T₀,0) for lean, stoichiometric, and rich conditions. The data is compared for 0.1 and 0.3 MPa. A consistent decrease in the LBV is evident with increase in pressure at all mixture conditions. Interestingly, LBV for the composition with X₉₃ = 0.1 is higher in comparison to CH₄/natural gas irrespective of the pressure. This clearly shows the efficacy of the ternary blend as an alternative for natural gas with lower carbon emissions (~ 25%). Also, LBV increases with the rise in temperature ratio, and the temperature exponent (calculated using Eq. 2.) tends to increase with pressure rise.

Figure 3. Variation of LBV with temperature ratio at (a) φ = 0.8, (b) φ = 1 and (c) φ = 1.2

Figure 4 shows the LBV variation with equivalence ratio at mixture temperatures of 300 K and 750 K for 0.1 and 0.3 MPa. At φ = 1 and 0.3 MPa pressure, LBV decrease by ~33% and ~27% at mixture temperatures of 300 K and 750 K respectively. This highlights the diminishing effect of pressure at higher mixture temperature. LBV of X₆₃ = 0.1 is higher than pure CH₄ at 0.1 MPa and is comparable at higher pressure (0.3 MPa). A good consistency is observed on comparison with the predictions of Okafor [6] and Li [15] kinetic models. Higher LBV predictions are observed for Shrestha [16] reaction mechanisms at stoichiometric and rich conditions.

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Figure 4: Variation of LBV with equivalence ratio at 0.1 and 0.3 MPa for temperatures (a) 300 K and (b) 750 K

4 Conclusion

Fundamental investigations on ammonia fuel blend have been reported to investigate a ternary blend composition with either higher or comparable laminar burning velocity (LBV) with methane. The key findings of this work include:

- A higher LBV of NH$_3$/H$_2$/CH$_4$-air mixture is observed in comparison to pure CH$_4$ at higher mixture temperatures of 500 K.
- At high pressure, the LBV of NH$_3$/H$_2$/CH$_4$-air mixtures (X$_{NH3}$ = 0.1) comply well with pure CH$_4$.
- NH$_3$/H$_2$/CH$_4$ ternary blend can be proposed as a potential alternative fuel to natural gas and extensive research can be further performed at engine relevant conditions to mitigate NOx emissions.

References

modelling of the laminar burning velocity of methane-ammonia-air flames at high pressures using a reduced reaction mechanism. Combust Flame (2019);204:162–75.


