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INVESTIGATION OF THE LAMINAR BURNING VELOCITY AND EXHAUST CHARACTERISTICS OF METHANE-AMMONIA-HYDROGEN TERNARY BLENDS

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ABSTRACT

Even though usage of hydrogen (H_2) in the energy sector has gained a considerable amount of traction over the last decade, its high flame speed restricts its feasibility as a drop-in replacement for existing fuels in applications such as internal combustion engines and gas turbines. In order to address some potential issues, ammonia (NH_3) can be used in conjunction with hydrogen, as its slow reaction kinetics offset that of H_2 without compromising decarbonization efforts. However, simply replacing natural gas (primarily methane, CH_4) with hydrogen/ammonia blends is not trivial due to compatibility issues associated with bulk combustion characteristics. In this study, methane-ammonia-hydrogen ternary blends are comprehensively investigated in terms of laminar burning velocity, flame morphology, stability, and emissions in order to understand transitional regimes between fossil-based approaches and carbon-free alternatives. This work presents the analysis of the combustion properties of different ammonia/methane/hydrogen blends at varied equivalence ratios in an optically accessible constant volume combustion chamber (CVCC) coupled with Z-type schlieren visualization. The emissions from each experimental trial were analyzed using a Fourier Transform Infrared (FTIR) Spectroscopy system for detailed speciation. Furthermore, a computational approach was implemented to validate the laminar burning velocity results using an established mechanism already tested for ternary blends.

Keywords: Hydrogen, Ammonia, Natural gas, Laminar burning velocity, Fourier transform infrared (FTIR) spectroscopy, Emissions

NOMENCLATURE

CVCC	Constant Volume Combustion Chamber
FTIR	Fourier Transform Infrared Spectroscopy
LBV	Laminar Burning Velocity
S_b	Flame Speed
DAQ	Data Acquisition System

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Φ	Equivalence Ratio
PPM	Parts Per Million
GWP	Global Warming Potential

1. INTRODUCTION

Combustion of hydrocarbon fuels is a prevalent power generation technique employed in different transportation modes, such as road, aviation, and marine, as well as in energy production via power plants and stationary power generation. With increasingly stringent emission regulations and the depletion of conventional fossil fuel reserves, the search for alternative, cleaner sources of energy have become a top priority to mitigate the environmental pollution and energy crisis. Alternative fuels that can produce minimal, or zero, carbon dioxide (CO_2) are getting more attention. In that regard, both hydrogen (H_2) and ammonia (NH_3) have gained significant attention due to their promising characteristics for a sustainable and low-emission energy future [1, 2].

Hydrogen has been considered as an excellent clean energy carrier due to its exceptional properties, such as high gravimetric energy density, low minimum ignition energy and wide flammability limit [3, 4]. These characteristics of hydrogen result in a highly energetic combustion event and rapid propagation velocity that is nearly ten times higher than that of methane [5]; therefore, using pure hydrogen in the existing combustion system poses potential challenges. Ammonia also a carbon free molecule, has been considered as a potential hydrogen carrier with advantageous properties such as high volumetric energy density, wide flammability range, high octane rating, and ease of liquefaction [6, 7]. Combustion of pure ammonia is challenging due to its higher ignition temperatures, low energy content, and slower laminar burning velocity. The emissions from pure hydrogen combustion are limited to water vapor, whereas pure ammonia combustion can generate nitrogen oxides. Studies have shown varying values for the laminar burning velocity (LBV) of pure hydrogen and pure ammonia. In general, the LBV of pure hydrogen at standard temperature and pressure is around 280-295 cm/s [8], while the LBV of pure ammonia is around 7-12 cm/s

[9].

Since the LBV of methane increases with the addition of hydrogen [10], researchers have used different inert gases with this fuel blend to suppress the increased flame speed [11]. It is also possible to achieve methane-like flame speeds for these blends by adding ammonia to the mixture as the reported LBV of ammonia-methane blends is lower than that of pure methane combustion [12].

The investigation of ternary blends of hydrogen, methane, and ammonia offers a unique opportunity to optimize the performance and emission characteristics of these fuel blends. Keeping the higher percentage of methane can potentially help implementation of these fuel blends in existing applications without any major modifications. By blending these fuels in different proportions, it is possible to achieve a synergistic effect that can enhance combustion efficiency, reduce pollutant emissions, and improve overall engine performance while keeping the flame speed similar to that of natural gas [13]. Furthermore, the use of ternary blends potentially enables the utilization of existing natural gas applications, especially internal combustion engines, thus facilitating a smoother transition to a zero-carbon fuel based economy.

In this research, we aim to provide an in-depth understanding of the combustion behavior of hydrogen-methane-ammonia ternary blends, focusing on fundamental combustion properties such as laminar burning velocity and flame stability. Both experimental and simulation techniques are employed to achieve a comprehensive understanding of the combustion process. This study's findings will contribute to the development of novel combustion strategies and engine technologies that can effectively utilize hydrogen-methane-ammonia ternary blends.

2. METHODOLOGY

2.1 Experimental Setup

The study involved an evaluation of the combustion characteristics such as flame speed (S_b), laminar burning velocity (LBV), and emissions of ternary blends consisting of 60% methane and varying proportions of NH_3 and H_2 between 10% to 30% for an initial pressure 1 bar, and the equivalence ratio (0.6, 0.8, 1.0). Figure 1 shows a schematic of the experimental system consisting of a constant volume combustion chamber (CVCC) [14, 15], gas cylinders, pressure transducer, a centrally located spark ignition system and the Fourier transform infrared (FTIR) spectroscopy. The combustion chamber has a cylindrical geometry with an internal length and diameter of 140 mm providing a total volume of approximately 1.93 liters. The chamber is constructed from high-quality stainless steel to ensure its structural integrity and durability under high pressure and temperature conditions. To obtain accurate and repeatable results during combustion experiments, a high-temperature o-ring is used for proper sealing of the chamber and a vacuum level of less than 500 mTorr is maintained to confirm proper sealing. For filling each individual gas, the gas lines were vacuumed each time, and Dalton's partial pressure method was used to sequentially fill each fuel (such as CH_4, NH_3, and H_2), oxidizer, and nitrogen gas. Once the gas filling was completed, a 3-minute wait time was considered to ensure proper mixing of the gases inside the chamber before ignition [16]. Two electrodes are positioned on opposite

sides of the combustion chamber and the ignition control, high speed camera images and pressure traces are stored automatically using an in-house program with a LabVIEW DAQ system. The test conditions for each experiment were repeated three times to ensure results that were repeatable with a confidence level higher than 95%. The spherically explaining flames were visualized with a knife edge based z-type schlieren. The optical system consists of halogen light source (FSI-1060-250, 250W), two condensing lenses, a pinhole, a knife edge, two concave mirrors (with internal diameters of 15.2 cm and focal lengths of 152.4 cm) and a high-speed camera (CMOS Edgertronic SC2+). Figure 1 illustrates the optical setup of the Z-type schlieren system used in the CVCC combustion vessel, showing the path of light from the halogen source to the high-speed camera. The time-resolved images of the spherical flame propagation were captured at 8000 frames per second with a resolution of 384×384 pixels. A scaling factor of 4mm/pixel was utilized to determine the radius of the flame with an in-house MATLAB code.

Mashruk et al.[13] experimented with ternary blends of CH_4-H_2-NH_3 and found that if the blends have more than 30% H_2 in it, then hydrogen starts to dominate the characteristics of the fuel blend. Because of the high reactivity and heat release rate of hydrogen, blends with high concentration of H_2 had issues including flashbacks and unstable flames [13]. Furthermore, better stability was achieved in flames generated by blends that had more than 10% NH_3. This study focuses on experimentally and numerically examining three different blends of CH_4-H_2-NH_3 in terms of laminar burning velocity and emissions with the initial configuration shown in Table 1.

2.2 Laminar Burning Velocity

Laminar burning velocity (LBV) is an important fuel property that depends on mixture composition, initial pressure, and initial temperature. It not only affects the combustion duration in applications [17], but is also used in the validatation of chemical kinetic mechanisms. For the laminar burning velocity calculations, the acquired images undergo a transformation to greyscale and subsequent binarization to identify the precise boundary of the expanding flame. Then, the equivalent diameter of the flame in each frame of the capture video is determined based on the circumference of the flame to correct for any non-circularity. The flame speed and stretch rate are then calculated [18]. The unstretched flame speed was calculated from the relationship between the stretch rate and flame speed using a linear fit. To calculate the laminar burning velocity of the gas mixture, the average unstretched flame speed was multiplied by the expansion factor. Additional details of calculating LBV of different mixtures from the schlieren images has been described in a previous publication [19].

2.3 Numerical Simulation

The experimental values were further validated by performing LBV calculations for those 3 specific ternary blends of $CH_4 - H_2 - NH_3$ in Cantera 2.6 using the detailed chemical kinetics from Li et al. [20], which includes 128 species and 957 reaction steps. This chemical mechanism has been tested for a wide range of applications with different levels of H_2 and CH_4
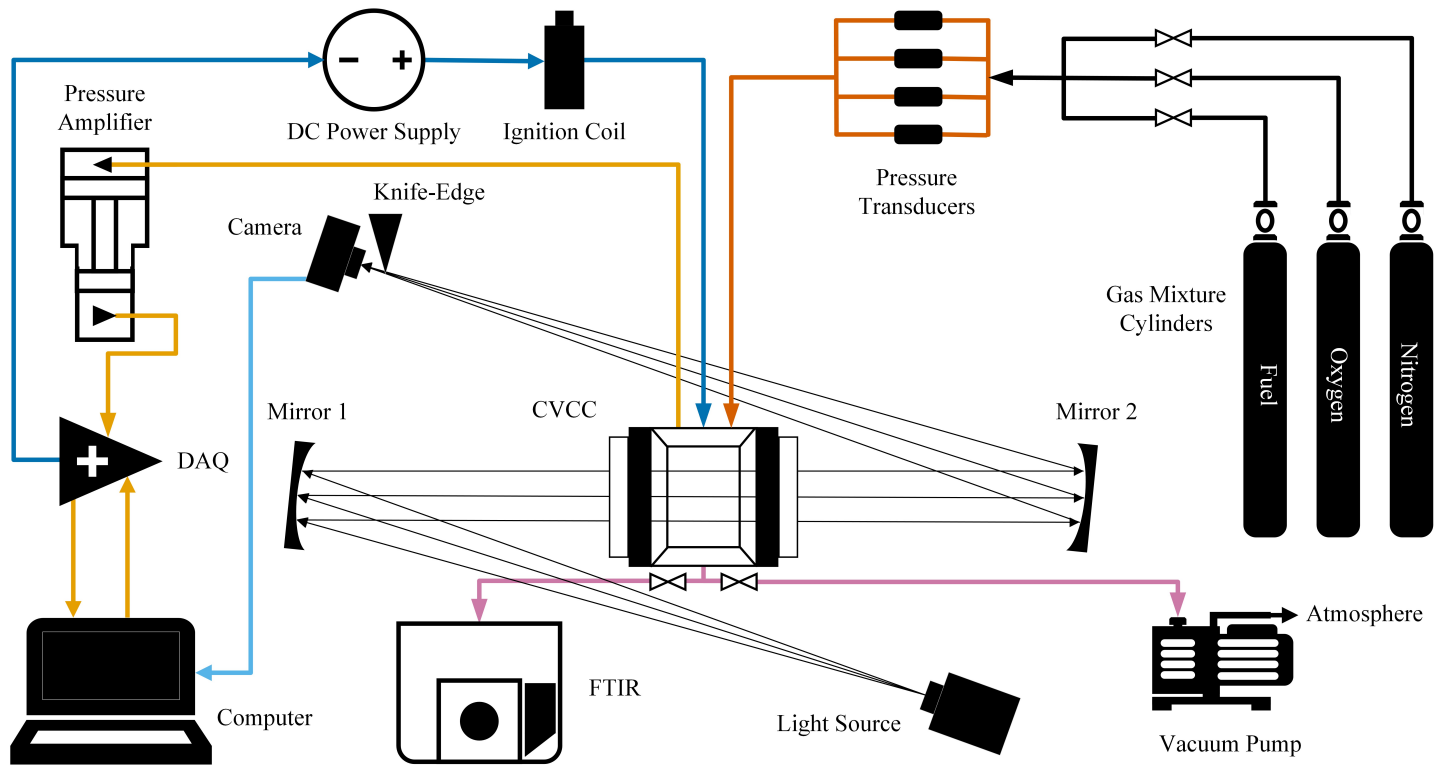
FIGURE 1: SCHEMATIC OF THE EXPERIMENTAL SETUP INCLUDING THE CVCC COUPLED WITH Z-TYPE SCHLIEREN VISUALIZATION, VACUUM PUMP, IGNITION SYSTEM, DAQ, AND FTIR.

TABLE 1: INITIAL CONDITIONS AND FUEL BLEND CONFIGURATION OF THE CASES TESTED.

Case	Gas Blends			Equivalence Ratio	Initial Pressure	Initial Temperature
	CH_4 (%)	H_2 (%)	NH_3 (%)			
1	60	10	30	0.8, 1.0, 1.2	1 bar	298 K
2	60	20	20	0.8, 1.0, 1.2	1 bar	298 K
3	60	30	10	0.8, 1.0, 1.2	1 bar	298 K

additions to NH_3 [13]. The initial pressure and temperature were set as the same as the experimental conditions at equivalence ratios ranging from 0.8 to 1.2.

2.4 Exhaust Sampling

Speciation was carried out by creating a calibration curve from reference spectra of each of the gases combined with a classical least squares fit in order to calculate the quantity of each exhaust species. gases were collected into a 200ml gas cell with a 2M optical path. The cell was equipped with a heater, temperature probe, and pressure transducer to use when comparing to reference spectra. Exhaust gases from each trial were dried of water and ammonia using an ice-bath heat exchanger. This elimination of water and ammonia enabled the quantification of CH_4, CO, CO_2, NO, NO_2 and N_2O.

2.5 Uncertainty Analysis

The errors associated in this study can be categorized in two major classes- error in preparing the gaseous mixture and error in the analysis of experimental data. The first type of error arises from the impurity of the gases used and the inaccuracy of the pressure transducers used to determine the partial pressures of the reactant gases while filling up the chamber. This error was minimized by using research grade gases with a purity level of 99.999% and high precision piezoresistive pressure transducers (Omega PX419) having an uncertainty level of ±0.08%.

Unlike the negligible errors of the first type, the compilation of errors made during the calculation of the flame front radius was significant and are depicted in the y-axis direction of LBV vs equivalence ratio plot. These errors were mitigated by repeating each experimental case three times. The standard deviation data generated from the repeated trials were utilized in determining the error bars.

Similar measures had been taken to ensure minimal error in the exhaust analysis procedure. The standard deviation data from the repetitive FTIR trials were used to evaluate the error bars shown in the y-axis direction of the exhaust sampling plots.
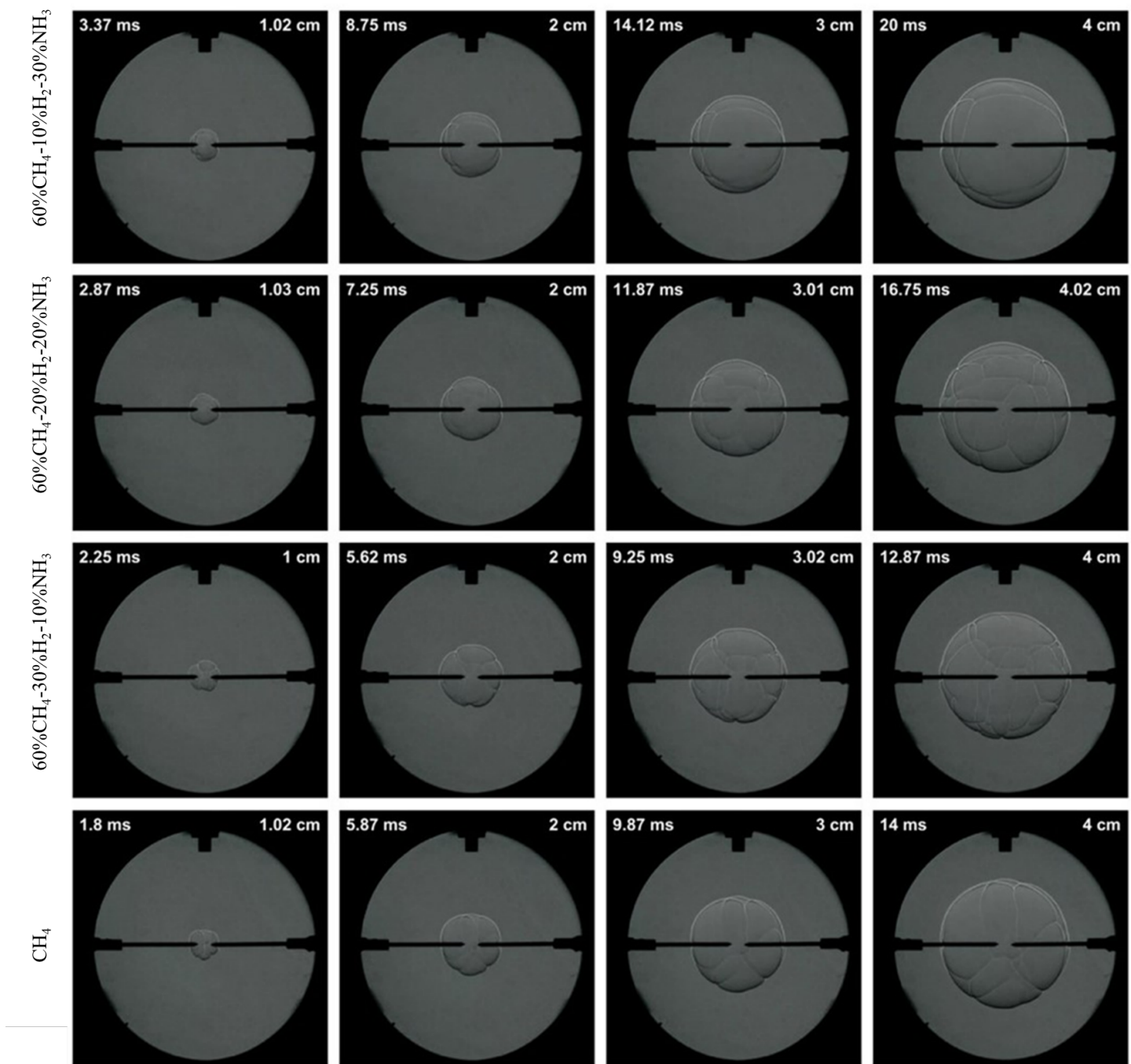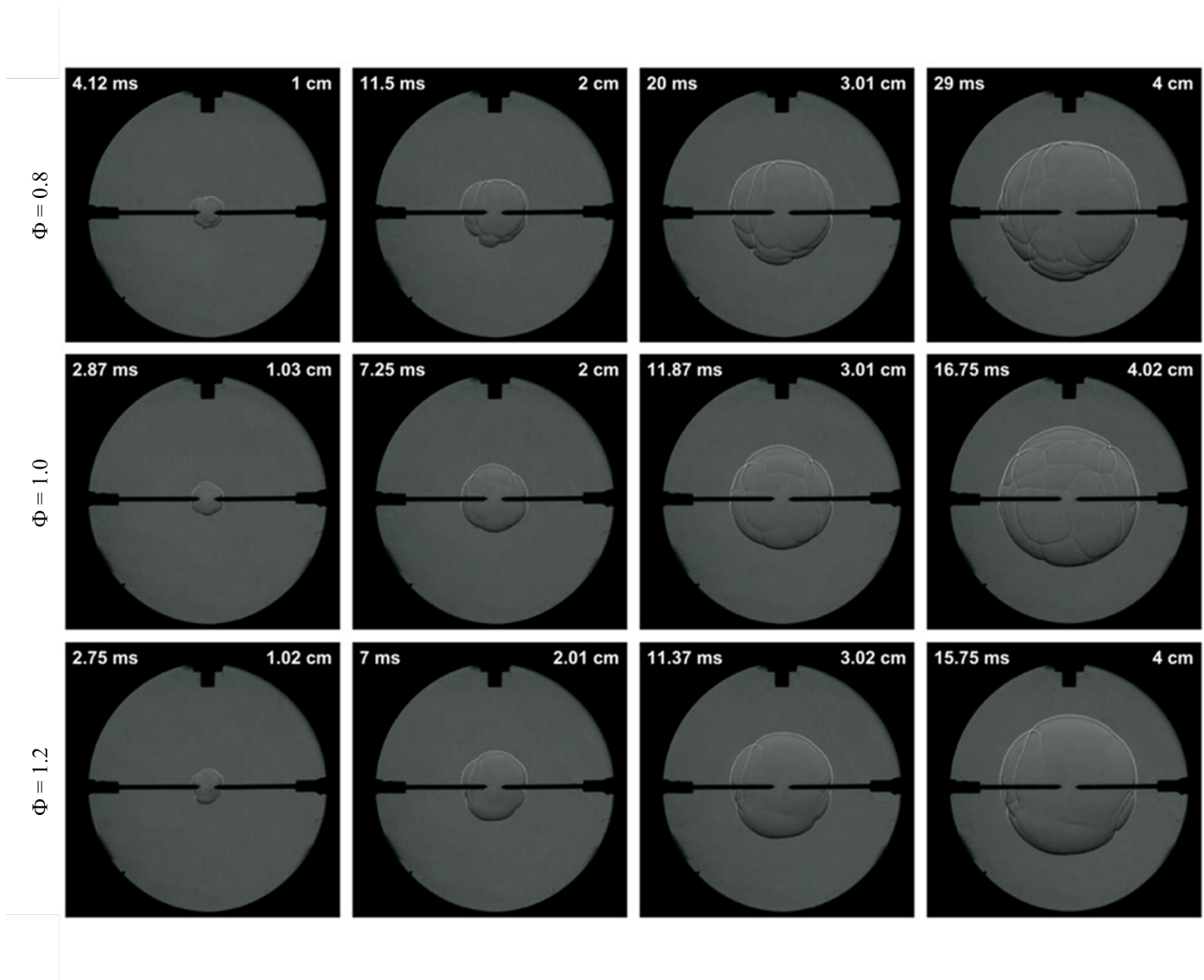
FIGURE 2: SCHLIEREN IMAGES OF SPHERICALLY EXPANDING FLAME FROM TERNARY BLENDS WITH VARYING AMMONIA/HYDROGEN RATIO AND PURE METHANE

FIGURE 3: SCHLIEREN IMAGES OF SPHERICALLY EXPANDING FLAME FROM 60%CH$_4$-20%H$_2$-20%NH$_3$ BLEND AT VARYING EQUIVALENCE RATIOS

3. RESULT AND DISCUSSION

3.1 Flame Morphology

Flame speeds of different ternary blends can be qualitatively assessed from the schlieren images of spherically expanding flame fronts inside the CVCC illustrated in Figure 2. Keeping the methane concentration constant at 60%, the centrally ignited flame front took 35% less time to reach a flame radius of 4 cm when the H$_2$ percentage was increased from 10% to 30% and NH$_3$ was decreased from 30% to 10%.

Figure 3 depicts the flame speeds for 60%CH$_4$-20%H$_2$-20%NH$_3$ blend at an initial pressure of 1 bar across different equivalence ratios. Due to the presence of H$_2$ and NH$_3$ in the fuel blend, the rich mixture of Φ = 1.2 is seen to be faster than the stoichiometric mixture, unlike the flame speed observed in pure methane combustion where the experiments implied that a mixture near stoichiometric conditions would generate the fastest LBV. These trends are discussed in detail in Section 3.2.

The instabilities seen on the spherically expanding flame surface, predominantly diffusive-thermal type at fuel lean conditions and hydrodynamic in nature at fuel rich configurations, become more prominent as the hydrogen concentration in the blend is increased [21].

3.2 Laminar Burning Velocity (LBV)

Figure 4 shows the comparison between experimental and simulated data for the laminar burning velocity of the tested fuel blends at three equivalence ratios (Φ = 0.8, 1.0, 1.2). In all the cases tested, the fuel rich mixture of Φ = 1.2 turned out to be the fastest. This increase in LBV was elevated even further as the hydrogen concentration was increased in the fuel blends. For the 60%CH$_4$-10%H$_2$-30%NH$_3$ the LBV saw an increase of 43.59% as the equivalence ratio was increased from 0.8 to 1.2. As the hydrogen concentration was increased to 20% and 30%, the increase in LBV was measured to be 59.20% and 70.59%,

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respectively.
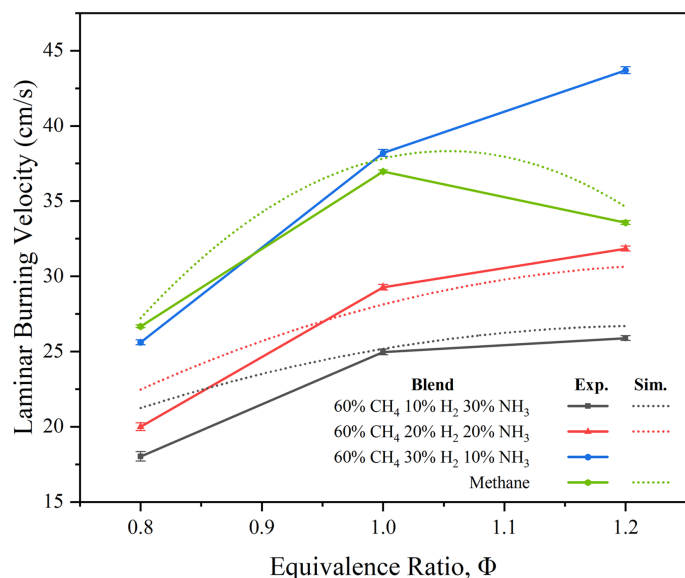
FIGURE 4: LAMINAR BURNING VELOCITY OF DIFFERENT BLENDS AT VARYING EQUIVALENCE RATIOS

When compared to pure methane combustion at fuel lean and stoichiometric conditions (26.67 cm/s for Φ= 0.8 and 36.97 cm/s for Φ = 1.0), the $60\%CH_4$-$30\%H_2$-$10\%NH_3$ blend comes the closest with a measured LBV value of 25.61 cm/s and 38.2 cm/s at similar equivalence ratios. The difference in their LBV values increases drastically at fuel rich conditions as pure methane's LBV falls off to be 33.57 cm/s while the ternary blend sees a rise in magnitude 43.69 cm/s at Φ = 1.2.

All these results were corroborated by the numerical simulations. It over-predicted the LBV values at fuel lean cases for all the blends but matched well with the experimental data at stoichiometric conditions. It over-predicted again at fuel-rich conditions for the blend with the least amount of hydrogen present. As the hydrogen concentration was increased in the blend, the mechanism started to under-predict the LBV value when compared to the experimental data.

3.3 Exhaust Analysis

The exhaust from the combustion trials is run through a Fourier Transform Infrared Spectroscopy (FTIR) unit. An inline ice-cooler condenses the water vapor from the the exhaust before directing it to the gas cell unit where exhaust speciation takes place, enabling the detection and measurement of unburned CH_4, CO_2, CO, NO_2, NO, and N_2O in the exhaust

3.3.1 Carbon Monoxide. CO is mainly produced from incomplete combustion due to a lack of oxygen. The exhaust data from the FTIR shows that replacing 40% CH_4 with blends of NH_3 and H_2 does not have a significant impact at fuel lean and stoichiometric conditions but results in a 34% reduction in CO generation when burning rich. This result is notable since the lack of availability of carbon does not completely translate to an equivalent reduction in CO, which reinforces the role of chemical kinetics in emissions analysis. Figure 5 also implies that by

increasing the NH_3 and decreasing the H_2 concentration reduces the CO production rate in exhaust even further.
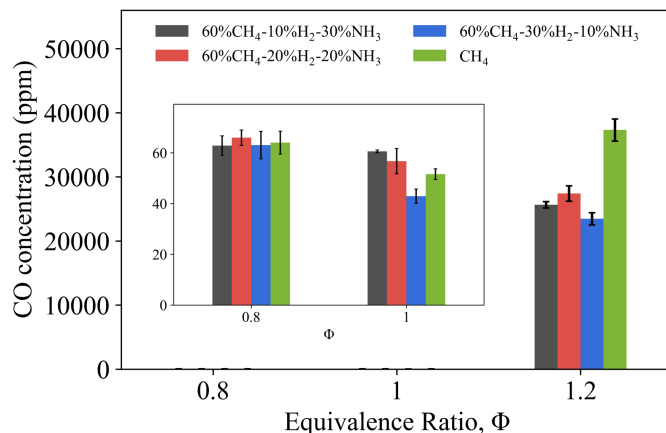
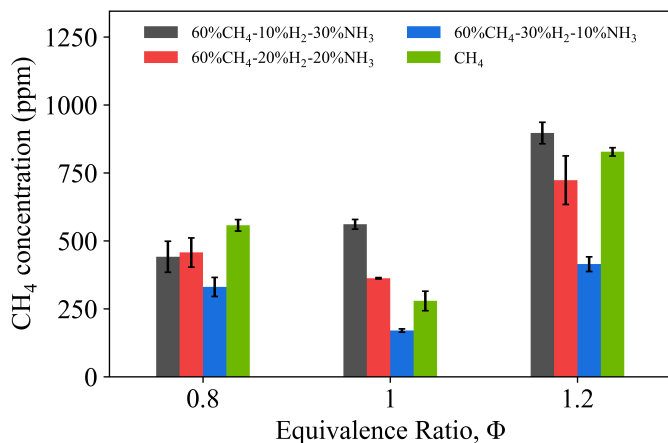FIGURE 5: CO CONCENTRATION IN THE EXHAUST AT VARYING EQUIVALENCE RATIOS

3.3.2 Carbon Dioxide. CO_2 is one of the major greenhouse gases produced from natural gas combustion. As expected, the FTIR data presented in Figure 6 shows that by replacing 40% CH_4 with blends of NH_3/H_2 it is possible to reduce the CO_2 production at all stoichiometries. It is also interesting to see that at fuel lean conditions, changing the ratio of hydrogen and ammonia in the blends does not have a significant impact on the production of CO_2, which is not the case at stoichiometric and fuel rich regions.
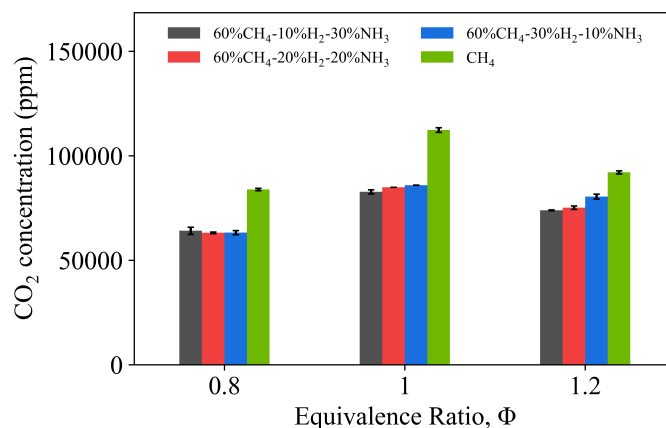
FIGURE 6: CO_2 CONCENTRATION IN THE EXHAUST AT VARYING EQUIVALENCE RATIOS

3.3.3 Unburned Methane. The FTIR data illustrated in Figure 7 produces expected results in terms of the high amount of unburned CH_4 seen at fuel rich region from pure methane combustion. But the blend of $60\%CH_4$-$10\%H_2$-$30\%NH_3$ produces under 500ppm of unburned CH_4 which matches that from stoichiometric methane combustion, making it a viable blend and suitable operating condition for different natural gas applications.

3.3.4 NO_x. Nitrogen oxides are formed by three different mechanisms [22]. The principal mechanism of NO_x formation

FIGURE 7: UNBURNED CH$_4$ CONCENTRATION IN THE EXHAUST AT VARYING EQUIVALENCE RATIOS

FIGURE 8: NO CONCENTRATION IN THE EXHAUST AT VARYING EQUIVALENCE RATIOS

FIGURE 9: NO$_2$ CONCENTRATION IN THE EXHAUST AT VARYING EQUIVALENCE RATIOS

FIGURE 10: NO$_X$ CONCENTRATION IN THE EXHAUST AT VARYING EQUIVALENCE RATIOS

in natural gas combustion is thermal NO$_x$, which occurs through thermal dissociation and succeeding reactions of oxygen (O$_2$) and nitrogen (N$_2$). Most NO$_x$ formed by this mechanism occurs in high temperature regions. NO$_x$ production rates have a positive correlation with three major factors: (1) oxygen concentration, (2) peak temperature, and (3) time of exposure at peak temperature. It is possible to reduce the NO$_x$ production by replacing some of the methane with blends of NH$_3$/H$_2$ to bring down the adiabatic flame temperature.

The second mechanism of NO$_x$ production transpires through early reactions of (N$_2$) and hydrocarbon radicals from the fuel. These reactions take place within the flame and are negligible compared to the amount of NO$_x$ formed by the thermal NO$_x$ mechanism.
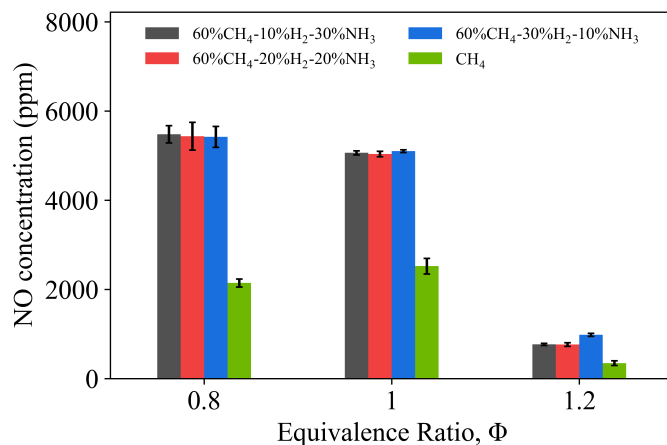
The third mechanism of NO$_x$ formation, called fuel-bound NO$_x$, is generated from the reaction of fuel-bound nitrogen with oxygen. Since methane does not contain nitrogen, NO$_x$ formation through this mechanism is typically insignificant. However, when using fuel blends containing NH$_3$, this mechanism begins to play a significant role in the total NO$_x$ production rate.

Figure 8 shows that all three blends produce less NO at fuel rich conditions when compared to NO generation data of pure methane at lean and stoichiometric configurations (i.e., natural gas engine operating conditions). At fuel lean and stoichiometric regions, the NO levels are very high when compared to the pure methane data due the increased presence of nitrogen. Varying the ratio of hydrogen/ammonia in the blends does not have an effect on the overall production of NO.
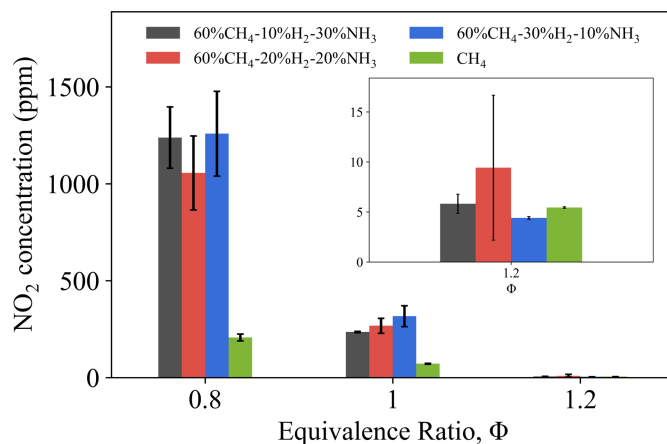
Due to the lack of oxygen in the fuel rich region, the production of NO$_2$ is the lowest not only for the tested blends but also for pure methane combustion, as shown in Figure 9.

Figure 10 represents the total NO$_x$ generated from the tested fuel blends and compares the data with pure methane combustion. The results imply that fuel rich operation results in a lower (around 68%) NO$_x$ emissions in comparison to fuel lean operation for all tested fuel blends containing NH$_3$ and H$_2$.
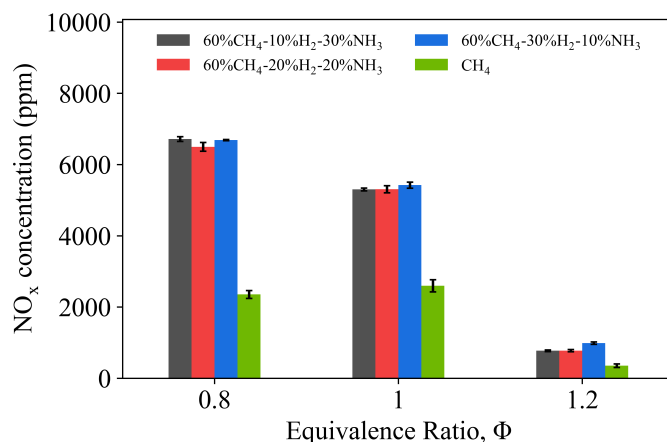
3.3.5 Nitrous Oxide. Nitrous oxide (N$_2$O) is one of the major greenhouse gases, with an estimated lifetime of 114 years

in the atmosphere and a global warming potential (GWP) value of 265 times that of carbon dioxide [23]. The high GWP value makes it critical to assess the N_2O production rate for any fuel blend, especially blends consisting of ammonia, which contains fuel-bound nitrogen.
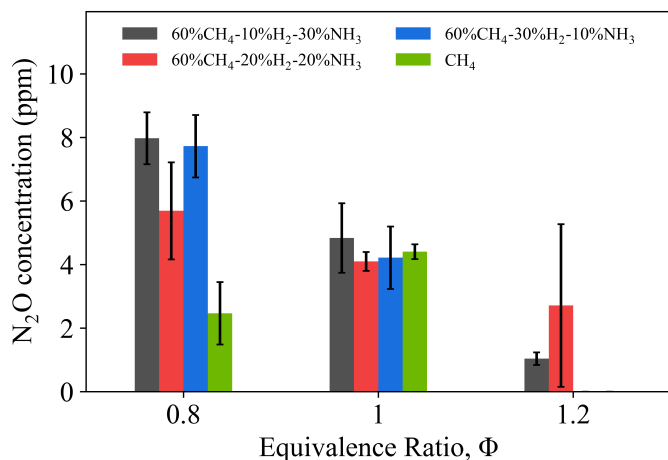
FIGURE 11: N_2O CONCENTRATION IN THE EXHAUST AT VARYING EQUIVALENCE RATIOS

Figure 11 illustrates the production of N_2O from the tested blends and pure methane at varying equivalence ratios. Even through N_2O generated by different blends were higher than pure methane combustion at fuel lean and stoichiometric condition, it is still under 10 ppm in all of the conditions tested. The rich cases produced even lower amount of N_2O, which were beyond the detectable range of the FTIR.

4. CONCLUSIONS

Three different blends of methane/hydrogen/ammonia were tested in an optically accessible CVCC to measure flame speeds and emissions. Experimented results were compared with pure methane combustion data and numerical simulations. An FTIR unit was deployed to compare the exhaust data at varying equivalence ratios to measure CO_2, CO, unburned CH_4, NO, NO_2, and N_2O for all blends. The results from the experiments can be summarized as the following-

- Hydrodynamic or diffusive-thermal instabilities appearing on the flame surface became more prominent with increasing hydrogen concentration and started to appear earlier as the mixture got leaner.

- Unlike pure methane combustion, which had the fastest LBV at stoichiometric conditions, all three tested blends had the fastest LBV at a fuel rich condition of $\phi = 1.2$. Both the measured and simulated LBV decreased as the mixture got leaner.

- The increase in LBV from $\phi = 0.8$ to $\phi = 1.2$ was 43.59% for 60%CH_4-10%H_2-30%NH_3 blend. This trend intensified further as the hydrogen concentration in the mixture was gradually increased.

- The 60%CH_4-30%H_2-10%NH_3 blend had a LBV of 43.69 cm/s at $\phi = 1.2$, which is about 18% faster than pure methane combustion at stoichiometric conditions. When both hydrogen and ammonia are set to be 20% each, the blend manifests a LBV of 31.84 cm/s which is 13% slower than stoichiometric methane combustion.

- at $\phi = 1.2$ all three blends tested succeeded in reducing total NO_x production by 68% when compared to that from pure methane combustion at fuel lean and stoichiometric regimes.

- The amount of CO can be reduced by keeping the ratio of hydrogen to ammonia low in the fuel blend. By bringing the hydrogen content down to 10% from 30%, there was an overall decrease of 18.18% in CO production.

The results show that the tested blends at particular conditions can match the flame speed of methane while reducing the CO_2, CO, and NO_x emissions. Future studies will explore other fuel blends and how they can improve emissions while maintaining a flame speed comparable to that found in existing systems.

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