Deflagration-to-Detonation Transition in Nitrous Oxide-Ethylene Mixtures and its Application to Pulsed Propulsion Systems

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In the past several years nitrous oxide has been more widely considered as a "safe", clean oxidizer for rocket propulsion systems, that was useable as both a monopropellant or as a bipropellant. Therefore, the present work investigated the use of nitrous oxide and ethylene as a bi-propellant mixture at elevated pressures. Earlier work with this bi-propellant mixture suggested that a steady detonation can be established in a combustion tube of larger L/d ratio and the associated flame acceleration prior to DDT could be studied more accurately. In order to use this bi-propellant mixture in a pulsed detonation engine quick flame acceleration is essential and this leads to a transition to detonation in a short duration of time. To study the flame acceleration and DDT behavior, tests were carried out in a combustion tube with L/d = 68 and detonation pressures were recorded using high pressure (100,000 psia) transducers. The detonation velocities were determined based on the time instances of measured pressure peaks and the distances between the transducers. Additionally, the pre-compression observed in the combustible mixture before transitioning to a detonation was also studied and described in this paper. Finally, the run-up distances from these tests were determined and compared with values for different fuel-oxidizer mixtures from literature.

Nomenclature

X_d	=	run-up distance normalized by tube diameter
k	=	constant of proportionality
p_o	=	initial pressure of bi-propellant mixture
m	=	exponent of initial pressure in the dimensionless run-up distance equation
kpsia	=	kilo-pounds per square inch
DDT	=	Deflagration-to-Detonation Transition

I. Introduction

THE decomposition and combustion properties of nitrous oxide (N₂O) have been studied for nearly a century. Technical interest in nitrous oxide intensified in the 1960s because of the compound's positive heat of formation, resulting in heat release during decomposition. Also, upon complete dissociation the products were pure nitrogen (N₂) and oxygen (O₂). These characteristics made nitrous oxide a unique oxidizer for combustion systems,

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and ignited interest in using N_2O in supersonic wind tunnels to increase enthalpy in order to simulate high speed flight conditions. Well before the 1960s, a nitrous oxide and coal hybrid rocket was developed and tested at I.G. Farben in Germany which produced 10-kN thrust for 120 s [1]. This was followed by its usage in Luftwaffe aircraft engines during World War II to improve high-altitude performance. The gas was chilled and liquefied and sprayed under pressure into the engine intake manifold using the system known as Göring Mischung 1 or GM-1. Around the same time, NACA investigated supercharging radial engines with nitrous oxide [2].

A great deal of work was done by the Air Force in the 1960s to characterize the decomposition and combustion of nitrous oxide. In 1962, Laughrey et al. [3] studied the decomposition and detonability of pure nitrous oxide at elevated temperatures and pressures, and while they were not able to achieve detonation they did observe high rates of chemical reaction they categorized as explosions. They concluded that it appeared possible that a detonation wave in nitrous oxide could be established in a longer tube. Later in 1963, Jost et al. [4] studied the detonation and chemical kinetics of hydrazine and nitrous oxide to assess the viability of nitrous oxide as an oxidizer for rocket propulsion. They measured detonation speeds of hydrazine-nitrous oxide mixtures at low initial pressures (on the order of 0.1 atm) and found the speeds to be near the ideal Chapman-Jouguet (CJ) detonation velocity.

Additional interest in nitrous oxide had arisen in the past several years as it was being more widely considered as a "safe", clean oxidizer for rocket propulsion systems, that was useable as both a monopropellant or as a bipropellant [5]. Grubelich et al. [6] replaced toxic or cryogenic oxidizers typically used in hybrid engines with nitrous oxide in the late 1980's and tested it with hydroxyl terminated polybutadiene (HTPB) as the fuel. According to their work, the nitrous oxide hybrid engine can have longer burn times than conventional solid propellant rocket motors for a given geometry. Several years later in 2001, Tyll et al. [5] tested nitrous oxide with propane in a bipropellant rocket engine. The nitrous oxide was catalytically decomposed and this exothermic process ignited the propane to produce sustained combustion. Tyll and co-workers proposed that performance improvements could be achieved by replacing the low vapor pressure propane with ethylene, which had a similar vapor pressure as nitrous oxide. Using ethylene as the fuel, their analysis predicted an increase of the specific impulse by several seconds. In addition, DiSalvo et al. [7] tested their patented constant volume rocket motor with nitrous oxide and propane in pulsed motor mode generating brief chamber pressure pulses on the order of 500-700 psia using injector inlet pressures of only 40-50 psia. In 2011, DARPA announced the Airborne Launch Assist Space Access (ALASA) program designed to produce a rocket capable of launching a 100 lb. satellite into low Earth orbit with Boeing contracted to develop the launch system. Boeing intended to lower the complexity of the launch vehicle and thus costs by powering the rocket with a monopropellant comprised of a pre-mixed combination of nitrous oxide and acetylene [8].

In the present work, the use of nitrous oxide and ethylene as a bi-propellant mixture at elevated pressures was investigated. Earlier work with these bi-propellant mixtures at higher initial pressures showed an extremely fast transition to detonations in a short closed vessel; 24.5 inches long with a 4 inch inner diameter [9]. The average detonation velocities from this work were as shown in Figure 1. The drop in velocity after the initial overshoot in Figure 1 suggested that a steady detonation can be established in a combustion tube of larger L/d ratio and the associated flame acceleration prior to DDT could be studied more accurately. In order to use this bi-propellant mixture in a pulsed detonation engine quick flame acceleration is essential and this leads to a transition to detonation in a short duration of time. To study the flame acceleration and DDT behavior, ethylene/nitrous oxide mixtures were tested in a large L/d, closed tube for a range of initial pressures.
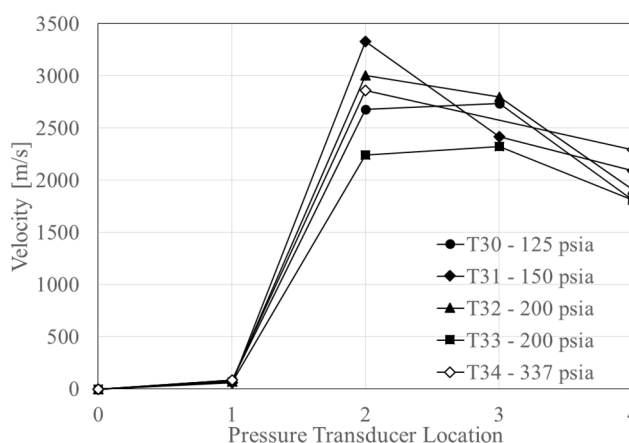

Figure 1. Detonation velocity vs. transducer location from previous etheylen/nitrous oxide tests in a short, closed tube [9].

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II. Experimental Setup

A 75 inch long closed combustion tube was used for this work with an inner diameter of 1.1 inches (L/d = 68), as shown in Figure 2. It was designed and fabricated at Zucrow Laboratories using a 1.5" XX-Heavy pipe, and was hydrostatically tested to 10,000 psia prior to installation. The ports on the side wall accommodated eleven PCB 109C11 transducers, 6 inches apart, to measure pressure peaks at each location along the length of the tube during the propagation of the combustion wave. One pressure transducer was installed in the end wall to measure the reflected pressure. These transducers were connected to a high frequency data acquisition system and recorded pressures at 600,000 samples/s/channel.

The setup was fabricated to supply nitrogen from the high pressure supply, and ethylene and nitrous oxide from respective bottles. Separate sonic venturis were used on the fuel and oxidizer lines to set mass flow rates of the propellants resulting in the appropriate final pressure for each test. A check valve was connected at each gas inlet on the tube to prevent back flow of gas mixture and combustion gases into the supply cylinders. The tube was heated to 100°F using a tape heater to prevent condensation of the propellant gases at high pressures. The ethylene and nitrous oxide gases flowed into the tube from opposing ports to facilitate mixing of the gases. The tube was purged with nitrous oxide before pressurization with the bi-propellant mixture and this ensured an atmosphere of nitrous oxide at ambient pressure thereby eliminating air or residual combustion gases in the tube.
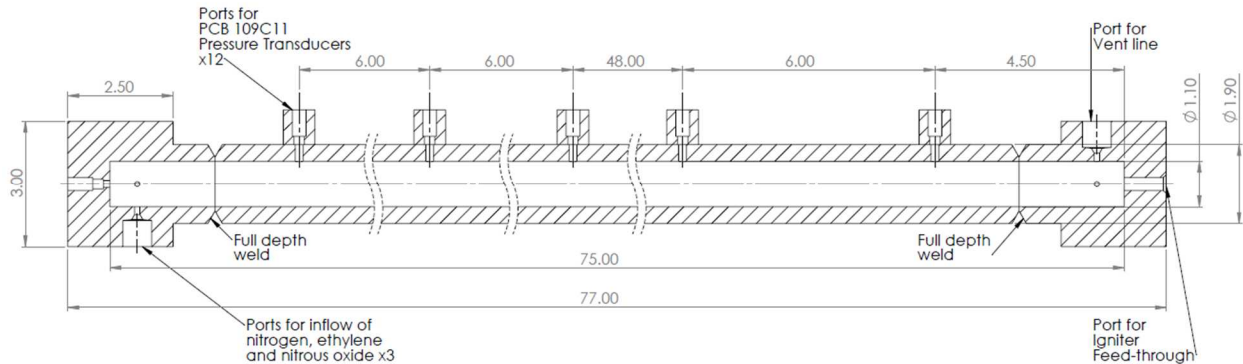

Figure 2. Diagram of the large L/d combustion tube used in the present work.

III. Experimental Results

The bi-propellant mixture was ignited by heating a nichrome wire 1/8" long soldered to the leads of a Kemlon feed through, as shown in Figure 3, mounted on the end wall. The nichrome wire was heated using an 18 V Li-Po battery switched by a solid state relay. Pressurizing the tube with the bi-propellant mixture was controlled by a LabVIEW auto sequence, and the gases were further allowed to mix via diffusion for 45 seconds before ignition. As part of the auto sequence, pressure and temperature from the low frequency instrumentation on the setup were recorded at a rate of 100 samples/s/channel. This information was used to estimate the actual O/F, initial pressure and initial temperature of the gas mixture prior to ignition. After each test the combustion gases were expelled through a vent line by opening two pneumatically actuated ball valves.
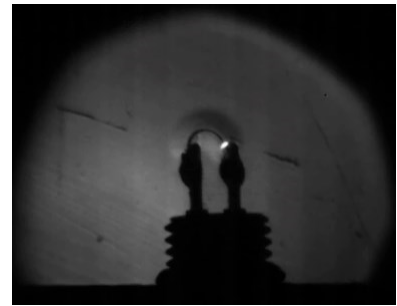

Figure 3: Nichrome bridge wire soldered to Kemlon feed through.

III. Experimental Results

A. Ethylene-nitrous oxide mixtures

Experimental runs were conducted with initial pressures from 40 to 150 psia at stoichiometric mixture ratios and the explosion pressures for each test were recorded. Based on the time instances of these pressure peaks and using the distance between pressure transducers (PTs), the propagation speeds of the combustion waves were determined. Multiple tests were carried out at each initial pressure to determine repeatability. The explosion pressures for one of the 50 psia tests were plotted in Figure 4 to illustrate the typical measurements in the tube during a test. The measured peak pressures from each location were averaged over multiple tests at the same initial pressure and were

plotted versus pressure transducer locations and shown in Figure 5. The recorded peak pressures were compared to the theoretical CJ detonation pressures calculated using the Shock and Detonation Toolbox [10] in Cantera [11], and tabulated in Table 1. This range of CJ pressures were also plotted in grey on Figure 5 for comparison.

Table 1: Theoretical CJ detonation pressures and velocities for different initial pressures.

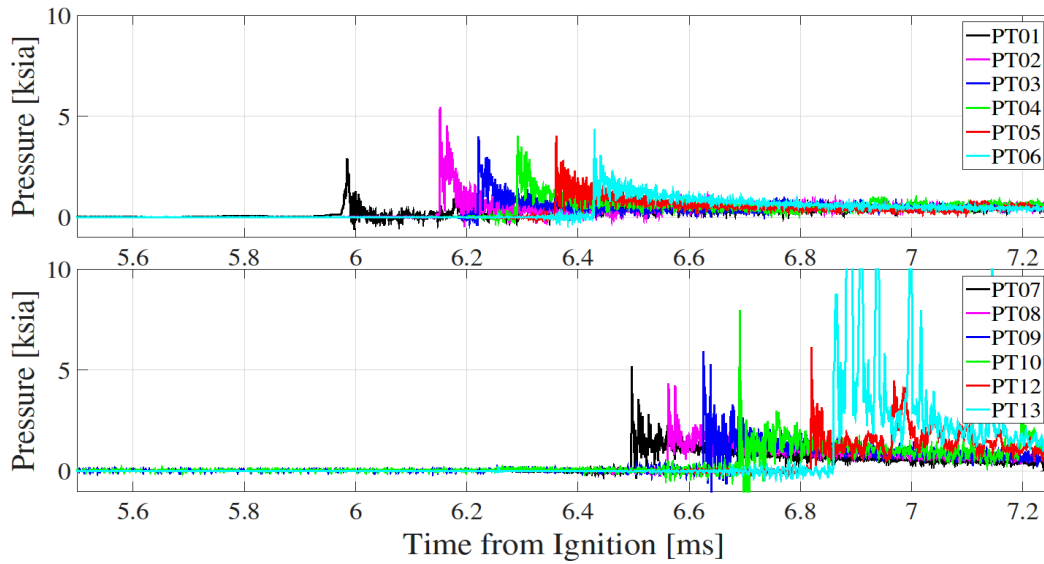| Initial Pressure [psia]	CJ Pressure (P_{CJ}) [psia]	CJ Velocity (D_{CJ}) [m/s]	Compressed Initial Pressure [psia]	Corresponding CJ Pressure [psia]	Corresponding CJ Velocity [m/s]
40	1575	2243	100	4045	2276
50	1983	2251	120	4876	2282
60	2393	2258	120	4876	2282
70	2804	2263	133	5420	2286
100	4045	2276	160	6507	2292
125	5086	2284	181	7450	2297
150	6133	2290	219	9051	2303

Figure 4: Pressure transducer signals for a stoichiometric ethylene/nitrous oxide test at 50 psia initial pressure.

The measured detonation pressures were higher than the theoretical calculations and on an average were 2.5, 2.4, 2.0, 1.9, 1.6, 1.45 and 1.46 times the CJ pressures for 40, 50, 60, 70, 100, 125 and 150 psia, respectively. The observation of detonation pressures higher than theoretical values was similar to the earlier test series [9]. During the deflagration process leading to the detonation the gases act like a piston and compresses the combustible mixture ahead of it, thereby increasing the "starting pressure of the mixture"; this results in a measured detonation pressure much greater than the theoretical value without pre-compression. This led to the observed over-pressure and with an increase in initial pressure there was a noticeable decrease in the average over-pressure factor. This drop in the over-pressure factor with increase in initial pressure indicated a lower compression of the combustible mixture due to faster DDT (shorter run up distance). The theoretical CJ pressure calculations indicated a linear correlation between in the CJ pressures and initial pressures which implied that the starting pressure of the mixture increased by the factors reported above. Thus, the gas mixture was compressed to pressures tabulated under "Compressed Initial Pressure" column in Table 1 which would result in detonation pressures and velocities tabulated alongside this column. Furthermore, these theoretical CJ pressures were close to the average pressures measured in the tube for each initial pressure. In addition to this, the detonation pressures oscillated as the wave propagated down the tube and this trend was more noticeable when pressure plots from individual tests were studied separately as opposed to the average values.
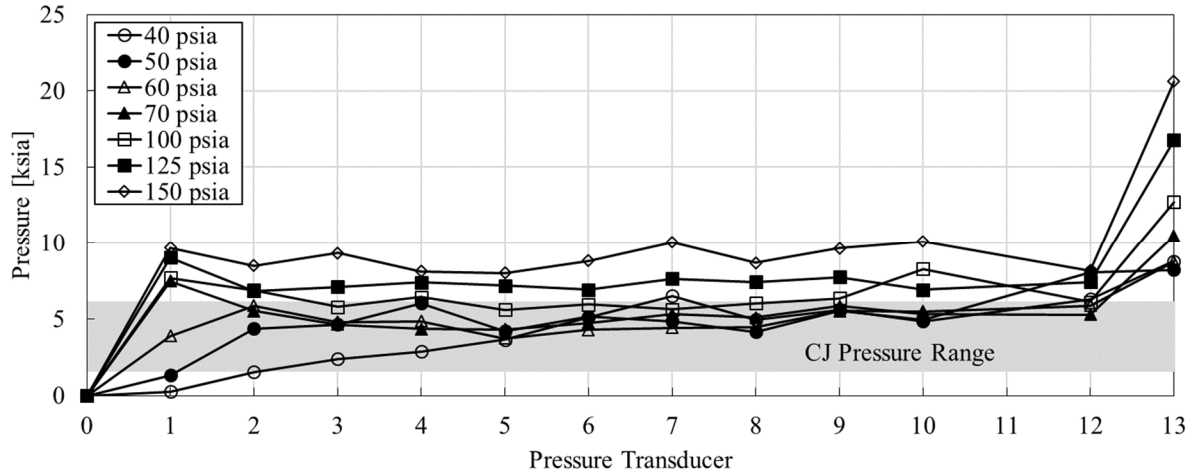
Figure 5: Peak pressures vs. transducer locations for the stoichiometric ethylene/nitrous oxide tests at initial pressures of 40 to 150 psia. The theoretical CJ detonation pressure range is indicated in grey.

One of the objectives of this work was to determine the repeatability of detonation parameters at a certain initial pressure. Multiple tests were conducted at each initial pressure and it was observed that detonation pressures at all transducer locations were variable. The variability of peak pressures at each initial pressure was compared by computing the standard deviation of the recorded pressures at every location and were plotted in Figure 6 for 50 and 100 psia. These two initial pressures were selected to show that the variability of peak pressures reduced with an increase in initial pressure and indicated better repeatability at higher initial pressures.
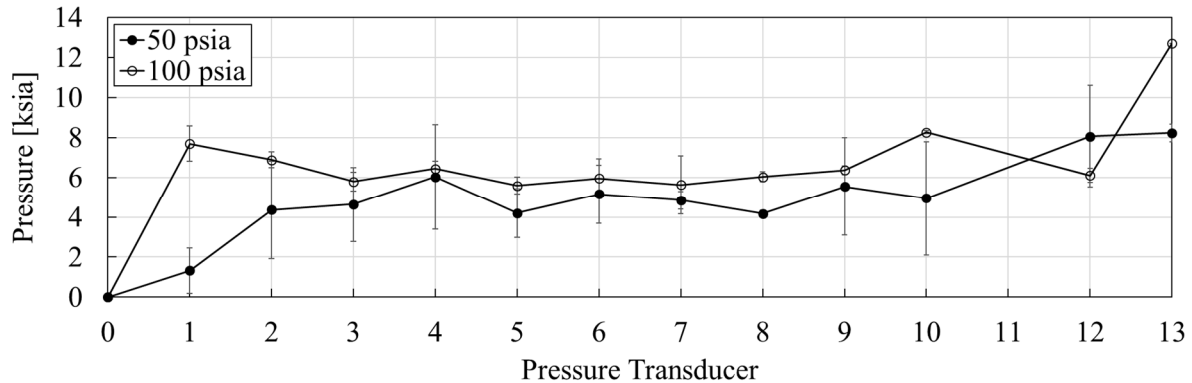

Figure 6: Variability of peak pressures represented as standard deviation from the mean for 50 and 100 psia initial pressure tests.

Further, based on the time instances of these pressure peaks and using the distance between the transducers, the propagation velocities of the combustion wave were estimated and were plotted versus pressure transducer locations in Figure 7. The CJ detonation velocities calculated using Cantera for these initial pressures were tabulated in Table 1 and the range of these values was indicated in grey on Figure 7. In comparison to CJ pressure, the CJ detonation velocity changed slightly when initial pressure was increased and hence the estimated velocities in Figure 7 were concentrated in a narrow band. Moreover, there was very little variability in the experimental detonation velocities, unlike detonation pressures.
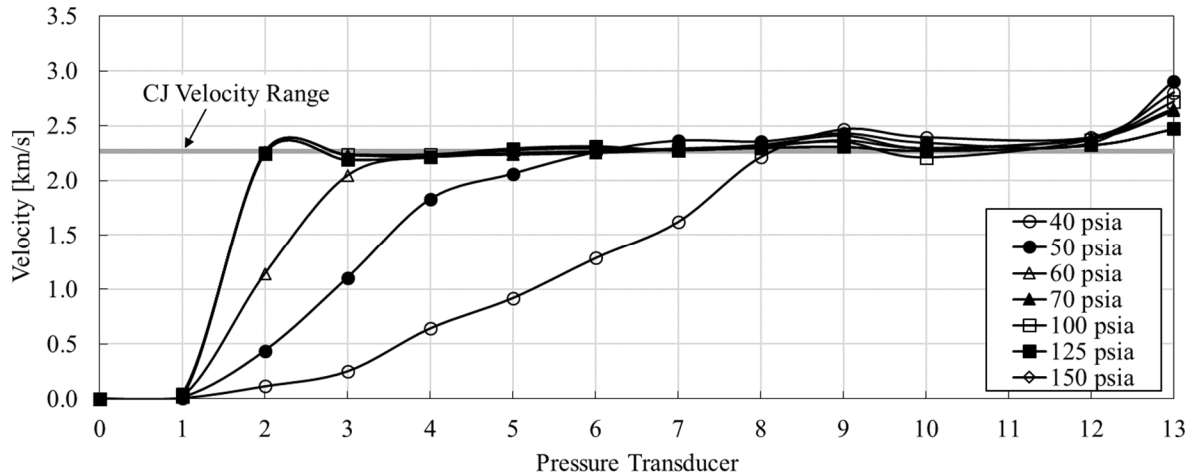
Figure 7: Velocity vs. transducer locations for the stoichiometric ethylene/nitrous oxide tests at initial pressures of 40 to 150 psia. The theoretical CJ detonation velocity range is indicated in grey.

The average velocity of the combustion wave between the igniter and the first transducer was less than 50 m/s, well below the CJ detonation velocity. The heated nichrome wire ignited the gases thermally and did not produce a shock, which would be necessary to directly initiate a prompt detonation. The nichrome wire igniter first initiated a deflagration that then transitioned to a detonation downstream from the igniter at different distances depending on the initial propellant gas pressure.

Bane et al. [12] studied undiluted hydrogen-nitrous oxide flames at low pressures which became unstable and turbulent nearly immediately following ignition. This increased flame surface area caused an elevated burning rate which led to significant flame acceleration. As with the hydrogen experiments, the nitrous oxide in the current mixture caused the deflagration to be unstable and turbulent. This resulted in its rapid acceleration to a critical velocity, beyond which the deflagration transitioned to a detonation. This process is discussed in detail by Lee [13] as part of the different mechanisms for deflagration-to-detonation transition (DDT) in smooth tubes.

B. Ethylene-oxygen mixtures

In order to compare the combustion properties of the above mixtures tests with air and oxygen as the oxidizers were carried out. These oxidizers were chosen due to the availability of detonation data from the literature that could be used for comparison. Firstly, tests were conducted with air at initial pressures from 50 to 125 psia, but none of the mixtures ignited with the nichrome wire ignition source. Finally, tests were conducted with oxygen at initial pressures from 30 to 125 psia. The mixtures at 30 and 50 psia did not achieve ignition, but explosion pressures were recorded for mixtures at 75, 100 and 125 psia, as shown in Figure 8. Similar to the tests with nitrous oxide, these mixtures produced explosion pressures higher than the calculated CJ pressure for ethylene-oxygen mixtures. The average over-pressure factor range was from 1.78 to 1.41, with higher factors associated with lower initial pressures. The average explosion pressure at a given initial pressure was lower for mixtures with oxygen when compared to those with nitrous oxide.
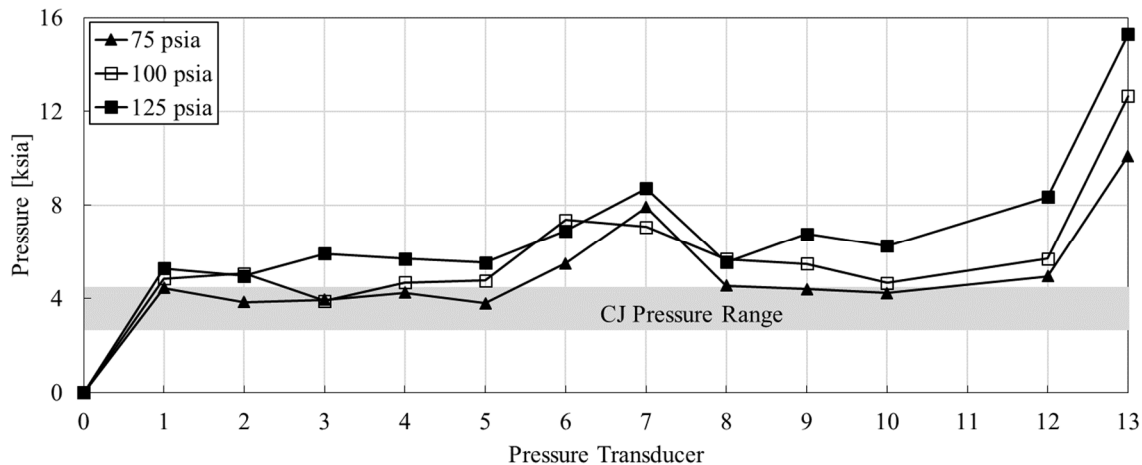
Figure 8: Peak pressures vs. transducer locations for the stoichiometric ethylene/oxygen tests at initial pressures of 75 to 125 psia. The theoretical CJ detonation pressure range is indicated in grey.

The combustion wave velocity was estimated for these mixtures as described in the previous sub-section and were plotted in Figure 9. It was observed that the average velocities were slightly higher than the CJ velocities for each of the initial pressures studied. These average velocities were also higher than those with nitrous oxide as the oxidizer. The determined velocities were more repeatable over multiple tests at the same initial pressure as compared to the pressure values, which had variabilities similar to the ethylene-nitrous oxide mixtures.
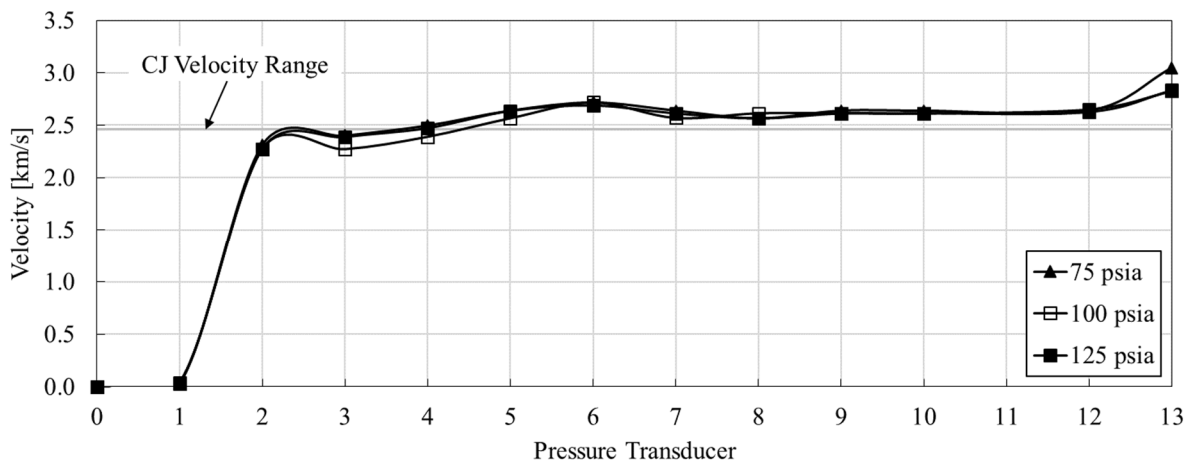

Figure 9: Velocity vs. transducer locations for the stoichiometric ethylene/oxygen tests at initial pressures of 75 to 125 psia. The theoretical CJ detonation velocity range is indicated in grey.

An explanation to the absence of ignition in mixtures with air and in low-pressure mixtures with oxygen was the low energy and small volume of the heated nichrome wire; below the critical ignition threshold for some of the mixtures. We believe that the energy released from the heated nichrome wire was sufficient to initiate the decomposition of nitrous oxide, which is exothermic in nature, and then this initiated the combustion of the overall mixture. However, with air and oxygen as the oxidizers there were no such exothermic decomposition reactions in the initial phase of the process that could ignite the fuel-oxidizer mixtures. Further investigation into this aspect can promote the use of nitrous oxide as the oxidizer when the use of higher energy ignition sources are inconvenient.

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IV. Theoretical Predictions

The theoretical detonation pressures and velocities for a range of initial pressures of the gas mixture were calculated using Cantera to compare with the experimental measurements. These calculations were performed for the following fuel-oxidizer combinations: ethylene-nitrous oxide, ethylene-air, and ethylene-oxygen. The combustion calculations with air and oxygen were performed for comparison and to understand the effect of different oxidizers. The results of these calculations were compared in Figure 10 and Figure 11.
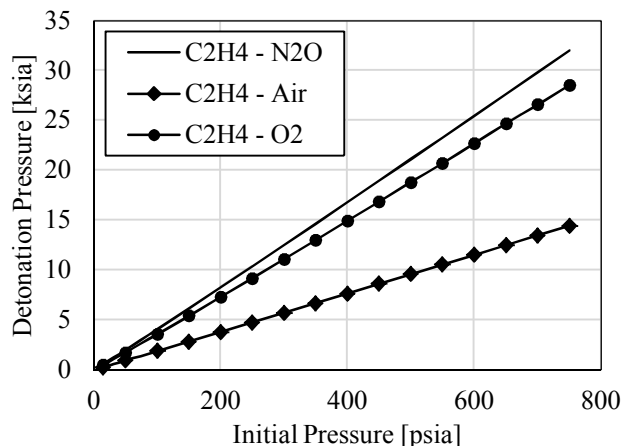

Figure 10: Theoretical CJ detonation pressures for stoichiometric gas mixtures vs. initial pressure.
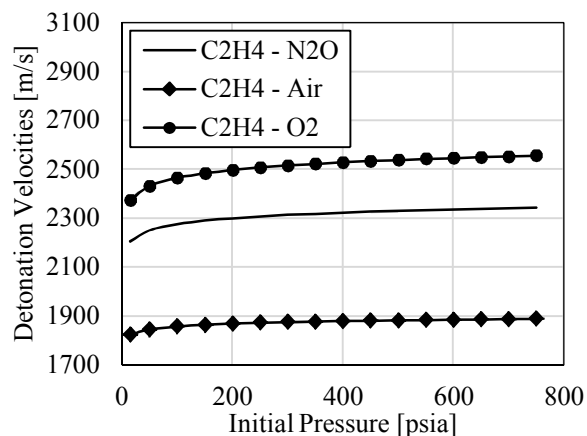
Figure 11: Theoretical CJ detonation velocities for stoichiometric gas mixtures vs. initial pressure.

The calculations were carried out at an initial temperature of $80.3^{\circ}F$ (300 K). For all cases, the CJ detonation pressure increased linearly with increasing initial pressure. The theoretical CJ detonation pressures were highest with nitrous oxide as the oxidizer and lowest with air as the oxidizer; the increase in the combustion pressure when changing from oxygen to nitrous oxide was approximately 11-13%. This trend was observed with the pressures measured in nitrous oxide and oxygen mixtures. Theoretical velocities for mixtures with oxygen were higher than those for nitrous oxide mixtures and this is concurrent with the measurements made during experiments. As discussed previously, the theoretical CJ detonation pressures were significantly lower than the pressures observed in the experiment. This discrepancy occurred because of the pressure increase by the deflagration before transitioning to a detonation.

V. Run-up Distances

Run-up distance is defined to be the distance between the source of ignition and that point in the tube where the wave first travels at the velocity of the stable detonation wave. The experiments described in the previous section can be used to estimate run-up distances for this bi-propellant mixture as a function of initial pressure (40-150 psia). The results were compared to run-up distance measurements of other fuel-oxidizer mixtures. Previous experiments by Bollinger et al. [14] [15] provided run-up distances for hydrogen-oxygen, hydrogen-nitrous oxide, methane-oxygen and carbon monoxide-oxygen mixtures. These run-up distances were non-dimensionalized with respect to the combustion tube diameter. The estimated dimensionless run-up distances for ethylene-nitrous oxide mixtures and those from literature were plotted against initial pressure on a log-log scale and shown in Figure 12.
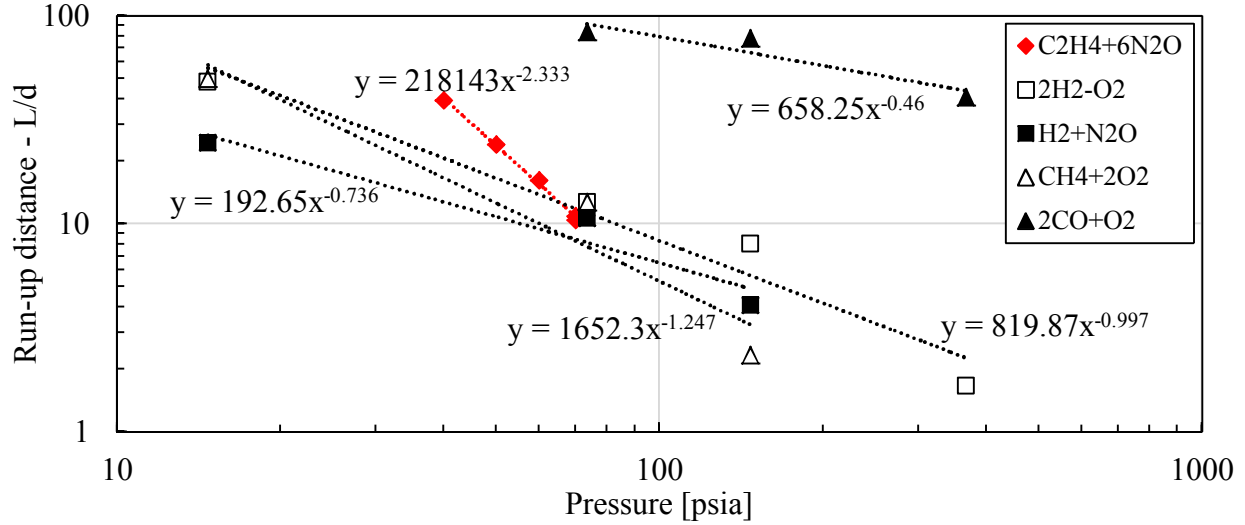
Figure 12: Dimensionless run-up distances for ethylene-nitrous oxide mixtures and various other mixtures for comparison.

The dimensionless run-up distance, X_d, decreased with increasing pressure, p_o, and Nettleton [16] suggested a power law of the form:

$$X_d = \frac{k}{(p_o)^m} \tag{1}$$

with k a constant. Trend lines attached to the run-up distances produced values of m in the range of 0.4 to 2.333, with the largest power associated with ethylene-nitrous oxide. Hence, the run-up distance for ethylene-nitrous oxide mixtures depended on the initial pressure as follows:

$$X_d = \frac{218143}{(p_o\,[psi])^{2.333}} \tag{2}$$

This equation suggested a faster drop in run-up distance as initial pressure increased for stoichiometric ethylene-nitrous oxide. This trend estimated a dimensionless run-up distance of 0.25 for ethylene-nitrous mixture at an initial pressure of 350 psi. This was shorter than the run-up distance for hydrogen-oxygen detonations at this pressure and the trend we observed indicated that the deflagration-to-detonation transition was almost instantaneous in ethylene-nitrous oxide mixtures at initial pressures higher than 350 psi. This instantaneous transition to detonations in ethylene-nitrous oxide mixtures at elevated initial pressures can improve the performance of pulsed detonation engines or similar propulsion systems which rely on quick flame acceleration to detonations.

VI.   Conclusion

The current series of tests were conducted to investigate detonations in ethylene-nitrous oxide mixtures in a large L/d combustion tube to understand their applicability to bipropellant propulsion systems. In all the tests conducted, the measured pressures significantly exceeded the theoretical CJ detonation pressures. Given that the ignition energy was intentionally far too low to achieve direct initiation, the DDT was due to the highly turbulent deflagrations which led to rapid acceleration to a critical velocity beyond which the deflagrations transitioned to detonations. In addition to this, Lee [13] suggested that the flame front couples with reflected pressure waves and causes localized explosions. These localized explosions creates a shock wave that will lead to an overdriven detonation immediately after transition. During the deflagration the gases behaved as a piston and compressed the combustible mixture ahead of the flame front thereby resulting in a detonation in mixtures of higher initial pressure as opposed to the initial pressure. Commonly known as pressure piling, this has the potential to cause higher pressures than a directly initiated detonation. The oscillations observed in the measured pressures were caused by detonation instability. The

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detonation wave velocity in some parts of the combustion tube exceeded the CJ velocity, indicating an overdriven detonation.

These tests were ignited with a heated nichrome wire and the energy was sufficient to exothermically decompose nitrous oxide, which initiated a combustion event in ethylene-nitrous oxide mixtures at 30 psia (no transition to detonation was observed). However, this method of ignition could not initiate combustion in ethylene-air mixtures even when the initial pressure was increased to 125 psia and could only initiate combustion in ethylene-oxygen mixtures at initial pressures of 75 psia and above. In order to achieve ignition in mixtures with air and oxygen, tests will be conducted in the future with a spark plug and a high voltage ignition system.

Finally, the dimensionless run-up distance for these tests were estimated and compared with run-up distances available in literature for various fuel-oxidizer mixtures. Although, ethylene-nitrous oxide mixtures have run-up distances higher than some fuel-oxidizer mixtures at low initial pressure, the trend exhibited by the estimated values indicated that quick deflagration-to-detonation transitions can be established in these mixtures at elevated initial pressures. This, in collaboration with the ability to ignite ethylene-nitrous oxide mixtures with low-energy ignition sources can be applied to existing pulsed detonation engine prototypes to investigate their effects on performance. Future work with ethylene-nitrous mixtures will include the development of a computational model to support these tests and understanding the chemical kinetics associated with the detonations.

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