

Methane–air detonation experiments at NIOSH Lake Lynn Laboratory

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ABSTRACT

The methane–air detonation experiments are performed to characterize high pressure explosion processes that may occur in sealed areas of underground coal mines. The detonation tube used for these studies is 73 m long, 105 cm internal diameter, and closed at one end. The test gas is 97.5% methane with about 1.5% ethane, and the methane–air test mixtures varied between 4% and 19% methane by volume. Detonations were successfully initiated for mixtures containing between 5.3% and 15.5% methane. The detonations propagated with an average velocity between 1512 and 1863 m/s. Average overpressures recorded behind the first shock pressure peak varied between 1.2 and 1.7 MPa. The measured detonation velocities and pressures are close to their corresponding theoretical Chapman–Jouguet (CJ) detonation velocity (D_{CJ}) and detonation pressure (P_{CJ}). Outside of these detonability limits, failed detonations produced decaying detached shocks and flames propagating with velocities of approximately $1/2 D_{CJ}$. Cell patterns on smokefoils during detonations were very irregular and showed secondary cell structures inside primary cells. The measured width of primary cells varied between 20 cm near the stoichiometry and 105 cm (tube diameter) near the limits. The largest detonation cell (105 cm wide and 170 cm long) was recorded for the mixture containing 15.3% methane.

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1. Introduction

Explosions caused by natural gas and coal dust accumulations continue to occur in underground coal mines in the USA. Since 1976, a total of 185 coal miners were killed and many were seriously injured as a result of underground coal mine explosions. Between 1976 and 2010, at least 25 explosions involving methane and coal dust occurred in the active areas of coal mines, resulting in at least 1 and up to 29 fatalities in each explosion. From 1986 to 2006, at least 12 known explosions involving methane alone occurred in the abandoned and sealed areas of coal mines (Zipf, Jr., Sapko, & Brune, 2007) including Sago Mine in West Virginia in 2006 with 12 miners killed, Darby Mine in Kentucky in 2006 with 5 miners killed, and Blacksville Mine in West Virginia in 1992 with 4 miners killed.

Researchers from the National Institute for Occupational Safety and Health (NIOSH) and the Naval Research Laboratory (NRL) are engaged in fundamental experimental and computational research

on explosion processes involving mixtures of methane and air. Explosive mixtures that form in coal mines are essentially mixtures of air and natural gas composed primarily of methane with small amounts of ethane and other alkanes. Though most accidental gas explosions in coal mines are deflagrations, the worst-case scenario involves detonations that can be extremely destructive and can generate explosion pressures up to 10 MPa on reflections. It is therefore important to know the conditions in which natural gas–air mixtures can or cannot detonate. This research is aimed at understanding explosion pressures that can develop in coal mines and the factors that lead to high explosion pressure and possible transition to detonation.

To carry out large-scale experiments with test mixtures of methane and air, researchers constructed an explosion tube measuring 105 cm in diameter and 73 m long located at the NIOSH Lake Lynn Laboratory (LLL) and about 60 km south of Pittsburgh. The methane used in these experiments is natural gas containing about 97.5% methane, about 1.5–1.7% ethane, less than about 0.05% other higher hydrocarbons, and the balance air and carbon dioxide. The experiments are being conducted in two phases. In the first phase, reported herein, detonations in test mixtures of methane and air are created by a direct initiation process that uses a near-stoichiometric, detonable mixture of methane and oxygen as

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a strong ignition source. In the second phase of the work to be reported later, researchers will examine detonations created through a deflagration-to-detonation transition (DDT) using weak spark ignition sources.

2. Previous experiments and methane–air detonations

Existing experimental data from methane and air explosion tests does not definitively answer this question: Given a flammable mixture of methane and air of sufficient volume and lateral extent that may exist in coal mine tunnels or entries, can the mixtures develop into a detonation from a weak spark ignition source? Laboratory-scale experiments examined three different methane–air explosion problems: 1) explosions of methane–air that are confined in pipes, 2) explosions of methane–air that are unconfined and in free space, and 3) explosions of methane–air that are partially confined by congested spaces, such as a process facility. The conclusion regarding the ability of methane–air to either detonate or develop high explosion pressures depended, in part, on the problem geometries under consideration.

The first problem involves explosive mixtures of methane–air confined within pipelines used for gas distribution or in process industries. In these experiments, the explosion spreads in one direction along the length of the tube. Using direct initiation of detonation, Kogarko (1958) and Wolanski, Kauffman, Sichel, and Nicholls (1981), p. 1651 found that the lower concentration limit (LCL) needed to sustain detonation ranged from about 6.3 to 8% methane in air, while the upper concentration limit (UCL) ranged from 13.5 to 14.5%. Recent work by Matsui (2002) found similar values for the LCL (7.5%) but a lower value for the UCL (11.5%). Bartknecht (1981), using a turbulent flame jet for ignition, shows that the tube length-to-diameter needed for a detonation to develop ranged from 75 to 125 depending on the tube diameter. In a review paper of detonation research in tubes, Lee (1984) summarized that on the order of 50–100 tube diameters are required for DDT to develop, starting with a weak ignition source in a smooth tube. The early work on methane–air detonation was conducted within smooth tubes less than 61 cm in diameter. Results showed that test mixtures of methane–air could sustain a detonation if it was initiated by an external, strong ignition source, and several researchers, such as Gerstein, Carlson, and Hill (1954) and Bartknecht (1981), were able to achieve detonation by DDT in smooth tubes. Some of the earlier work may have contributed to the perception that methane–air mixtures can only detonate under extraordinary circumstances not seen in the practical world. However, Lindstedt and Michels (1989) observed DDT with stoichiometric methane–air in 5 cm diameter tubes using Shchelkin spirals to create varying surface roughness of the tube.

In the 1970s and early 1980s, numerous research groups (Bull, Elsworth, Hooper, & Quinn, 1976; Nicholls, Sichel, Gaboriel, Oza, & Van Der Molen, 1978; and Parnarouskis, Lind, Raj, & Cece, 1980) examined the second problem—explosions of unconfined methane–air—which could develop from the catastrophic release of liquid natural gas during transport or storage. Unlike the prior discussion with explosions in tubes, the explosion in this case develops freely in all three spatial dimensions. These research groups used large volumes of methane–air or methane–oxygen–nitrogen mixtures and tried to sustain detonation in the test mixture using a high explosive charge as the initiator. None of the experiments produced sustained detonation in a stoichiometric methane–air mixture. Bull et al. (1976) supported by Nicholls et al. (1978) suggested that 22 kg of solid explosive is required to produce detonation in an unconfined stoichiometric methane–air cloud, which is a highly unlikely occurrence. No experiments with large volumes of unconfined methane–air mixtures produced

detonation either by direct initiation or DDT, and researchers concluded that detonation with unconfined methane–air is unlikely. This early work with unconfined methane–air mixtures also contributes to the impression that methane–air mixtures are very unlikely to detonate. However, work by Kuhl et al. (1972) and Strehlow and Baker (1975) shows that devastating overpressures can develop with an accelerating flame or a rapid deflagration, and that detonation is not necessary to achieve high dangerous overpressures.

Recent studies have focused on the third problem—explosions of methane–air that is partially confined in congested spaces. This kind of explosion could occur if a methane–air mixture develops within a process facility, which contains obstacles such as pipes, tanks, reaction vessels, support structures, and other equipment. The obstacles both confine the flow, which acts to accelerate the flame, and interrupt the flow path. This leads to turbulence and additional flame acceleration. The increased flame speed leads to the development of blast and shock waves, which in turn may lead to detonation in the explosive mixture. Ciccirelli and Dorofeev (2008) summarize recent studies of criteria for the onset of detonations in obstructed channels based on cell size, tube diameter, blockage ratio (BR), and other geometric factors. The minimum diameter to observe the onset of detonations is $d > \lambda$ where d is the unobstructed passage diameter and λ is the detonation cell size (Peraldi, Knystautas, & Lee, 1986; Ciccirelli & Dorofeev, 2008).

Moen, Funk, Ward, and Rude (1983) reported the first measurements of the detonation cell size for stoichiometric methane–air at about 28 cm. Lee (1984) and Bartknecht (1993) report a detonation cell size of about 30 cm, while Shepherd (2006) gives a range from 25 to 35 cm for stoichiometric methane–air. Kuznetsov et al. (2002) provided cell size data for a range of methane in air compositions. At 8.5% methane, the measured cell size is 44 cm; at 9.5% it is 19 cm, and at 12%, it is 33 cm.

Early research on gaseous detonation considered the "run-up length", i.e. the distance required to develop a supersonic flame with respect to the combustion products which then undergoes DDT. For smooth tubes, empirical observations show that the run-up length may range from 50 to 100 times the tube diameter (Lee, 1984; Bartknecht, 1993; van Wingerden et al., 1999; Kolbe & Baker, 2005). Based on theoretical models, Ciccirelli and Dorofeev (2008) provided relationships for the length required by a flame to reach supersonic velocity with respect to the combustion products, but not DDT. For methane–air in a smooth tube, this length is about 80 tube diameters. In obstructed channels with a BR of 0.3, this length decreases to about 20 diameters, and for a BR of 0.6, it decreases to about 10 diameters.

Recent research developed a geometrical criterion for the development of detonation in obstructed tubes. The characteristic geometrical size, L , necessary to sustain detonations is:

$$L > 7\lambda$$

where λ is the detonation cell size (Dorofeev, Sidorov, Kuznetsov, Matsukov, & Alekseev, 2000; Dorofeev, 2002; Ciccirelli & Dorofeev, 2008). Here, L is computed as:

$$L = D / (1 - d/D)$$

where D is the tube diameter and d is the unobstructed tube diameter.

Kuznetsov et al. (2002) conducted the most recent detonation experiments with stoichiometric methane–air in 17.4- and 52.0-cm diameter tubes with BRs of 0.3 and 0.6, respectively. In some experiments, a sustainable detonation developed by DDT. In other cases, the detonation could not be sustained. This ambiguity has caused many people in the US coal mining industry to question

whether high pressure gas explosions or detonations could occur in a coal mine.

3. Design and construction of the explosion tube

In designing an explosion tube for methane–air, researchers considered three criteria: 1) the minimum diameter to support detonation, 2) the minimum length to develop a supersonic flame, and 3) a suitable geometry to support detonation. In addition, the explosion tube should have dimensions similar to typical coal mine tunnels or entries.

By the minimum diameter criterion ($d > \lambda$), a 30 cm-diameter tube is required to observe detonation with a stoichiometric methane–air mixture in a smooth, unobstructed tube. Fuel-rich or fuel-lean test mixtures will require a larger tube diameter to observe detonation. The 105 cm diameter tube should enable observation of detonation in a wide range of methane–air mixtures. At a BR of 0.3 and 0.6, the unobstructed tube diameter d is 88 and 66 cm, respectively, which is sufficient to support detonation with a stoichiometric mixture and should be sufficient to support detonation with lean or rich methane–air test mixtures.

The length-to-diameter ratio is about 70. If the tube were smooth, i.e., no blockages, it is possible, but appears unlikely, that this tube is long enough to accelerate the flame to supersonic velocity in the test mixtures. However, with a BR of 0.3 or 0.6, the tube length is adequate to accelerate the flame.

For a 105 cm-diameter tube with a BR of 0.3 and 0.6, and unobstructed diameters of 88 and 66 cm, the characteristic geometrical size, L , is 6.5 and 2.8 m, respectively. In both cases, L is greater than the length, 7λ , required to sustain detonation, which is 2.1 m.

Typical coal mine tunnels or entries measure 6 m-wide by about 2 m-high. The equivalent diameter for these rectangular openings is about 3 m. The explosion tube diameter is 105 cm (1.05 m) and is thus similar in scale to coal mine tunnels or entries.

The explosion tube at NIOSH LLL, shown in Fig. 1, consists of 1) the tube; 2) the supporting systems for gas injection, mixing, sampling, and analysis; 3) the ignition system; 4) the diagnostic equipment; and 5) a control system. The steel tube has a wall thickness of 9.5 mm, minimum yield strength of 248 MPa, and an ultimate tensile strength of 414 MPa. For stoichiometric methane–air mixtures, the quasi-static CJ detonation wave pressure is 1.66 MPa, and that pressure could persist for time periods up to 0.010 s. Based on hoop stress calculations with this quasi-static



Fig. 1. NIOSH LLL explosion tube with control building in the distance.

pressure, the safety factor against deformation is about 2.70, and the safety factor against rupture is 4.50. Shock waves could reflect from the detonation tube walls at a pressure about 2.54 times greater than the CJ pressure or 4.4 MPa (Landau & Lifshitz, 1987), and they might have durations on the order of a few milliseconds. Numerical simulations of methane–air detonation in channels (Kessler, Gamezo, & Oran, 2009) show that the localized areas of the tube where detonation waves might reflect have dimensions measuring a few tens of centimeters square. In these small areas, the tangential stress is about 243 MPa, and the safety factors are about 1.02 for deformation and about 1.70 for rupture. Higher transient pressures of 7–14 MPa could develop when newly formed detonations resulting from DDT propagate in a shock-compressed gas. The highest dynamic pressure recorded so far in the NIOSH LLL explosion tube is about 7 MPa, and the tube shows no evidence of deformation or damage.

Prior to filling the explosion tube with methane to obtain the desired test mixture composition, a 0.15 mm-thick plastic diaphragm is placed over the open end of the tube. The gas mixing system enables creation of homogeneous methane–air test mixtures inside the tube with predefined composition. A 2.25 kw blower fan draws gas through 6 inlets and an intake manifold, and it exhausts through another manifold with 7 outlets. This system can circulate the test mixture in the tube in about 3 min, and mixing times are about 30 min.

A sample draw system enables collection of test mixture samples remotely for analysis with an infrared gas analyzer. Three samples are collected along the length of the tube to ensure that the test mixture is mixed homogeneously and has the desired composition. Two samples of the gas mixture are drawn after the mixing is completed, and these are analyzed off-site using gas chromatography. In most of the experiments with this detonation tube, researchers are able to obtain the desired volume % methane in the test mixture to within $\pm 0.2\%$.

For direct initiation of detonation in the methane–air test mixture, a plastic bag filled with a stoichiometric methane–oxygen mixture is inflated at the closed end of the tube after methane–air mixing in the tube is complete. The bag is made of 0.15 mm plastic film and has a cylindrical shape with a diameter of about 1 m. Detonation in the methane–oxygen mixture is ignited with a non-electric blasting cap that is placed at about 0.5 m from the closed end of the tube.

The tube is equipped with 23 quartz–piezoelectric-type pressure transducers and 23 light sensors located as shown in Fig. 2. The light sensors are silicon phototransistors with a rise time of about 2 μ s and a spectral response from 600 to 1100 nm. Sensors are mounted at the end of a small tube that is 15 cm long with an inside diameter of 0.32 cm. The field of view for the light sensor is about 5 cm on the opposite side of the 105 cm-diameter detonation tube.

The piezoelectric pressure sensors have a range of 0–6900 kPa and $\leq 1 \mu$ s rise time. In early experiments with the detonation tube, the pressure sensors were threaded directly into the detonation tube steel. The speed of sound in mild steel is about 5900 m/s, and extraneous vibrations induced in the steel by the methane–air combustion arrive at the pressure sensor prior to the arrival of the detonation wave, which travels on the order of 1800 m/s. To shield the pressure sensors from this noise, the pressure sensors are decoupled from the tube using the mount shown in Fig. 2. The pressure sensor is suspended over a hole in the detonation tube by double-studded silicone gel anti-vibration mounts.

The sensors are connected to two separate 24-channel National Instruments CompactRIO and eight-slot backplane with a 3-million-gate field-programmable gate array. The first CompactRIO for the light sensors has six analog modules, which are 4-channel, 16-bit, 0–5 V DC and simultaneously sampled at 50 kHz. The

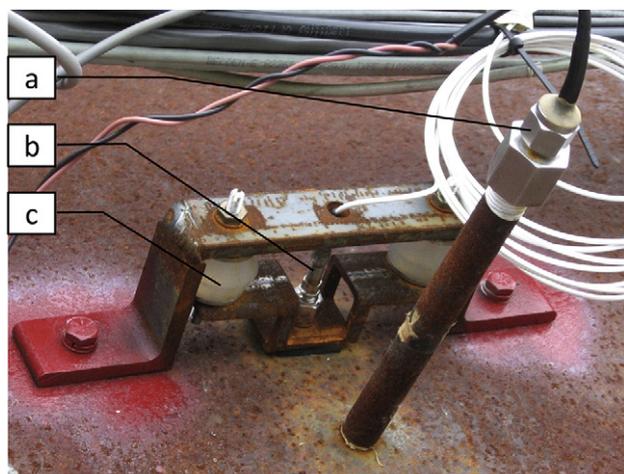


Fig. 2. Light (a) and pressure sensors (b) and anti-vibration mount (c).

second CompactRIO for the pressure sensors has six modules that are 4-channel, ± 5 V, 24 bit integrated electronic piezoelectric (IEPE) sensors, and also simultaneously sampled at 50 kHz. For synchronization of the light and pressure signals, one channel from each CompactRIO records a 50 Hz (± 2 V) square wave produced by a signal generator.

The time resolution for all recorded signals is 20 μ s, as defined by the data acquisition system (DAS) sampling frequency. The time error between two signals generated by a wave propagating between two sensors located 3 m apart can reach 40 μ s. If the detonation velocity is 1800 m/s, the uncertainty in position of the detonation wave is 0.072 m in 3 m or 2.4%. Therefore, the uncertainty in the measured detonation velocity is also 2.4% or ± 43 m/s. If the velocity is constant, the uncertainty is significantly reduced using multiple sensors at greater distances apart.

Before each experiment, two smokefoils are placed between 0.15 and 2.8 m from the open end of the tube. The 1.2 m by 1.2 m smokefoils are made of 1.6 mm-thick aluminum sheets rolled to a 0.5 m radius of curvature. Smokefoils are secured to the inside top of the tube with bolts around the perimeter of the foils. The upstream edge of each foil is inserted into a welded metal slot. The foils are covered with soot produced by acetone flames.

All control for detonation experiments with the tube is done remotely in the LLL control building located about 300 m away from the detonation tube. Firing the primary shot fire circuit triggers the data acquisition system. In case the primary shot fire circuit fails to ignite the methane-oxygen bag and the test mixture, two additional independent shot fire circuits are available to ignite the flammable mixture using electric matches. It is also possible to vent the test mixture to atmosphere using the gas mixing system.

4. Results from direct initiation experiments

During 2009 and 2010, more than 30 explosion tests were conducted to determine the detonation characteristics of various mixtures ranging from 4 to 19% methane in air. Each experiment attempted direct initiation of detonation in the test mixture using a bag containing about 2.9 m³ of methane-oxygen mixture as an igniter. The diagnostic equipment used in these experiments allowed for the identification of detonations using four methods: 1) comparison of measured flame and shock velocities to the computed D_C values, using pressure and light sensors placed in pairs every 3 m on the tube wall; 2) comparison of the recorded pressures to the computed pressure profiles for ZND detonations; 3) measurement of the separation between the shock and the flame using the time difference between pressure and light signals recorded at the same location; and 4) the patterns produced on smokefoils placed near the open end of the tube.

Measured detonation parameters are compared to theoretical values computed with CHEETAH 3.0 (Fried, Glaesemann, Souers,

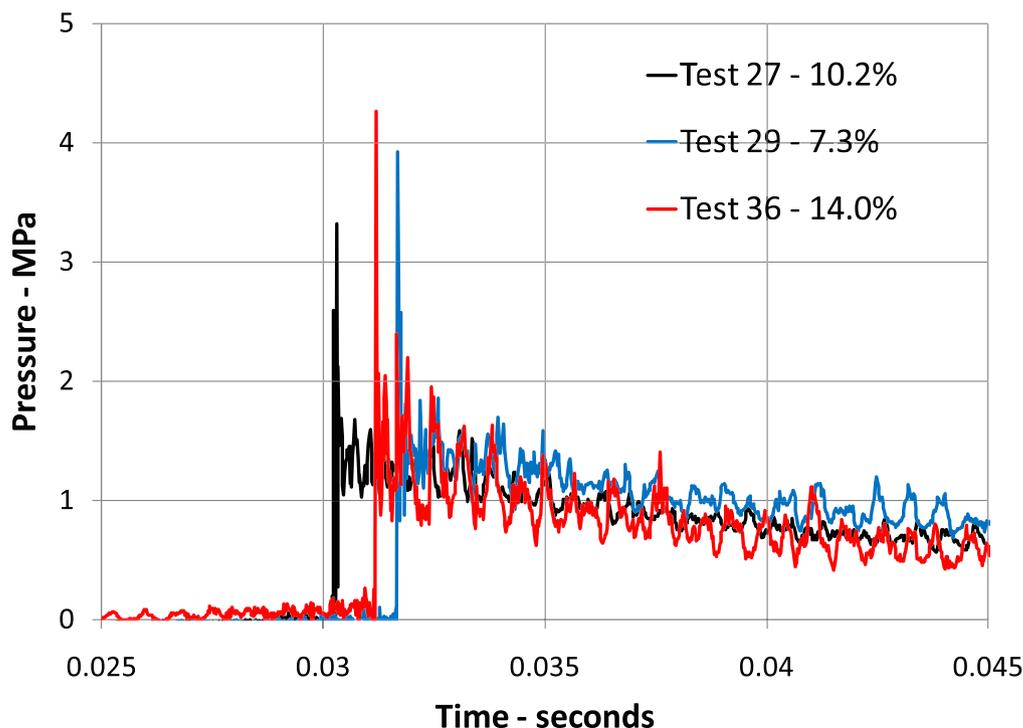


Fig. 3. Typical pressure-time records for fuel-lean, near-stoichiometric, and fuel-rich mixtures of methane in air.

Howard, & Vitello, 2000). CHEETAH is a thermochemical program that solves thermodynamic equations to find chemical equilibrium of reactant and product species. For input, CHEETAH requires reactant species, in this case methane and air, and their initial temperature and pressure. CHEETAH determines the equilibrium product species and calculates their final temperature and pressure, along with the constant volume (CV) explosion pressure, the CJ detonation wave pressure, and the CJ detonation wave velocity.

Fig. 3 shows typical pressure-time records from three different tests at 7.3, 10.2, and 14.0% methane in air, representing fuel-lean, near-stoichiometric, and fuel-rich mixtures. The igniter bag containing the methane-oxygen mixture is about 3 m long and affected pressure and light measurements in the first 10 m of the detonation tube. As expected, pressure measurements in this region were higher. The measured detonation velocity, determined from the time difference between stations using both pressure and light sensors, was also slightly higher in this region. Beyond 10 m in the tube, however, pressure-time records at each sensor were consistent in magnitude and shape. Measured detonation velocity between stations using both pressure and light sensors indicated a constant detonation velocity and therefore a stable detonation.

Table 1 summarizes the detonation data extracted from these tests as compared to theoretical values computed with CHEETAH. The measured detonation pressure is estimated as the pressure immediately behind the leading shock signal. By inspection of Fig. 3, the measured detonation pressure is estimated with an accuracy of about ± 0.3 MPa. The relative error on these detonation pressure measurements is about 20%. As indicated in Table 1, the estimated detonation pressures are lower than theoretical calculations by 1–15%.

The measured detonation velocity is computed between measuring stations located beyond 10 m, where the detonation is unaffected by the ignition source. Based on measured first arrival times, Fig. 4 shows the position of the shock and flame versus time for test 27 using a near-stoichiometric test mixture. If the detonation wave is stable, it is possible to measure its velocity between measuring stations more than 3 m apart and obtain greater precision. As indicated in Table 1, the estimated detonation velocities are within $\pm 2\%$ of theoretical calculations.

Fig. 5 shows the smokefoil record for test 27 using a near-stoichiometric test mixture. The cell patterns are irregular and the cell sizes are large compared to other fuel-air mixtures. Measurements of the detonation cell size were done by direct observation of the smokefoils using a tape measure for scale. Although photographs of the smokefoils were taken, no attempt has yet been made to enhance the images digitally to extract

Table 1

Comparison between theoretical and measured detonation parameters for fuel-lean, near-stoichiometric, and fuel-rich mixtures of methane in air.

Test number	29	27	36
Methane in air mixture (%)	7.3	10.2	14.0
CJ detonation pressure—CHEETAH (MPa)	1.539	1.758	1.594
Measured detonation pressure (MPa)	1.5	1.5	1.55
Difference (%)	-2.5	-14.7	-2.8
CJ detonation velocity—CHEETAH (m/s)	1702	1828	1781
Measured detonation velocity (m/s)	1689	1831	1756
Difference (%)	-0.8	+0.2	-1.4
Number of detonation cells observed	3	8	4
Range for detonation cell size (cm)	46 to 68	22 to 30	51 to 78
Average detonation cell size (cm)	55	25	61
Computed detonation cell size (cm)	70	27	60
Difference from computed (%)	-21	-7	+2
Time between pressure spikes (sec)	0.00025	0.00006	0.00023
Cell size (Dorofeev et al., 1992) (cm)	42	11	40
Difference from computed (%)	-40	-59	-33

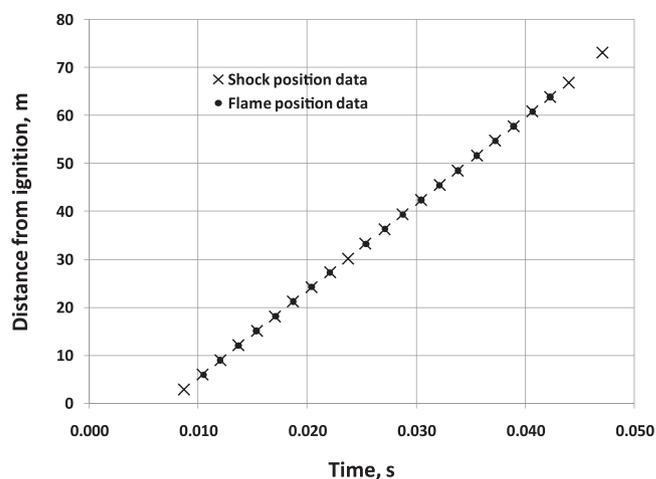


Fig. 4. Shock and flame position versus time for test 27 with 10.0% methane in air.

detonation cell information. Table 1 indicates the number of detonation cells observed, their size range, average size, and the expected size based on a model developed by Gavrikov, Efimenko, and Dorofeev (2000). The agreement between the model and measurements is reasonable considering the precision of the cell size measurements.

Fig. 6 shows the data points of measured detonation velocity versus methane in air % for 30 experiments over a range of mixtures of methane in air. The theoretical CJ detonation velocity versus methane in air % is also included on Fig. 6 as a solid line. Detonation in the test mixture is sustainable from about 5.3 to about 15.5% methane in air. Test mixtures less than 4.9% or greater than 16.0% methane in air did not sustain combustion, either by detonation or deflagration, when ignited by the strong methane-oxygen ignition source used in these experiments. The difference between the measured and theoretical detonation velocities is less than 10 m/s or 0.6%. These experiments confirm earlier methane-air experiments by Lindstedt and Michels (1989), which measured the detonation velocity at 89% of the theoretical value using a 5 cm



Fig. 5. Smokefoil record of test 27 with 10.0% methane in air. Average cell size is about 25 cm. Note 1 m-long scale.

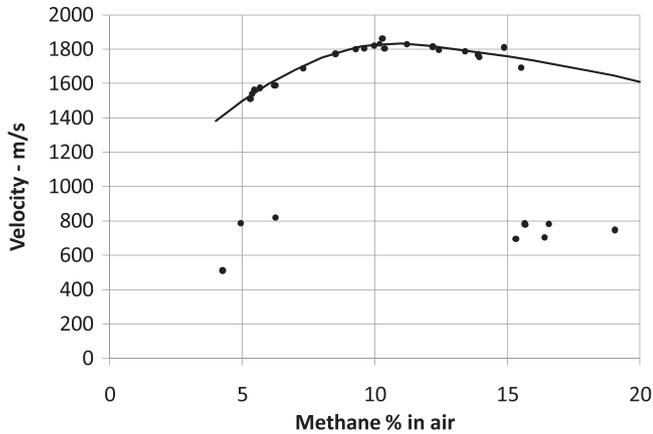


Fig. 6. Measured detonation velocity (data points) and CJ detonation velocity (solid line) versus methane in air.

diameter tube. Near the limits, the difference may increase to about 30–40 m/s or 2.5% less than theoretical expectations. Beyond the detonation limits, the shock velocity decreased to about 500–800 m/s for mixtures less than 5% methane in air and to about 700–800 m/s for mixtures greater than 16% methane in air. These observed shock velocities are about $\frac{1}{2} D_{CJ}$.

During Tests 17 through 24, the igniter bag was filled with 3.25 m³ of methane-oxygen mixture, and the possibility exists that the bag broke prematurely or leaked into the test mixture, thus corrupting the results. Rupture of the igniter bag did occur during Test 25, which is not included in this data set. From Test 26 forward, the igniter bag was filled with a smaller quantity of methane-oxygen mixture, which solved the earlier possible problem. Data from Tests 17 through 24, except for Test 18, is still included in Figs. 6 and 7, and the agreement between the measured and theoretical detonation velocities is good.

Fig. 7 shows the data points of measured detonation pressure versus methane in air % for the experiments. The theoretical CJ detonation pressure versus methane in air % is also included as a solid line. The measured detonation pressure is just a visual estimate of the steady-state CJ detonation pressure after passage of the main shock wave in the pressure-time records. The error in these detonation pressure measurements is estimated at ± 0.3 MPa. There is good agreement between measured and theoretical values of detonation pressure, considering the magnitude of the error in

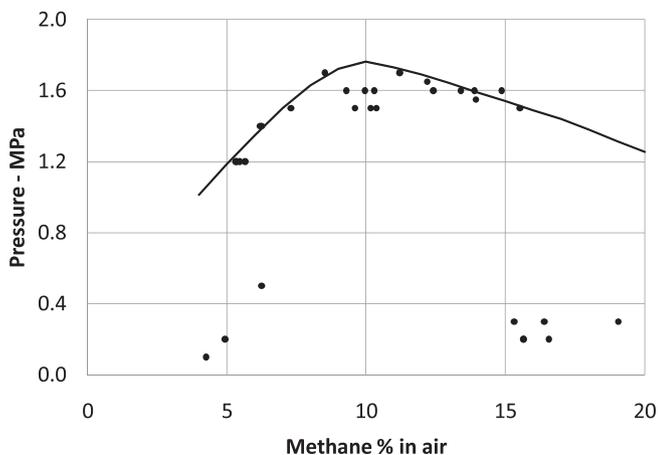


Fig. 7. Measured detonation pressure (data points) and CJ detonation pressure (solid line) versus methane in air.

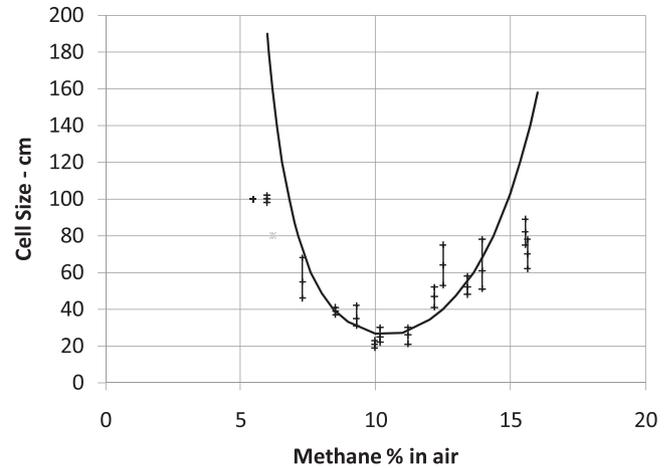


Fig. 8. Measured detonation cell size (low, high, and average data points) and cell size model (solid line) versus methane in air.

the visual estimates of CJ detonation pressure. The maximum CJ pressure, 1.77 MPa, occurs in about 10% methane in air and declines as the lower and upper limits are approached. The visual estimates of CJ detonation pressure near 10% methane in air appear low; however, no significance is attached to this apparent discrepancy considering the magnitude of our estimation error. Even at the limits of 5.3 and 15.5%, CJ pressures are still high at about 1.2 MPa and 1.5 MPa, respectively. Thus, a methane-air mixture develops variable but substantial pressures over the entire detonable range.

Fig. 8 presents measured detonation cell size as a function of mixture composition. Smokefoils were deployed on most tests from 27 onward. Tests in the range of 5.3–15.5% methane in air produced useful soot tracks and those outside the range did not. The number of cells in a foil ranged from 1 to 8, depending on the mixture. As seen in Fig. 8, the variability of the cell measurements is on the order of 30% due to the irregularity of the cell patterns for methane-air. Fig. 8 also includes the computed cell size based on a model by Gavrikov et al. (2000). Good agreement exists between the measured and theoretical cell sizes considering the high variability of the cell size measurements.

As shown by Dorofeev, Efimenko, and Kochurko (1992), the average time between spikes in the pressure-time records multiplied by the detonation speed provides a measure of the detonation cell width. Preliminary analysis of the pressure-time records shown in Fig. 3 yields the time between pressure spikes and an estimate of the cell size shown in Table 1. For a stoichiometric methane-air mixture (test number 27), the measured cell size measure using the pressure spike method is about 11 cm which is about 50% lower than that determined from Smokefoil observations. More detailed analysis using all the available pressure-time records is underway to obtain better estimates of the detonation cell size using the pressure spike method.

5. Conclusions

Using a direct method to initiate detonation in test mixtures, detonation is sustainable in methane in air mixtures over a range from 5.3 to about 15.5%. The range for the lower detonation limit is between 4.9 and 5.3% methane in air, and the range for the upper detonation limit is between 15.5 and 15.7% methane in air. This new range almost encompasses the entire normal combustion limits of 5–16% methane in air as reported by Cashdollar, Zlochower, Green, Thomas, and Hertzberg (2000). This range is larger than that observed by Wolanski et al. (1981), p. 1651, who reported a range of

7–13% for methane in air. The methane used in these experiments is natural gas containing about 97.5% methane and from 1.5 to 1.7% ethane; however, the presence of ethane is considered negligible. The detonation velocity varied from 1512 to 1863 m/s over this composition range, and the measured detonation velocities agreed with computed D_{CJ} values to within 2.5%. The measured pressure behind the peak shock pressure ranged from 1.2 to 1.7 MPa, and agreed with computed CJ pressure to within experimental error associated with this pressure measurement. Detonation cell patterns recorded with smokefoils showed irregular cell patterns for the methane–air test mixtures. Near-stoichiometric mixtures produced a detonation cell size of about 25 cm. With a 7% test mixture, cell size increased to about 55 cm, and with a 15.4% mixture, a single cell measuring 105 cm-wide by 170 cm-long was recorded.

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