ABSTRACT

The main purpose of this work is to investigate the influence of methane addition in methane-hydrogen-air mixture ($\varphi = 0.8 – 1.6$) on the critical conditions for transition to detonation in a 90-deg wedge corner. Similar to hydrogen-air mixtures investigated previously [1], methane-hydrogen-air mixtures results showed three ignition modes, weak ignition followed by deflagration with ignition delay time higher than 1 ms, strong ignition with instantaneous transition to detonation, and third with deflagrative ignition and delayed transition to detonation. Methane addition caused an increase in the range of 3.25 – 5.03% in the critical shock wave velocity necessary for transition to detonation for all mixtures considered. For example, in stoichiometric mixture with 5% methane in fuel (95% hydrogen in fuel) in air, the transition to detonation velocity was approx. 752 m/s (an increase of 37 m/s from hydrogen-air) corresponding to $M = 1.89$ (an increase of 0.14 from hydrogen-air) and 75.7% (an increase of 4.7% from hydrogen-air) of speed of sound in products. Also, similar to hydrogen-air mixture, the transition to detonation velocity increased for leaner and richer mixture. Moreover, it was observed that methane addition in general increased the pressure limit at the corner necessary for transition to detonation.

Keywords: hydrogen, methane, detonation, transition to detonation, shock focusing, shock reflection

1.0 INTRODUCTION

Hydrogen due to its properties is considered as one of the most promising energy carriers of the future. The common use of hydrogen in domestic, transport and energetics appliances will generate problems of safety nature as hydrogen is very easy to ignite, highly flammable in air and if the flammable H2-air cloud covers congested area the flame can go through DDT process (deflagration to detonation transition). Deflagration to detonation transition is not always predictable process, it is not easy to find and identify the exact time and location of the transition to detonation [2]. Understanding flame acceleration and DDT is also crucial for detonation propulsion systems (especially in PDE – Pulse Detonation Engines) and explosion safety [3,4]. Today’s industries place a high focus on safety measures, those measures are the reason why the wide utilization of hydrogen-enriched natural gas in the energy industry is limited by certain critical safety issues, such as the embrittlement problem of hydrogen in the transportation system [5], the enhancement of release and gas accumulation rates [3,4], and the reduction in the minimum ignition energy (MIE) [8]. Many papers investigated the flammability limits and explosion characteristics of methane-hydrogen mixtures [8,10-14]. Results showed that the methane addition to hydrogen reduced the flammability range, laminar burning velocity, and maximum pressure rise rate, in other words, methane addition reduces mixture reactivity and therefore reduces the risk of combustible gas explosions and its severity [9]. Also, the effect of methane addition on the detonation velocity and detonation cell size was investigated in a previous research [13], and results showed that this addition increases the detonation cell size and delays the deflagration to detonation transition in the tube. Another experimental study investigated the effect of methane addition to shock wave propagation, self-ignition and flame development of hydrogen-based mixtures [11]. Similar to [9], results show that methane addition can extremely reduce flame intensity and inhibit flame development. Moreover, results also show that the existence of methane in hydrogen-based mixtures has a great effect

EFFECT OF METHANE ADDITION ON TRANSITION TO
DETONATION IN HYDROGEN-AIR MIXTURES DUE TO SHOCK
WAVE FOCUSING IN A 90 – DEGREE CORNER

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on the self-ignition possibility, because methane addition reduce the shock wave overpressure and temperature.

In general, there are two main mechanisms responsible for transition to detonation which are shock reflection or shock focusing, and instabilities and mixing processes [14]. For the case of flame acceleration in a duct, where a shock wave is produced by this flame, transition to detonation becomes much more probable when the shock interacts with a corner or a concave wall that can produce shock focusing. As the wave reflects/focuses, the temperature and pressure locally can generate a hot spot which can cause the fuel-air mixture to reach its detonation threshold.

The main focus in this research is to investigate the transition to detonation due to shock focusing in a 90 – degree wedge corner and quantify the influence of methane addition to hydrogen on the transition phenomenon. Ignition features are one of the most important characteristics of fuels. Previous researchers have investigated the auto-ignition behavior in a shock tube, the existence of weak and strong ignition modes in different fuels, and the boundaries/limits of the modes [1,15–18]. Based on these published papers, three main types of ignition modes were observed for transition to detonation investigation, weak ignition (deflagration), strong ignition (detonation), and deflagrative ignition with delayed transition to detonation (delayed detonation). Results from a research that was done recently [18] show three ignition modes induced by 90 – degree shock focusing in a stoichiometric methane-oxygen mixture with 81.01 Vol.% of argon dilution and an initial pressure of 0.12 bara: peak local ignition mode (weak ignition) with an incident shock wave velocity of 689.18 m/s, boundary ignition mode (delayed transition to detonation) with an incident shock wave velocity of 768.05 m/s, and strong ignition mode (direct transition to detonation) with an incident shock wave velocity of 925.92 m/s. The three ignition modes are similar to the ones obtained for hydrogen-based mixtures in [1], which suggests that the three ignition modes will be observed for methane-hydrogen-air mixtures. One of the parameters that influence the ignition in transition to detonation due to shock focusing in hydrogen-based mixtures is the reflector shape, as it was investigated numerically and experimentally in previous works [15,19,20]. In general, results show the dependence of ignition modes on Mach number, hydrogen concentration and reflector shape.

To the best of author’s knowledge, there are no available experimental results regarding the effect of methane addition to hydrogen-based mixtures on the transition to detonation focusing in 90 – degree corner at an initial pressure of 1 bara. Therefore, the main aim of this research is to investigate the effect of methane addition on critical flow parameters necessary for transition to detonation focusing in 90 – degree corner in a set of 5% methane – 95% hydrogen in air mixtures at different equivalence ratios being initially at 1 bara. The main critical parameters that will be investigated are shock wave velocity, shock wave velocity relative to speed of sound in products, shock wave velocity relative to speed of sound in reactants, ignition delay time, and maximum pressure in the corner. It’s expected that the methane addition will cause the shock wave velocity needed for transition to detonation and maximum pressure in the corner to increase. The experimental results and the knowledge gained from this research would imply a practical solution for detonation prevention safety in applications using hydrogen-based mixtures and are beneficial to understanding the physics of shock wave focusing and its mechanism of shock wave focusing-induced detonation in this specific mixture.

2.0 EXPERIMENTAL SETUP AND PROCEDURE

Experimental work was conducted using the experimental stand described in [1]. The 1.5 m long detonation tube with a 0.11 × 0.11m cross-section shown in is used in these experiments. The initial part of the tube is filled with 6 × 6 mm mesh layers made of 1 mm diameter wire as shown in Figure 2; this mesh is used to accelerate the flame at short distance to fast deflagration regime with leading shock wave at velocity in the range of 600 - 900 m/s. Velocity at the exit of acceleration section can be controlled by number and order of mesh layers for a specific fuel - air mixture, and the generated shock wave is dependent on the flame acceleration process. The further side of the tube is closed with a lid equipped with 90 – deg wedge corner that is used to reflect the leading shock wave. Five pairs of sensors were used in these experiments, starting with four Piezoelectric pressure sensors (113B26 type with a
maximum pressure range of 6.7 MPa) and four in-house ion probes were placed side-on at the top wall of the tube. The last pair of pressure sensor (113B22 type with a maximum pressure range of 33.3 MPa) and ion probe were placed in the wedge tip 20 mm apart symmetrically to the tube main axis. All ion probes were supplied with DC battery. By postprocessing the data from all the sensors, velocity of the shock wave and flame, pressures, ignition delay time and ignition mode in the corner can be obtained, and that way one can quantify the conditions necessary for transition to detonation. Velocity of the shock wave is obtained by extrapolating linearly the velocity measured by the last three pressure sensors. This was necessary due to the fact that the leading shock wave velocity decreased along the tube length. Pressure in the corner is obtained from PS5. Finally, ignition delay time in the corner is obtained by comparing the time of arrival of the shock wave and flame occurrence in PS5 and IP5 sensors, respectively.

The mixtures tested were 5% methane - 95% hydrogen with air with equivalence ratios of 0.8, 1.0, 1.3 and 1.6. The mixtures were prepared by means of partial pressure method using digital manometer with 1 mbar accuracy and then stored horizontally for 24 h before the tests. At the beginning of each test, the tube was evacuated using a vacuum pump and it was checked for leaks. Afterwards it was filled with 1 bara of the fuel-air mixture and was left to stabilize for 5 min before ignition. The pressure sensors and

Figure 1. Experimental setup [1]

Figure 2. Acceleration section [1]
ion probes signals were recorded with 2 MHz frequency after the ignition using data acquisition system (DAS) that was activated by a time sequencer (to synchronize ignition and DAS recording start time). The initial pressure and temperature for all experiments conducted for this research were 1 bara and 298 ± 3 K, respectively. Finally, the experimental procedure used in this research is the same as the one used by Rudy for hydrogen-air mixtures [1].

3.0 RESULTS

Total number of experiments conducted was 70 and the results obtained showed three types of ignition modes similar to those for hydrogen-air mixtures [1]. The first mode is deflagrative ignition in the corner and it was observed for lower velocities. This ignition mode was characterized by an ignition delay time higher than 1 µs and a single pressure peak obtained from PS5 with a pressure value lower than observed in other ignition modes. The peak pressure value decreases after the reflection to a value of approximately 1 MPa. The second mode is direct transition to detonation in the corner and it was observed for higher velocities. For direct transition to detonation, the ignition delay time was less than 1 µs and higher maximum pressure from PS5 was obtained. Finally, the third observed mode is deflagrative ignition with delayed transition to detonation and it was characterized by two pressure peaks recorded by PS5 and an ignition delay time higher than 1 µs.

Figure 3 shows examples of sensors histories for all recorded ignition modes. Starting with the first graph (left) which shows a deflagration with a maximum pressure of approx. 6 MPa and an ignition delay time between the pressure from PS5 and signal from IP5. The middle graph shows a detonation case with a maximum pressure of approx. 8.5 MPa and the PS5 and IP5 signals activates simultaneously for this case. Finally, the third graph shows a delayed detonation case where PS5 recorded two pressure peaks, and second peak has a higher value than the first one, which is similar to the data obtained for hydrogen-air mixtures [1].

Figure 4 presents the ignition delay time calculated from PS5 and IP5 as a function of reflection velocity at different equivalence ratios. Points from hydrogen-air are added for reference. For methane-hydrogen-air mixtures, IDT for transition to detonation cases is given as 1 µs for reference only, however, the real values for these tests are in the range on 0 - 0.5 µs which is within the measuring accuracy of the data acquisition system. On the other hand, for higher equivalence ratios (φ = 1.28, 1.6), it has been observed that methane addition causes a higher ignition delay time than 1 µs for detonation cases, and that is probably due to the change in the reactivity of hydrogen-air mixture after doping 5%.
of methane. Delayed detonation cases had an IDT in the range of 2 - 5 µs. Comparing all graphs presented in Figure 4 with the results obtained for hydrogen-air mixtures [1], it is visible that methane addition to hydrogen increased the transition to detonation shock velocity limits for all equivalence ratios. One can also see that most delayed detonation cases occurred between the deflagration and detonation ignition modes even with the methane addition and the general behavior of IDT as a function of shock wave velocity is similar to the one obtained for hydrogen-air mixtures.

Figure 4. Ignition delay time in the corner as a function of reflection velocity for mixtures: $\phi=0.8$ (top left), $\phi=1$ (top right), $\phi=1.28$ (bottom left), and $\phi=1.6$ (bottom right).

Figure 5 shows the limits for shock wave velocity, velocity relative to speed of sound in reactants and velocity relative to speed of sound in products limits for transition to detonations. The red dashed line represents the transition to detonation limits for 5% methane – 95% hydrogen in air mixtures at different equivalence ratios, and the black solid line represents the transition to detonation limits for hydrogen-air mixtures at different equivalence ratios obtained in [1]. As predicted, methane addition to hydrogen-air causes transition to detonation limits to shift up. However, the behavior of the mixture at different equivalence ratios is similar to hydrogen-air where transition to detonation limit is characteristic U shape and the lowest transition to detonation velocity is achieved at an equivalence ratio of 1.0. Also, for leaner and richer mixtures higher velocities are necessary for transition to detonation. The transition velocity for stoichiometric mixture was approx. 752 m/s (an increase of 37 m/s from hydrogen-air) corresponding...
to $M = 1.89$ (an increase of 0.14 from hydrogen-air) and 75.7\% (an increase of 4.7\% from hydrogen-air) of speed of sound in products. In other words, the shock velocity necessary for transition to detonation is higher for methane-hydrogen-mixtures. For the other equivalence ratios, the increase is presented in the figure next to each limit point. Finally, the percentage of increase was calculated as the absolute difference between methane-air mixture and hydrogen-air mixture divided by the value for hydrogen-air mixture.

Figure 5. Limits for transition to detonation in 90-deg wedge corner: shock wave velocity (left), velocity relative to speed of sound in reactants $a_R$ (center), velocity relative to speed of sound in combustion products $a_P$ (right). $H_2$ + air limit taken from [1]

Figure 6 presents the maximum pressure values recorded by PS5 for all the tests. The dashed red line shows the minimum pressure in the corner that was recorded for successful transition to detonation in methane - hydrogen - air mixtures, where the solid black one represents the same limit for hydrogen – air [1]. Similar to shock wave velocity, methane addition to hydrogen increased the value of pressure at the corner necessary for transition to detonation. This increase in PS5 max in the corner is in range of 0.1 – 1MPa depending on the equivalence ratio. Also, similar to hydrogen-air mixtures, the value of corner pressure necessary for transition to detonation increases for more lean and rich mixtures. Mainly, methane addition caused an increase in the corner pressure measured by PS5, comparing to the values obtained from hydrogen-air mixtures [1]. One can also conclude that the larger increase in the maximum pressure necessary for transition to detonation was recorded for rich mixture of 1.6 equivalence ratio.

Figure 6. Maximum pressure recorded by PS5. $H_2$ + air limit taken from [1]
4.0 CONCLUSIONS

The research presented in this paper was focused on the influence of methane addition to hydrogen-air mixtures on critical flow parameters necessary for transition to detonation due to focusing in 90-degree wedge corner. Investigated critical parameters are shock wave velocity, shock wave velocity relative to speed of sound in products, shock wave velocity relative to speed of sound in reactants, ignition delay time, and maximum pressure in the corner. 5% methane addition to hydrogen-air mixtures with a range of equivalence ratios between 0.8 and 1.6 being initially at 1 bara were considered and following facts might be concluded:

➢ Three ignition modes (deflagration, detonation, delayed detonation) were visible for methane-hydrogen-air mixture. However, unlike hydrogen-air mixtures where delayed detonation cases occurred for rich mixtures (φ = 1.28, 1.6), delayed detonation cases occurred for all equivalence ratios for methane-hydrogen-air mixtures.

➢ Methane addition to hydrogen-air mixtures causes the shock velocity limit necessary for transition to detonation to increase. $V_S$ limit for stoichiometric mixture was approx. 752 m/s (an increase of 37 m/s from hydrogen-air) corresponding to $M = 1.89$ (an increase of 0.14 from hydrogen-air) and 75.7% (an increase of 4.7% from hydrogen-air) of speed of sound in products.

➢ The maximum pressure values recorded by PS5 in case of detonation are in the range of 8.1 – 12.8 MPa for 0.8 - 1.6 equivalence ratio range. Similar to shock wave velocity, methane addition to hydrogen increased the value of pressure in the corner necessary for transition to detonation, especially for rich mixtures.

REFERENCES


