EXPERIMENTAL DETERMINATION OF CRITICAL CONDITIONS FOR HYDROGEN-AIR DETONATION PROPAGATION IN PARTIALLY CONFINED GEOMETRY

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ABSTRACT

An experimental investigation was performed to determine critical semi-open channel height (h^*) in which hydrogen-air detonation may propagate. Three types of gaseous mixture composition were used: 25%, 29.6% and 40% of hydrogen in air. Experimental setup was based on rectangular (0.11 x 0.11 x 2 m) test channel equipped with acceleration section (0.11 x 0.11 x 1 m) and data acquisition system with 5 pairs of pressure transducers and ion probes. Different channel heights h in range of 15 - 40 mm were used in the test channel. The critical height h^* was defined for each investigated mixture. Additionally, sooted plates technique was used to determine detonation cell sizes λ and their relationship to h^* . The results showed that detonation in stoichiometric H₂-air mixture may propagate in semi-open channel only when the channel height is equal to or exceeds 3λ . For less reactive mixtures this critical relation reaches 3.1λ or 3.6λ for mixtures with 25% and 40% of hydrogen in air respectively.

1.0 INTRODUCTION

Combustion behavior of hydrogen-air mixtures in partially confined geometries is very important from the practical point of view. Hydrogen leakage in a confinement may lead to creation of a flammable mixture within explosion or detonation limits at the top of the room. Such a phenomena might occur in nuclear reactors, chemical plants or fuel cell containers. The effect of geometry on detonation for uniform mixtures of hydrogen and air has been investigated by many researchers - CALTECH database contains many of the data obtained [1] including critical tube diameter and minimum tube diameter. It has been established that for smooth channels the detonation propagates when the critical tube diameter d^* is $d^* = 3 \lambda$ where λ is an average detonation cell size [2]. The most probable scenario with hydrogen release in confinement is the creation of non-uniform hydrogen-air mixture. Such release processes were investigated by numerous researchers [3-7]. A significant investigation was performed by Whitehouse et al. [5] where propagation of vertical flame front was studied in uniform and non-uniform, stratified hydrogen-air mixtures in a vertical 10.7 m³ cylinder with ignition sources located at the top and the bottom of the cylinder. It was reported that for the top ignition velocity of the flame and combustion pressure were significantly higher for mixtures with concentration gradient than for homogeneous mixture. Furthermore the flame was propagating even in a mixture with average hydrogen concentration below the downward flame propagation limit - the local hydrogen concentration in gradients at the top igniter was above this limit what allowed for combustion [5]. There was also carried out some experiments for semi-confined hydrogen-oxygen mixtures [8]. It was established that for stoichiometric hydrogen-oxygen mixture bounded by nitrogen the detonation propagates in the channel with height at least equal to 5.08 mm.

Obstacles located along the path of the propagating flame in a confined geometry may cause flame acceleration and in the worst case scenario deflagration to detonation transition (DDT) [6-7]. This phenomenon was widely studied for well premixed and homogenous gas mixtures, however there is

still little experimental data on non-uniform hydrogen-air mixtures. In the work of Lamoureux et al. [9] there was presented a comparison between flame front acceleration in non-uniform and homogenous hydrogen air mixtures. Moreover, the authors investigated the effect of the obstacles on the flame speed. There were two very important conclusions from this work. The first one is that the lean hydrogen air flame can be strongly accelerated if the expansion ratio is greater than 3.75 corresponding to a 10.5 % hydrogen-air mixture. The second one is that the maximum flame velocity achieved in a non-uniform mixture with negative gradient and uniform mixture was the same. There have also been conducted several experiments on the deflagration to detonation transition (DDT) in semi-open, obstructed geometries both for uniform and non-uniform hydrogen-air mixtures [10-12]. Kuznetsov et al. [10] performed experimental studies about DDT of homogenous hydrogen-air mixture in a partially confined, obstructed channel. It was observed that detonation onset depends on the layer thickness which must be 13-14 times greater than detonation cell width λ . It was also established that obstacle orifice size is a strong factor for sustained detonation - for blockage ratio equal to 0.5 detonation would not propagate if the orifice size is less than 3λ . These conclusions were confirmed in similar experiments performed by Grune et. al [11] and showed that the channel width has little influence on flame acceleration or DDT and the main parameters responsible for these phenomena are gas layer thickness and mixture reactivity. For non-uniform hydrogen-air mixtures a significant attribute having an impact on detonation propagation is concentration gradient [11-14]. Grune et al. [12] reported that for maximum hydrogen concentration gradient at the top of the channel smaller than 16% and the gradient slope equal to $\sim 0.7\%$ /cm the detonation will decay. In the work [14] it was found that for the gradient lower than 0.2 H₂/cm the critical conditions for effective flame acceleration to sonic speed are the same as for uniform mixtures. It was also reported that steeper hydrogen gradient results in more reactive mixtures and higher hydrogen concentration at the top of the channel which allows the flame to accelerate to sonic speed.

Despite these investigations, the critical conditions of uniform hydrogen-air mixture detonation transmission in semi-confined, unobstructed geometry is not widely described. Some experimental large scale investigations were performed by Rudy et al. [15]. In this case the critical thickness h^* of stoichiometric hydrogen-air mixture was approximately equal to 3 cm which confirms the relation with detonation cell size $h^*/\lambda \approx 3.4$. That condition was confirmed by Gaathaug et. al [16]. In their work it was reported that for 30% H₂ in air mixture the detonation propagates if $h^*/\lambda = 3.7$. For stoichiometric hydrogen-oxygen mixture reported by Dabora [8] h^*/λ ratio is in range of 2.4 - 3.6 depending on λ size used [1]. The aim of this work was to precisely determine the critical layer thickness h^* and h^*/λ ratio for detonation propagation in semi-confined, non-obstructed channel filled with uniform, stoichiometric and non-stoichiometric hydrogen-air mixtures.

2.0 EXPERIMENTAL STAND AND PROCEDURE

2.1 Experimental stand

Experiments were conducted in a channel consisting of acceleration section (0.11x0.11x1 m) and test section (0.11x0.11x2 m). The detonation onset was initiated in 0.35 m long turbulence generator made of 6x6 mm metal grid layers located inside the acceleration section. Detonation wave moved along the remaining part of the acceleration section and then propagated in the test section which was a rectangular channel divided into two volumes by a thin 3 µm plastic film (HDPE). Top volume had dimensions (HxWxL) hx0.09x2 m where height h was varied by a moveable channel bar. Lower volume (0.045x0.09x2 m) was filled with air. In order to measure pressure and flame occurrence along the channel five pairs of pressure transducers and ionization probes were placed at the top wall of the tube. One additional pressure transducer was used as a trigger for data acquisition system. For detonation cell size measurements a sooted plate was placed at the end of the test section. Cell size measurements were done in a smooth tube. The geometry of the experimental facility and the cross-sections of the tubes are presented in Fig. 1. Mixtures investigated were prepared in cylinders by partial pressure method and stored horizontally for at least 24 hours.



Figure 1. Experimental setup scheme. Sensors distances are related to ignition point. Red colorflammable mixture, blue color – air channel.

2.2 Experimental procedure

Before each experiment open side of the channel was covered with a 3 μ m plastic film stretched on the moveable C-shape bar. The air was evacuated by a vacuum pump and the whole tube was filled with hydrogen-air mixture. Then hydrogen-air mixture located in volume confined by a plastic film was replaced by air flowing from one side of the test section channel towards the other. This process was continued until the oxygen concentration sensor placed in the outlet pipe indicated 20.9% value. Next, to diminish possible diffusion through the plastic film acceleration section and top channel in test section were flushed by fresh hydrogen-air mixture for approximately 3 min. The initial conditions of gases filling the channel were ambient pressure (~0.1 MPa) and temperature (298±3 K). Three hydrogen-air mixtures were used in the experiments: 25% 29.6% and 40% of hydrogen in air. The ignition process was started by a spark plug placed in the middle of the acceleration section flange.

3.0 Results

3.1 Detonation in smooth channel

Prior to experiments in semi-confined channels, each mixture was tested in an empty tube (0.11x0.11x3 m) in order to check the detonation velocity and cell size measurements. Sooted aluminium plate was installed at the end of the tube. Figure 2 presents the post-processed image of the detonation cells footprints in the mixture with 29.6% of hydrogen in air. As the recorded structure was not regular at least 120 cells were measured for each mixture to get the median and average cell sizes. Cell size measurements were done with slide calliper and 0.5 mm accuracy. Cell sizes bar charts for each mixture are presented in Fig. 3. Comparison between referenced works [17-20] and cell sizes obtained shows good agreement with the results of Ciccarelli et al. [18,20]. Figure 4 presents typical pressure and ion probes traces for mixture with 25% H₂ in air (Fig. 4 left) and the detonation mean velocities between sensors obtained in smooth tube in comparison with ideal CJ values (Fig. 4 right). The experimentally obtained velocities are only slightly below the ideal CJ values.



Figure 2. Cell size image for mixture with 29.6% of hydrogen in air. Arrow shows the propagation direction. Image post-processed.



Figure 3. Cell size histograms for mixtures with 25%, 29.6% and 40% of hydrogen in air.



Figure 4. Comparison between cell size measurements and referenced works [17-20].



Figure 4. Example of pressure gauges and ion probes signals for 25% H₂ in air mixture in smooth tube (left) and comparison between velocities obtained in smooth tube for all mixtures considered and theoretical CJ-calculations (right).

2.3 Detonation in partially confined channels

Experiments in partially confined channels were prepared for hydrogen concentrations in the mixture equal to 25%, 29.6% and 40%. The height of the channel was variable in range of 15 - 45 mm with 5 or 2.5 mm step. Figure 5 shows sensors recordings for channel heights equal to 22.5 mm (Fig. 5 left) and 17.5 mm (Fig. 5 right) in stoichiometric H₂-air mixture. Left diagram in Fig. 5 shows case with detonation propagating up to the end of the test section. First sensor indicates pressure curve shape similar to presented in the case of smooth tube as the first sensor pair is placed in the smooth part of the acceleration section. The following pairs of sensors are placed in the test section thus their indications are considerably different. Shortly after the pressure peak indicating detonation front a sudden pressure drop to around 0.25 MPa occurs due to the gas expansion into the air placed in lower part of the test section. In case of detonation failure (Fig. 5. right) only first pair of sensors in the test section indicated detonation front, the following pairs of sensors show pressure and flame front decoupling and the velocities recorded oscillate around the products sound speed (Fig. 6.) In general, for all of the mixtures considered the scheme of detonation failure is very similar.



Figure 5. Detonation propagating up to the end of test section in 22.5 mm channel (left) and detonation failure in 17.5 mm channel shortly after passing first pair of sensors in test section (right).

The differences are mainly in that the detonation fails before or shortly after reaching the first pair of sensors in the test section. In all of the experiments with 25% and 29.6 H₂-air mixture detonation reached first pair of sensors in the test section (placed 0.17 m downstream the test section). For 40% H_2 in air detonation failed before reaching the first pair of sensors in the test section. That is visible in Fig. 6 as lower velocity values for a second set of points (L = 0.954 m). Moreover, with decreasing the channel height the detonation failure takes place faster in the test section as the velocities in second set of points decrease with decreasing channel height. Another conclusion is that even for cases with detonation propagating up to the end of the tube velocity deficiency may be observed. This deficiency increases as the channel height decreases and the channel length increases. The detonation velocity deficiency was observed by the Dabora [8] who concluded that the expansion of the products behind the detonation front decreases the velocity and 8-10% decrease is necessary to quench the detonation. In this research maximum observed detonation velocity deficiency related to C-J velocity is equal to 6.2%, 7.4% and 8.2% for mixtures with 25% 29.6% and 40% H_2 in air respectively. As the test section length is limited to 2 m we may not exclude that the detonation might fail in longer tube as the velocity slightly but progressively decreases along the channel. Based on the experiments performed critical partially confined channel heights were specified for each mixture investigated. These values are summarized in Table 1. Additionally, the h^*/λ relations are specified for both λ values: median (λ_{med}) and average (λ_{ave}) . For stoichiometric H₂-air mixture the h^*/λ relation is very close to 3 which slightly limits the values available in the literature: 3-4 [15], 3.7 [16] and 2.4-3.6 [8]. Additionally, the h^*/λ relation seems to increase as the mixture becomes less reactive. This critical relation presented in Fig. 7. seems to be linear under conditions considered.



Figure 6. Velocities for various semi-open channel heights and mixtures. Sketch at the lower part of the figure presents the schematic cross-section of the tubes.

Table 1. Critical partially confined channel heights for which detonation may propagate in various H_2 -air mixtures with corresponding h/λ relations.

H ₂ concentration in air	25%	29.6 %	40%
Critical channel height h* [mm]	40	22.5	25
λ_{med} [mm]	11	8	8
λ_{ave} [mm]	11.12	7.69	8.24
h^*/λ_{med} [-]	3.64	2.81	3.13
h^*/λ_{ave} [-]	3.6	2.93	3.03



Figure 7. Relation between h and h/λ for all mixtures considered; dotted line connects h^*/λ_{ave} values.

3.0 CONCLUSIONS

Experiments with detonations in stoichiometric (29.6 % H₂) and non-stoichiometric (25% and 40% H₂) hydrogen-air mixtures were performed in a 3 m long channel with 2 m long test section with separated partially confined channel. The aim of the study was to precisely specify critical semi-open channel height h^* in which detonation may propagate. By decreasing the semi-open channel height detonation velocity deficiency was observed. This deficiency progressively increased along the test channel reaching value of 8.2 % relating to ideal CJ velocity. For channels with height lower than critical one, detonation failure was observed and velocities following decreased to the value of products sound speed. Additionally, the investigation included detonation cell size measurements for the investigated mixtures in order to specify h^*/λ relation. The results obtained showed that the critical semi-open channel height is equal to 40, 22.5 and 25 mm for mixtures with 25%, 29.6% and 40% H_2 in air respectively. Corresponding h^*/λ ratio is in range of 2.8 - 3.64 depending on the mixture and method of λ size determination. In general, the h^*/λ ratio is very close to 3 for stoichiometric H₂-air mixture and increases as the mixture becomes less reactive. The results obtained limit the values available in the literature for stoichiometric mixture: 3-4 [15], 3.7 [16], 2.4-3.6 [8] and extends the critical relation h^*/λ to non-stoichiometric mixtures. The h^*/λ relation increases linearly as the channel height increases. However, due to the geometrical limitations of the experimental stand, additional experiments should be performed in larger scale to cover wider range of hydrogen concentrations in air and confirm the observed linear dependence.

ACKNOWLEDGEMENTS

Funding for this investigation was provided by The Dean of Faculty of Power and Aeronautical Engineering of Warsaw University of Technology, grant no. 504/01503/1131/42.

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