

Deflagration to Detonation Transition in Binary Fuel H₂/CH₄ with Air Mixtures

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1 Introduction

The future rarefaction of oil source and concerns about global warming makes hydrogen the most valuable candidate as fuel replacement. But to be effective, the gas needs an appropriate distribution network all over countries. The use of the already existing natural gas (methane) distribution network can be a way to fulfill this objective and the distribution of H₂/CH₄ mixture is envisaged. Thus, the knowledge of combustion and detonation characteristics of hydrogen/methane - Air mixtures (with a volume ratio of H₂ in the fuel higher than 0.5) is needed to permit correct sizing of distribution devices. Moreover, security application needs data on the possibility of self explosion with air in the case of high pressure fuel release or in the case of low energy initiation (electric spark) of accidental hazardous mixtures and deflagration to detonation transition (DDT).

Few data are available in the literature on the deflagration to detonation transition of H₂ - Air and CH₄ - Air mixtures. Sorin et al. [1] present data on DDT of H₂ - Air mixture at ambient condition in a 26 mm i.d. tube containing a spiral of blockage ratio (BR) of 0.5. They found that run up distance for detonation onset L_{DDT} is approximately equal to 37 cm. In addition Kuznetsov et al. [2] show that for stoichiometric CH₄ - Air mixtures at ambient conditions, the run up distance is equal to 12m, that is 32 times higher than H₂ - Air L_{DDT} . This length was obtained in a tube of 520 mm i.d. and with orifice plate obstacles of BR = 0.3. Despite the difference of tube diameter, close to limit value of diameter related to criterium of existence of detonation in a tube [3], we can see that H₂ and CH₄ represent opposite detonation sensibility with air. No data concerning detonability of binary mixture of H₂/CH₄ with air are available.

The aim of the present study is to obtain deflagration to detonation transition data (L_{DDT}) of H₂/CH₄ - Air mixtures as a function of molar fraction x of H₂ into binary H₂/CH₄ fuel mixture, equivalent ratio Φ of the mixture and initial pressure P_0 at ambient temperature ($T_0 = 293$ K). A tentative of correlation between L_{DDT} and detonation cell size λ is done.

2 Experimental Device

The DDT data were obtained in a 6 m long stainless steel tube divided in a 2 m long section with 19 pressure transducer locations (separated by 100 mm) and a 4 m long tube with a transducer located

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at the end of the tube (cf. Fig. 1). The ignition is obtained using an automotive spark plug with energy deposition of around 15 mJ. In order to enhance the detonation transition, a 2.8 m long schelkin spiral, with a blockage ratio $BR = 1 - (d/\varnothing)^2 = 0.5$ and a pitch equal to the diameter $\varnothing = 52$ mm, is installed in the tube. For each experimental condition, at least 7 shots were done. The L_{DDT} corresponds to the average length measured. The pressure transducers used (KISTLER 603B) has a response time of $1\mu s$, suitable to shock and detonation measurements.

The mixtures used for the study follows the formula: $\Phi[xH_2 + (1-x)CH_4] + (2-1.5x)\text{air}$ with Φ the equivalence ratio and x the ratio of H₂ in binary H₂/CH₄ fuel mixture. The initial pressure P_0 of the mixture can be varied from 0.2 to 2 bar. During the study, x and Φ were varied from 1 to values for which DDT was not observed.

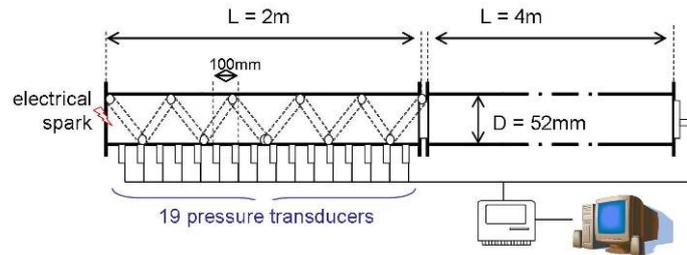


Figure 1. Experimental device for L_{DDT} measurement.

3 Results and Interpretation

During the DDT process, the propagation of the flame in the tube induces a precursor shock whose time of arrival at different position is determined from the pressure signal. The $L-t$ diagram can be drawn and the local velocity of the wave (i.e. between two transducers) can be deduced. The Figure 2 shows an example of the evolution of the wave velocity along the tube for $x = 0.9$ and $\Phi = 1$ mixture. Points indicated in the figure are the local velocity of each shot, the red and blue lines are respectively the average experimental self sustained (D_{spiral}) detonation velocity and the Chapman-Jouguet (D_{CJ}) detonation velocity. We can notice, first, a strong increase of shock velocity up to $L \sim 0.35$ m and, after an overshoot, a plateau. Then, after a new acceleration up to a velocity higher or of the same order of D_{CJ} , DDT occurs. $L = L_{DDT} \sim 0.85$ m corresponds to the location where $D = D_{\text{spiral}}$ is first reached in the $D-L$ diagram.

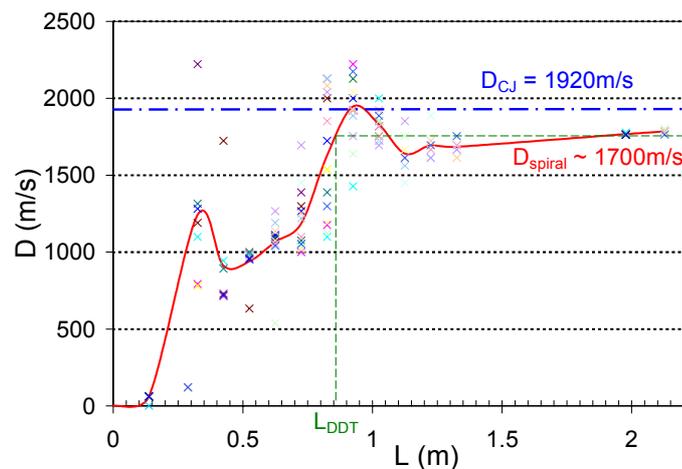


Figure 2. Evolution of shock and detonation velocity in the 2 m long transducer section with spiral for H₂/CH₄ - Air mixture with $x = 0.9$ and $\Phi = 1$.

The evolution seen on Figure 2 is typical of DDT in spiral section as observed in [1] for $\varnothing = 26$ mm i.d. tube. This evolution can be detailed in three phases: (i) a low velocity flame propagation (due to laminar flame after the ignition), (ii) a rapid acceleration to fast deflagration (around 1100 - 1200 m/s) and (iii) a transition to detonation identified by a resulting overdriven detonation peak. As noticed in [1], the detonation velocity in the spiral section is lower than D_{CJ} (D_{spiral} around $0.87 \cdot D_{CJ}$ for $d/\lambda \sim 2$), due to the spiral momentum losses on the detonation propagation regime.

The experimental results of L_{DDT} are shown in Figure 3. Figure 3-left summarizes the dependency of L_{DDT} on initial pressure P_0 . L_{DDT} seems to vary like a power -0.8 of initial pressure ($P_0^{-0.8}$) for the different mixtures studied ($0.8 < x < 1$ and $0.7 < \Phi < 2$). Figure 3-right displays the evolution of L_{DDT} with equivalence ratio Φ for different x at $P_0 = 1$ bar. $L_{DDT}(\Phi)$ is a U-shape curve with a minimum at $\Phi = 1.2 - 1.4$ rich mixture. L_{DDT} ranges from 0.1 to 1.3 - 1.4 m with the exception of $x = 0.9$ and $\Phi = 1.8$ where L_{DDT} is about 2 m, DDT being not observed systematically in this case. For $x < 0.7$, DDT doesn't occur at all, so this configuration allows the onset of detonation only for $x \geq 0.7$.

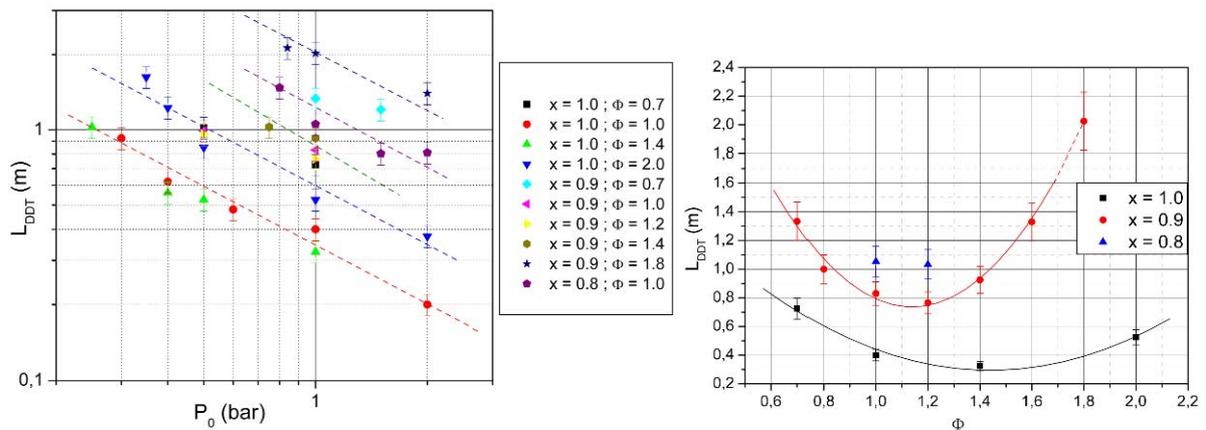


Figure 3. L_{DDT} in H₂/CH₄ - Air mixtures in 52 mm i.d. tube with schelkin spiral. Left: as a function of initial pressure P_0 . Right: as a function of equivalence ratio Φ at $P_0 = 1$ bar.

The Figure 4 represents L_{DDT} as a function of detonation cell size λ for the mixtures studied. The dependency of λ to equivalence ratio Φ at ambient conditions ($P_0 = 1$ bar, $T_0 = 293$ K) for various x (cf. Fig. 5) is provided from reference [4]. Cell sizes at initial pressures different from $P_0 = 1$ bar are deduced from Fig.5 assuming the relationship $\lambda \sim P_0^{-1.15}$ [5]. Varying x from 1 to 0.8 makes the cell size to increase from 10 to 30 mm at stoichiometric ratio. The same influence is seen for the transition to detonation, L_{DDT} varies from 0.4 to 1.1 m. So the introduction of a weak volume of CH₄ in the mixture substantially increases the chemical induction time and therefore decreases the detonability. We remark from Fig. 4 that, for $x \geq 0.8$, a linear evolution of L_{DDT} with cell size as far as $\lambda \leq 3$ cm. More precisely it seems to correspond to $L_{DDT} \sim 30 - 40\lambda$. Particularly for $P_0 = 1$ bar, L_{DDT} is close to 40λ . A small amount of CH₄ (up to 20%) added in the fuel does not change the correlation $L_{DDT}(\lambda)$ obtained with H₂ - Air mixture except for $x = 0.9$ and $\Phi = 1.8$ where the previous correlation fails (cf. Fig.4). Thus, because L_{DDT}/λ ratio are the same for obstacles laden tubes of different diameters ($\varnothing = 26$ mm [1] and 52 mm), it seems that the reactivity of the mixture and the ability of auto-ignition behind a shock wave (i.e., induction length) influences significantly the DDT run up distance observed in H₂/CH₄ - Air mixtures.

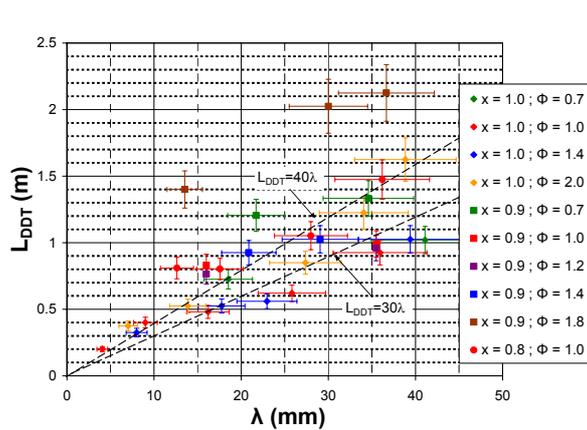


Figure 4. L_{DDT} as a function of detonation cell size λ in H_2/CH_4 - Air mixtures.

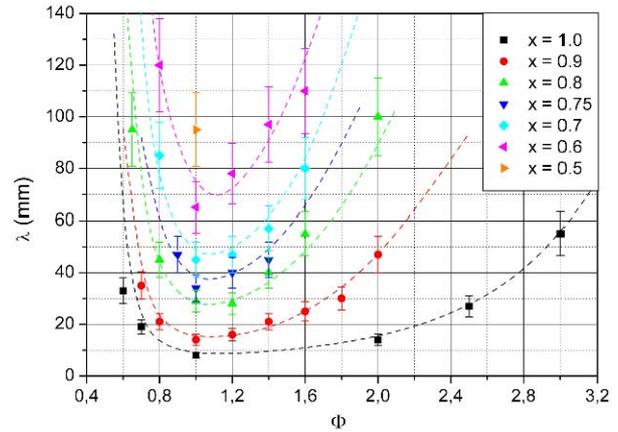


Figure 5. Cell size λ for H_2/CH_4 - Air mixture as a function of equivalence ratio Φ and H_2 volume ratio x in fuel at $P_0 = 1$ bar, $T_0 = 293$ K obtained in 52 106 mm i.d. tube (Bozier et al. [4]).

4 Conclusion

Deflagration to detonation transition (DDT) was studied in H_2/CH_4 - Air mixtures in a $\varnothing = 52$ mm i.d. tube with spiral of blockage ratio of 0.5. Different H_2 volume ratio x in binary fuel H_2/CH_4 , equivalence ratio Φ and initial pressure P_0 are considered. L_{DDT} was determined from velocity-distance diagram. It was found that the introduction of a small amount of CH_4 ($x \geq 0.8$, i.e. ratio of CH_4 less than 20% in the fuel) desensitizes the mixture compared to H_2 - Air mixture and increases the run-up distance L_{DDT} to obtain transition to detonation. The fuel binary mixtures studied behaves like H_2 - Air mixture, i.e., the length of transition obeys the linear law $L_{DDT} \sim 30-40\lambda$. This correlation indicates that, in certain conditions (obstacle laden tube), the deflagration to detonation transition depends strongly on chemical kinetics behind a shock wave propagating in the choking regime conditions of the mixture.

References

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