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Cellular structure of detonation wave in hydrogen-methane-air mixtures

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Abstract

The paper reports on an experimental study of the cellular structure of detonation waves in hydrogen-methane-air mixtures. Experiments were performed in a 6-m circular cross section tube with inner diameter of 140 mm (full diameter of 170 mm) and a 0.8-m driver section tube with inner diameter of 90 mm. The initial conditions of stoichiometric hydrogen-methane-air mixtures were 1 atm and 293 K with varoius hydrogen content. The average detonation cell size for stoichiometric hydrogen-methane-air mixtures obtained by the smoked foil technique during the experiments was calculated with the 2D Fourier transform. Using the Cantera program, the detonation cell width was computed as a function of initial pressure and molar methane fraction in fuel for a stoichiometric methane-hydrogen-air mixture at initial temperature 295 K. The results of the computations were compared against the experimental results.

Keywords: Detonation, Cell Size, Cellular Structure, Hydrogen, Methane

1. Introduction

An increasing interest is being shown in methane, a main component of natural gas (ca 90%), as an alternative fuel. This is motivated by a general desire to reduce exhaust emissions. Whereas natural gas seems to be the cleanest of all the fossil fuels [1], hydrogen too has been investigated as a single fuel in many experimental studies. Unfortunately, hydrogen is very difficult to store, handle and transport on the scale required by industry due to its low density, extremely low boiling point (at 1 atm: -253°C or 20 K) and wide range of flammability limits (from 4% to 75%). That is why a hydrogen-methane mixture is used in a variety of combustion devices such as ICE engines (e.g. HCCI), gas turbines, various types of burners with premixed and diffusion flames and in public pipeline transport. Another factor militating against hydrogen is the virtual absence of infrastructure anywhere in the world for transporting hydrogen gas. The construction of extensive pipeline networks for hydrogen gas comparable to the existing natural gas pipeline networks would require massive investment. Although it is conceptually possible to provide financing for dedicated hydrogen pipelines, construction of extensive hydrogen networks is not expected in the short term. An alternative that at least merits discussion is the partial use of the existing natural gas pipelines to transport hydrogen. Adding hydrogen to a methane-air mixture increases the flammability limits and stability ranges of the flame. It also decreases autoignition delay times and emissions of exhaust gases. Unfortunately, the presence of hydrogen in flammable mixtures dramatically increases the risk of explosion and in particular

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cases, especially during pipeline transport, of detonation.

Adding hydrogen to a methane-air mixture increases the flammability limits and stability ranges of flame and the operating conditions can then be shifted towards leaner conditions [1–3]. Hydrogen addition decreases autoignition delay times and emissions of exhaust gases. Furthermore, it causes of reaction zone shape and creates better transport conditions for methane. The presence of hydrogen in combustible mixtures greatly increases the risk of explosion and especially detonation due to the high overpressures associated with a detonation wave. Although a detonation wave can be initiated directly by the deposition of a large amount of energy in a very small volume of the mixture, typically such ignition sources are not present in most industrial settings. Explosions almost universally start by the ignition of a flame from either an electrical spark or the autoignition of the mixture in contact with a superheated surface. Under certain conditions a flame can accelerate and undergo transition to a detonation wave, which is also known as the DDT process.

Due to the high energy required for detonation initiation directly in fuel-air mixtures, the worst case scenario in industrial conditions can easily be avoided in the open air. Sometimes a spontaneous detonation can be associated with DDT and direct initiation caused by a turbulent jet of combustion products.

Hydrogen and methane are currently the most important energy carriers, but their use is determined by safety conditions. Generally speaking, hydrogen is much more dangerous than methane, hence the latter is widely used and considered thus far as publicly acceptable [4] in respect of explosion and detonation hazard.

Chaumeix et al. [5] validated a detailed kinetic mechanism for the oxidation of hydrogen-methane-air mixtures in detonation waves. They performed a series of experiments on auto-ignition delay times using a shock tube technique coupled with emission spectrometry for $H_2/CH_4/O_2$ mixtures highly diluted in argon. The CH_4/H_2 ratio was varied from 0 to 4 and the equivalence ratio from 0.4 to 1. The temperature range was from 1,250 to 2,000 K and the pressure behind reflected shock waves was between 0.15 and 1.6 MPa. They also proposed a correlation

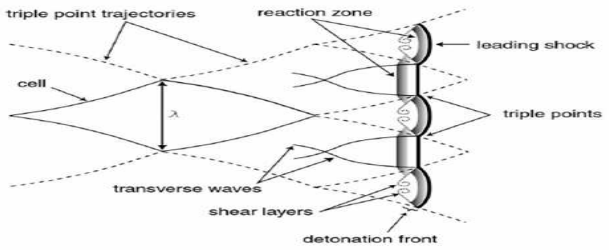

Figure 3: The cellular structure of the detonation wave [6]

between temperature (K), concentration of chemical species (mol/m^3) and ignition delay times. The experimental auto-ignition delay times were compared to the modelled ones using four different chemical reaction mechanisms taken from the literature (see Figures 1 and 2).

A detonation wave of gaseous fuel-air mixture has a multidimensional cellular structure consisting of a cellular pattern that could be experimentally determined using smoked foil. The width of the detonation cell correlates with several detonation dynamic parameters, e.g. initiation energy or critical tube diameter, and also reflects the stability of the detonation wave.

The cellular structure of the detonation wave is shown schematically in Fig. 3. There are 3 kinds of waves in a detonation wave, namely Mach stem, incident shock and transverse waves. Mach stem and incident shock waves travel in the same direction as the detonation front. Reflected shock waves are transverse to the detonation front. The velocity of the reflected shock waves is equal to the sound speed in combustion gases. Fig. 3 also shows triple point trajectories.

The collision of a pair of neighboring triple points results in the formation of a strong leading wave that initiates chemical reactions after a very short induction time. The leading shock wave propagating through the cell decays in strength. It is common to use Mach reflection terminology in describing the progression of the leading shock. In the first part of the cell when the strong leading shock is created from the triple-point collision it is referred to as the Mach stem. In the latter part of the cell leading up to the triple-point collision it is referred to as the incident shock.

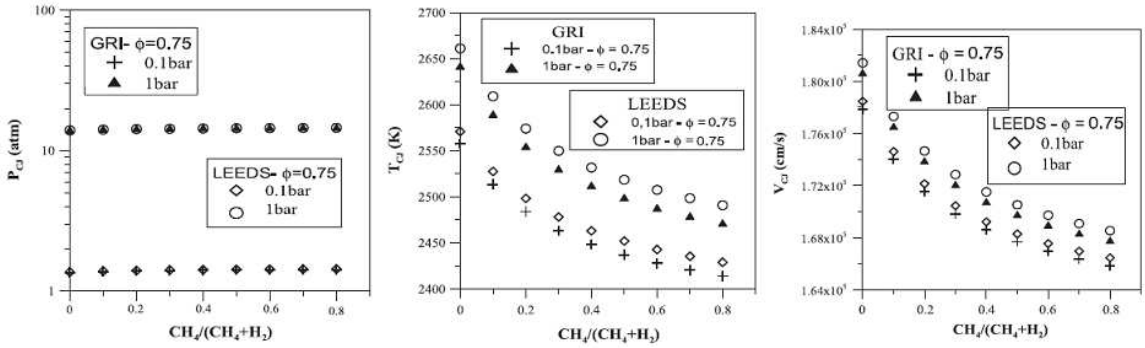

Figure 1: Evolution of CJ-parameters versus methane content in the mixture for two different initial pressures, 10 and 100 kPa and an equivalence ratio of 0.75. The mixture was initially at 298 K [5]

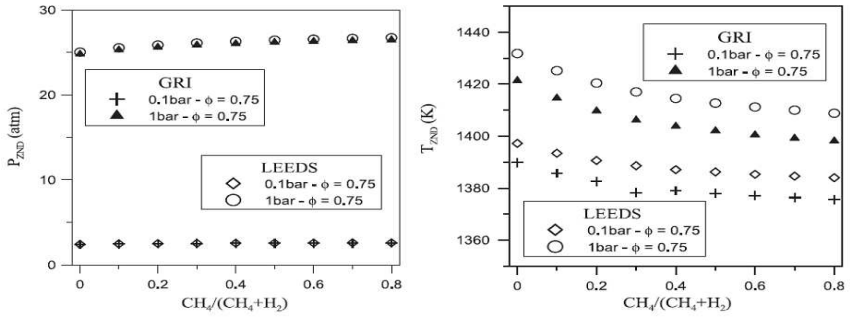

Figure 2: Evolution of ZND-parameters versus methane content in the mixture for two different initial pressures, 10 and 100 kPa and an equivalence ratio of 0.75. The mixture was initially at 298 K [5]

The objective of this study was to find experimentally the size of the cellular structure of detonation waves in hydrogen-methane-air mixtures.

2. Experimental

The experimental study was performed in a 6-m circular cross section tube with inner diameter of 140-mm (full diameter of 170-mm) and a 0.8-m driver section tube with inner diameter of 90 mm. The experimental tube consisted of four sections (2×2-m and 2×1-m) jointed together. The initial conditions of stoichiometric hydrogen-methane-air mixtures were 1 atm and 293 K with various hydrogen content. The flame propagation and pressure wave were monitored by pressure transducers and ion probes. Pressure transducers were positioned at several locations along the channel to collect data concerning development of the detonation. The experimental set-up is shown schematically in Fig. 4 and in the photographs in Fig. 5.

The average detonation cell size for stoichiometric hydrogen-methane-air mixtures obtained by the smoked foil technique during experiment was calculated with the 2D Fourier transform written by Hebral and Shepherd [7], as the subscript for Matlab application. For example, the cell size for the stoichiometric H_2-air mixture in our experimental set-up was 14-mm, and for the addition of 10% CH_4 in the mixture – 48-mm.

Figure 7 shows the cell sizes versus the number of cells for the stoichiometric hydrogen-air mixture. The blue line in Fig. 9 shows the average detonation cell size, which was equal to 14-mm, based on our image processing calculations. The picture in Fig. 7 presents the smoked foil with the detonation cell pattern of the same case.

Figure 7 shows the cell sizes versus the number of cells for 10% methane in the stoichiometric hydrogen-methane-air mixture. The blue line in Fig. 7 shows the average detonation cell size which was equal to 48 mm, based on our image processing

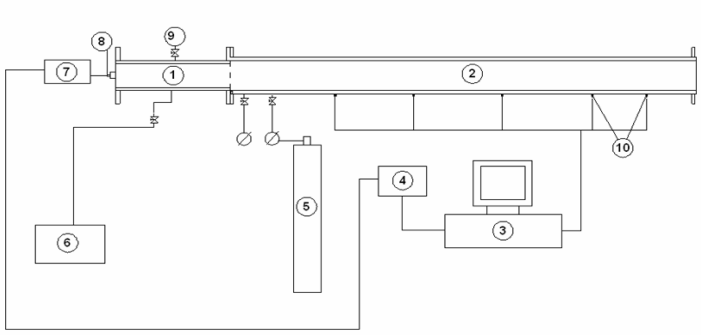

Figure 4: Experimental set-up where: 1 – driver-section tube, 2 – test-section tube, 3 – PC and data acquisition system, 4 – time sequencer, 5 – hydrogen-methane-air cylinder, 6 – vacum pump, 7 – ignition device, 8 – ignition plug, 9 – dillution valve, 10 – pressure transducers and ion probes

Figure 5: Photographs of the experimental set-up

calculations.

Furthermore, the experimental data were compared with the computational results for the detonation cell size, using Cantera software for chemical kinetics.

The main goal of the present work was to combine detonation cell width λ with ignition induction zone length Δ for detonation in hydrogen-methane-air mixtures. Detonation cell width λ is the characteristic feature of the mixture and provides information on reactivity. If the value of the detonation cell width is known it becomes possible to determine detonation limits and the energy required for direct initiation of the detonation. Previous numerical simulations of detonation propagation did not enable proper computation of the three dimensional cellular structure of the detonation wave and in consequence did

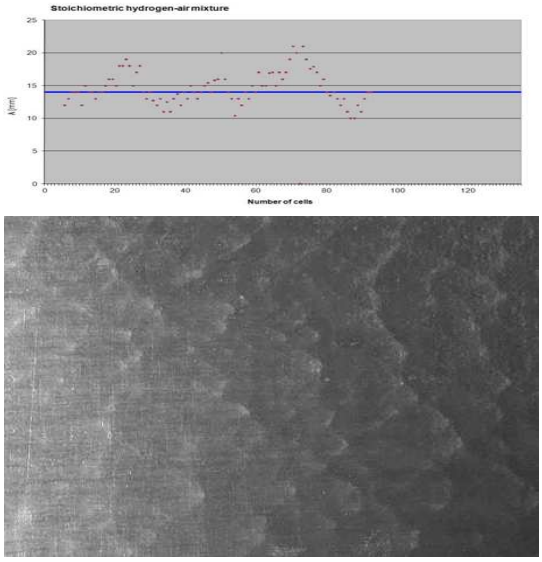

Figure 6: Detonation cell sizes for the hydrogen-air mixture

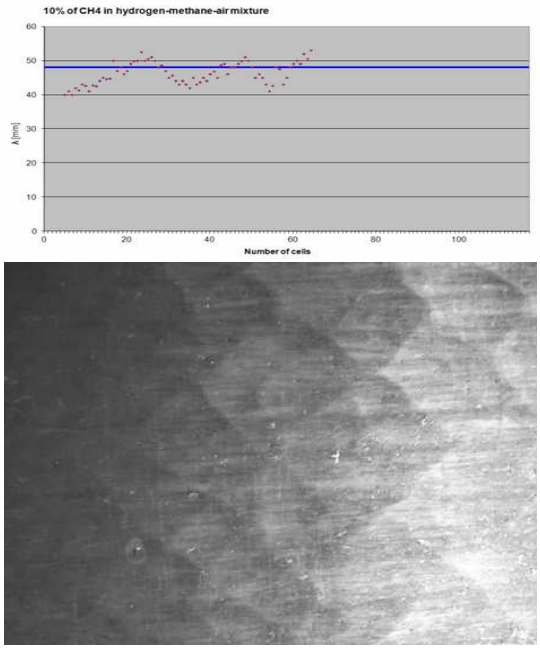

Figure 7: Detonation cell sizes for 10% of CH_4 in hydrogen-methane-air mixture

not allow one to determine with sufficient accuracy the value of detonation cell width λ. The best way to determine the value of λ is through experimental study.

Numerous analyses have shown, that detonation cell width λ can be related to ignition induction zone length Δ for the detonation process. In this work linear correlation λ=A·Δ was used, where A is a proportionality parameter. Using a model of the propagation of the detonation wave proposed by Zeldovich, von Neumann and Döring, it becomes possible to compute ignition induction zone length Δ with good accuracy. If the values of the ignition induction zone length and proportionality parameter A are known, the value of detonation cell width λ can be calculated. Ignition induction zone lengths Δ were computed using the ZND program. Computations with the ZND program are based on detailed chemical kinetics mechanisms.

Using Cantera, the detonation cell width was computed as a function of initial pressure and molar methane fraction in fuel for a stoichiometric methane-hydrogen-air mixture at initial temperature 295 K. The results of the computations were compared against the experimental results obtained in the laboratory for the same mixture and initial conditions. A chemical kinetics mechanism called LUTZ88 was chosen for computation of the λ value of the detonation wave cell size for hydrogen-methane-air mixtures. The mechanism involves 39 species and 154 reactions. Calculations were made with CANTERA and SDT toolbox. Ignition induction zone length Δ was computed using the ZND model. Particular detonation wave cell sizes were determined by the equation:

$$\lambda = A \cdot \Delta$$

where factor A equals 19. Figure 8 presents computational results with initial T=295 K, where x means mole fraction of methane in the mixture.

3. Conclusions

The computed detonation wave cell sizes for hydrogen-methane-air mixtures provide an overall review of certain risks and hazards which might occur during pipeline transport of such mixtures. Transporting hydrogen in mixtures with natural gas through existing natural gas pipelines would result in higher levels of safety than would be the case with pure hydrogen, especially in the context of detonation parameters. Furthermore, such mixtures could transform hydrogen into an energy carrier all over the world, but storage and transport problems in the volumes required for large industrial use means that

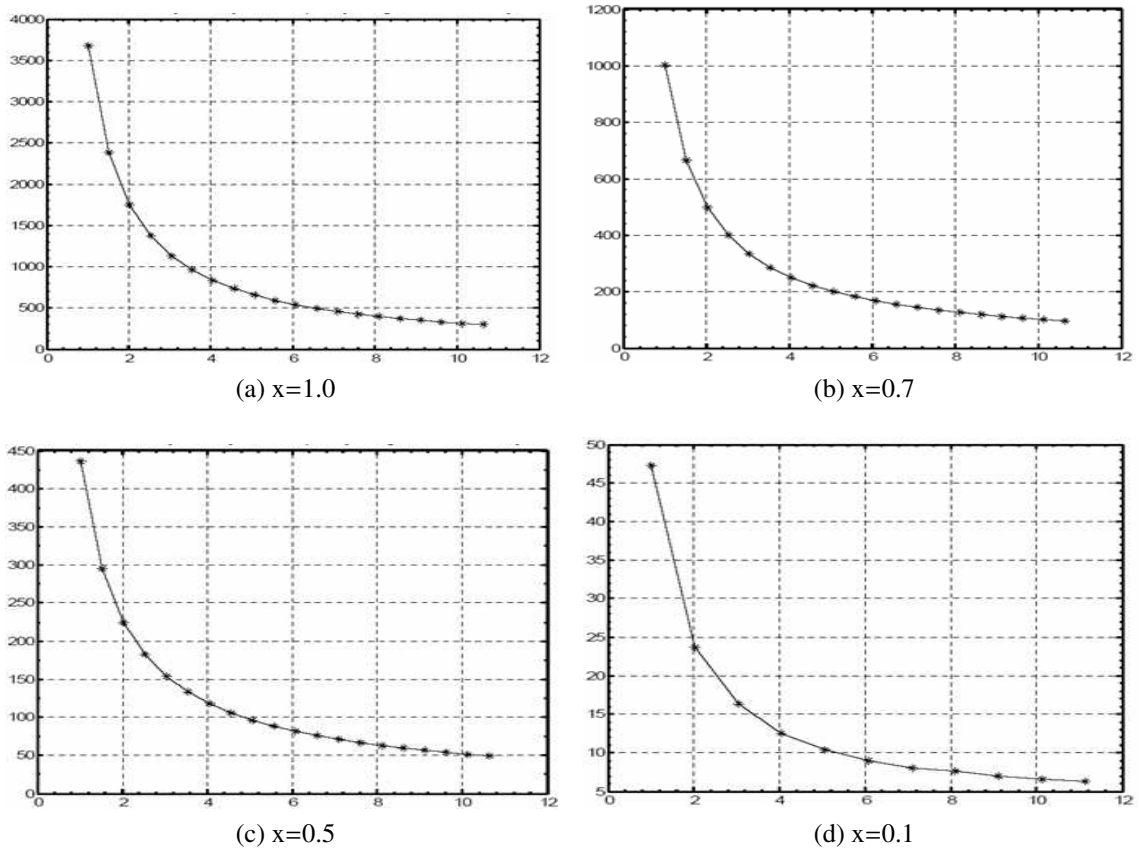

Figure 8: Detonation wave cell size as a function of initial pressure of hydrogen-methane-air mixtures for CH_4 mole fraction in the mixture: 1.0, 0.7, 0.5 and 0.1. Horizontal axes are $P_1 \times 10^4$, Pa; and vertical axes are in mm

little progress appears achievable in the foreseeable future.

Acknowledgement

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References

[1] G. Karim, I. Wierzba, Y. Al-Alousi, Methane-hydrogen mixtures as fuels, International Journal of Hydrogen Energy 21 (7) (1996) 625 – 631.

[2] J. Gauducheau, B. Denet, G. Searby, A numerical study of lean CH4/H2/air premixed flames at high pressure, Combustion Science and Technology 137 (1998) 81–99.

[3] J.-Y. Ren, W. Qin, F. Egolfopoulos, T. Tsotsis, Strain-rate effects on hydrogen-enhanced lean premixed combustion, Combustion and Flame 124 (2001) 717–720.

[4] M. Carcassi, F. F., Deflagration of H2-air and CH4-air lean mixtures in a vented multi-compartment environment, Energy 30 (2005) 1439–1451.

[5] N. Chaumeix, S. Pichon, F. Lafosse, C. Paillard, Role of chemical kinetics on the detonation properties of hydrogen/natural gas/air mixtures, International Journal of Hydrogen Energy 32 (2007) 2216–2226.

[6] J. Austin, F. Pintgen, J. Shepherd, Lead shock oscillation and decoupling in propagating detonations, in: 43rd AIAA Aerospace Sciences Meeting and Exhibit, no. AIAA-2005-1170, Reno, 2005.

[7] J.-P. Hebral, J. E. Shepherd, User guide for detonation cell size measurement using photoshop and matlab, Technical Report FM00-6, Graduate Aeronautical Laboratories, California Institute of Technology (2000).