EXPERIMENTS ON FLAME ACCELERATION AND DDT FOR STOICHIOMETRIC HYDROGEN/AIR MIXTURE IN A THIN LAYER GEOMETRY

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ABSTRACT

A series of experiments in a thin layer geometry performed at the HYKA test site of the KIT. The experiments on different combustion regimes for lean and stoichiometric H2/air mixtures were performed in a rectangular chamber with dimensions of $20 \times 90 \times h$ cm³, where h is the thickness of the layer (h = 1, 2, 4, 6, 8, 10 mm). Three different layer geometries: (1) a smooth channel without obstructions; (2) the channel with a metal grid filled 25% of length and (3) a metal grid filled 100% of length. Detail measurement of H2/air combustion behavior including flame acceleration (FA) and DDT in closed rectangular channel have been done. Five categories of flame propagation regimes were classified. Special attention was paid to the analysis of critical condition for different regimes of flame propagation as function of the layer thickness and roughness of the channel. It was found that thinner layer suppresses the detonation onset and even with a roughness, the flame is available to accelerate to speed of sound. The detonation may occur only in a channel thicker than 6 mm.

1.0 INTRODUCTION

1.1 Hydrogen Fuel Cells

The hydrogen fuel cell is the advanced source of energy based on the electrochemical potential of reaction between hydrogen and oxygen taking place at the electrodes (cathode and anode):

$$^{-2e^{-}}$$
 H2+0.5O2 $\stackrel{-}{\Rightarrow}$ H2O (1)

This is very promising energy carrier with respect to safety and CO, CO2 and NOX pollution into the atmosphere in comparison with hydrocarbon fuel internal combustion engines and gas turbines. Usually, one elementary fuel cell needs a supply of hydrogen and oxygen to the electrodes and an exhaust of water as a product of reaction. Hydrogen is delivered from a composite high-pressure tank (700 bar). Oxygen comes from atmosphere and water returns to atmosphere through the exhaust pipe. To provide the desired level of energy for car engine, 300-400 or even more, elementary fuel cells are combined into the fuel cell stack. One of the examples (Mirai) consists of 370 cells (single-line stacking) which provides 114 kW (155 PS) power.

The fuel cell stack consists of assembly of elementary fuel cells, hydrogen and oxygen (air) feed lines, exhaust pipeline, electric cables and supporting devices (Figure 1). The fuel cell stack is protected by metal jacket with a super electric and heat isolator. It could also be a gap between the fuel cell stack and a metal jacket with some atmosphere consisting mainly of air. The gap size is controlled by the breakdown voltage. Typical gap size is ~10 mm or ~ 5 mm with additional insulating foil. The leak of hydrogen from elementary fuel cells and gas feed line may lead to formation of hydrogen—air mixture in a gap between fuel cell stack and metal jacket. In presence of electric spark, the hydrogen-air mixture may burn with different flame propagation regimes depending on mixture reactivity and geometry of gaseous gap. Depending on the flame propagation regime, different internal pressure loads affecting the side walls may occur. Mechanical response of external metal jacket and integrity of the fuel cell with respect to hydrogen explosion pressure load is of the great importance for safe use of hydrogen fuel cells for hydrogen driven vehicles.

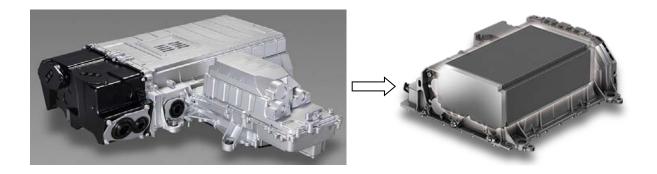


Figure 1. Typical fuel cell design [1].

1.2 Flame Propagation in a Layer Geometry

Different flame propagation regimes, the mechanisms of flame acceleration and DDT transition in different geometries were recently investigated. An influence of a specific layer geometry was investigated at KIT in a thin 2D-layer [2-3]. The layer thickness was changed from 2 to 6 mm in between two squared glass window of different dimensions 20x20 cm² and 50x50 cm² or two aluminum plates of 1x1 m². Hydrogen-air and hydrogen-oxygen mixtures were tested to investigate the flame dynamics and to evaluate the flame instability characteristics.

For lean mixture of $10\% H_2$ in air, thermo-diffusion and Landau-Darrieus instabilities lead to the development of a double-mode cellular structure of the flame surface. The primary thermo-diffusion instability appears almost immediately after the ignition, leading to quite stochastic, relatively small-size mode of cellular structure. Then, the large-size self-similar mode of cellular structure due to Landau-Darrieus instability develops with a growing cell size looking similar to the flower petals. The shape of flame front is horizontally asymmetric due to the effect of buoyancy because flame velocity in upper direction is higher than to the bottom. Almost the same behavior demonstrates 15% H2-air combustion but with higher velocity and less influence of buoyancy (Fig. 2). Since the Markstein number becomes positive for hydrogen-air mixtures above $15\% H_2$ it reduces the sensitivity of the flame to thermo-diffusion instability. This is why very smooth stable flame surface can be seen for mixtures with 30 and 50% H2 in air at the initial stage of the process because the positive Markstein number and the Lewis number Le > 1 (Fig. 2).

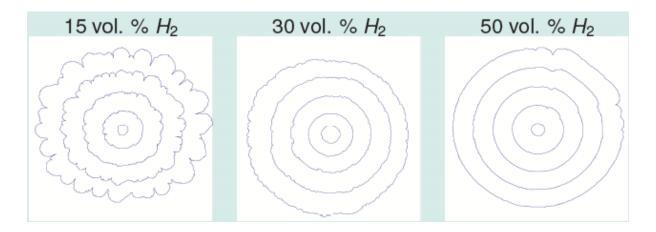


Figure 2. Flame propagation of lean H_2 – air mixture for 15, 30 and 50% H_2 in air in a 6-mm gap layer.

After a flame radius of the order of 10-15 cm, the cellularity of the flame surface appears only due to the Landau-Darrieus instability. The flame finally accelerates 1.4-1.6 times compared to laminar flame speed due to the flame wrinkling and folding according to thermal diffusion and Landau-Darrieus

instabilities mechanisms. The same flame acceleration factor 1.4 was theoretically found for real hydrogen-air mixtures depending on mixture reactivity and expansion rate of combustion products in papers [4-5] using Sivashinsky equation analysis [6].

Except the own flame instability due to thermal-diffusion and Landau-Darrieus, additional mechanisms of flame acceleration, like shear layer or boundary layer producing turbulent flow ahead the flame might also be involved into the further flame acceleration to sonic velocity and then even to detonation. It was found the minimum size or so called run-up-distance to deflagration-to-detonation transition (DDT) for certain layer geometry should be not less than 20 cm from ignition point [2-3]. Since the chamber was separated from ambient air with plastic film, significant effect of the nature of interface between the test mixture and surrounding atmosphere was discovered in the tests. Figure 3 shows the case of detonation onset within a shear layer between the tested hydrogen-oxygen mixture and ambient air. The mixture was ignited in upper corner at the centerline position which is not covered by light beam. Two sides at the ignition position were closed and two opposite were open (shown in Fig. 3, right). Hydrogen-air mixtures behaves in the same way with a difference that the detonation may occur in several times thicker layer with a longer run-up-distance.

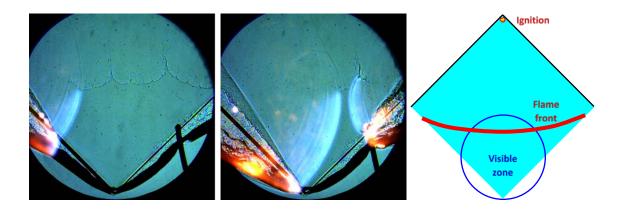


Figure 3. Example of flame propagation and detonation. Layer size 50x 50cm² h=6 mm, two sides open, 60 % H2 in oxygen, ignition in upper corner (the scheme shown right).

All phenomena discussed now are relevant to a smooth thin layer geometry. Of course, the penetration of any obstructions will promote the flame acceleration to speed of sound and then reduce the run-up-distance to detonation. Some disadvantage of obstruction inside the layer could be suppression of flame acceleration due to heat losses to obstructions. In general, the problems of flame acceleration and DDT in a fully closed thin layer are not investigated yet. Such a scenario of hydrogen explosion may exactly appear inside a metal jacket of hydrogen fuel cell stack.

1.3 Effect of a Layer Thickness on Flame Propagation Velocity

According to paper [7-8] the scale effect becomes of the importance when dimensionless scale factor L_T/δ is order of 100, where L_T is the integral length scale (tube diameter D in a channel geometry); δ is the laminar flame thickness as a measure of chemical reactivity, $\delta = \chi/S_L$; χ is the thermodiffusivity. If the ratio L_T/δ is less than 100, only slow subsonic flames due to local extinction phenomena could be expected in the experiments. The reason is that the turbulence generation and the flame acceleration could be suppressed in a narrow gap by some energy losses due to viscous friction, heat exchange and other mechanisms.

Thus, the scale problems might be very important for thin layer geometry, like for the gap between fuel cell stack and jacket of the fuel cell housing. Since the characteristic size of the gap is only 5-10 mm, the most reactive flames with laminar flame thickness less than 50-100 mkm will be able to accelerate to sonic velocity. For hydrogen-air mixtures at normal conditions, it corresponds to the

mixtures with hydrogen concentration more than 16-18% with laminar flame thicknesses less than 50-100 mkm (calculated by Cantera [9]).

One more reason for flame deceleration and even quenching could be heat losses and steam condensation leading to loss of "piston" energy. This phenomenon is more pronounced in smaller scale with increased specific surface of the system [10, 11]. When the flame is passing through a channel or an orifice of the size L(D), the energy losses are proportional to the surface of cold walls. If specific surface is too large, the heat losses to side walls may lead to flame quenching if the ratio $L/\delta < 42$ [12]. The ratio D/δ associates with Peclet number $Pe = D_Q/\delta$. The critical Peclet number Pe = 42 also corresponds to so called quenching distance (diameter) D_Q which is less than $42 \cdot \delta$. According to this formula, the calculated quenching distances for 14%H2 and 16%H2 in air are 8.5 mm and 4.2 mm, respectively. This means that for hydrogen concentrations less than 14%H2 we may expect the flame extinction in a layer of 8 mm. For hydrogen-air mixture of 16%H2, we also may expect an extinction of the flame in a layer thickness of 4 mm or less.

The quenching phenomenon is more pronounced in smaller scale with increased specific surface of the system. Table 1 shows the calculated by Cantera [9] values of critical layer thickness L leading to local flame extinction (Q) or to flame acceleration (FA) limit calculated according to [7-8] for lean hydrogen-air mixtures.

Hydrogen	Initial	Initial	Laminar flame	Flame	Quenching
	temperature	pressure	thickness	acceleration limit	limit
%H2	To, K	Po, bar	δ, mm	$L_{\rm FA}$, mm	$L_{\rm Q}$, mm
9	293	1	0.150	15.0	6.3
10	293	1	0.107	10.7	4.5
11	293	1	0.081	8.1	3.4
12	203	1	0.064	6.4	2.7

Table 1. Theoretical flame acceleration (FA) and quenching (Q) limits for a layer geometry.

Thus, the flame may accelerate to speed of sound if the layer thickness exceeds the flame acceleration limit $L_{\rm FA}$. Another mechanism of laminar flame acceleration is so called "finger" flame behavior, when the flame dynamics is governed by the geometry factor or increasing flame area with a distance. As the gap size decreases, the specific area of the flame gets higher, causing the burning velocity to increase for 2D-geometry as follows [2-3]

$$S_f = \frac{dx}{dt} = \frac{A}{A_0} \sigma S_L = \left(1 + \frac{x}{h}\right) \sigma S_L, \tag{2}$$

where $A = 2\pi x^2 + 2\pi xh$ is the visible flame area including the area in a funnel between the flame and solid surface which is proportional to the flame radius x in a layer with a constant gap h; A_0 is the planar flame area assumed to be ring-shaped with a radius x, $A_0 = 2\pi xh$; $\sigma = \rho_u/\rho_b$ is the expansion ratio of combustion products compared to the unreacted mixture; S_L is the fundamental laminar flame speed. This means that for "finger" flames in a layer geometry, the visible flame velocity is inversely proportional to the gap size h. In general, such a mechanism only allows to reach the speed of sound for flame propagation velocity. Then, additional mechanisms for flame acceleration like shock-flame interactions, shear layer or boundary layer producing turbulent flow ahead the flame might also be involved into the further flame acceleration to sonic velocity and even to detonation. It was found that the possibility of detonation onset and then propagation fits to detonation cell size λ [13] but up to now there are not enough experimental data on critical conditions for detonation propagation in a thin layer geometry.

1.4 Scope of the Work

The main goal is detail measurement of H_2 /air combustion behavior including flame acceleration (FA) and DDT in a closed layer geometry in order to provide experimental data for numerical code validation for combustion processes in a layer of hydrogen–air mixtures. Critical conditions for different flame propagation regimes with respect to the safety depending on channel gaps and obstructions to be investigated.

2.0 EXPERIMENTAL DETAILS

A series of hydrogen combustion and detonation experiments was done in a layer geometry to model an enclosure with a thin gap between two plates. The facility was vertically installed a front of reflecting screen. The relative position of light source, high speed camera (Fastcam SA1.1, Photron), test facility and reflecting screen should provide required distances for optical system to get proper quality of schlieren images.

The experimental facility consists of a rectangular channel with inner dimensions of 900x200x (1-10) mm³ (LxWxh), which is made of PVC frames to fix the gap size h = (1, 2, 4, 6, 8, 10) mm between two Plexiglas windows and to keep isolated the test mixture from ambient atmosphere (Fig. 4). Principal positions of igniter (red cross), three pressure sensors (blue diamonds) and gas inlet (black ring, top left) and outlet (black ring, bottom right) are shown in Fig. 4. Wooden profiles were used as flanges to fix together all elements of the assembly by 24 metal C-clamps. Two Plexiglas windows of 12 mm thickness were used as side walls. They played a role of transparent windows for optical access as well. Another important reason was the possibility to mount pressure sensors and a spark plug directly to the windows.

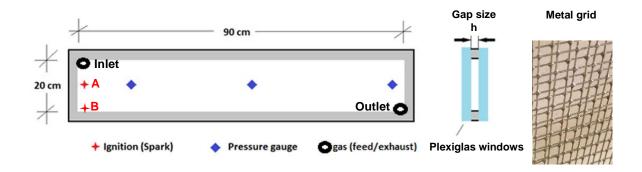


Figure 4. Scheme of the channel with narrow gap. Ignition positions A and B are shown in red. Metal grid consists of 2 layers of metal grid 6.5x6.5x0.6 mm. It may fill the gap in entire length or in 25% from ignition side.

To record the overpressure history during experiments a set of 3 fast dynamic pressure transducers (Type PCB M113B28, characteristic rise time $1\mu s$) and a fast data acquisition system (500 kHz) were used. Sample rate of the pressure records was established in the range of 1 μs . Pressure sensors were installed at the centerline with coordinates from end of the channel x_1 =150 mm (P1); x_2 =500 mm (P2); x_3 =850 mm (P3). The pressure transducers are mounted to the side wall in flush position with the internal surface to do not disturb the pressure signal profile.

As discussed in introduction, the presence of roughness promotes the flame acceleration and reduces the run-up-distance to fast sonic flame or even to detonation. To do that, one or two layers of fine metal grid (size $6.5 \times 6.5 \times 0.6$ mm) were placed inside the channel because real roughness of glass windows cannot be provided. The efficient blockage ratio depending on gap size changes from BR = 0.6 for 1- and 2-mm channels to BR=0.12 for 10-mm channel (Table 2). Another variable was to

change the congestion by filling entire gap with a metal grid (100% congested), to fill only a quarter of the channel (25% congested) or to keep the channel free of any obstructions (0% - smooth channel).

Table 2. Blockage ratio of channels with metal grid.

Channel width, mm	1	2	4	6	8	10
Blockage ratio	0.6	0.6	0.3	0.2	0.15	0.12
Note	1 layer	2 layers				

Lean and stoichiometric hydrogen-air mixtures from 4 to 30% H2 at ambient initial conditions (~20 °C; ~1000 mbar) were tested. The mixtures were chosen to cover the domain from flammability limit to flame acceleration and detonability limits according to diagram of state for flame propagation regimes [7, 13]. Main thermodynamic and combustion properties of hydrogen-air mixtures to be tested were calculated using STANJAN [14] and Cantera [9] packages and given in Table 3.

Table 3. Combustion characteristics of tested hydrogen-air mixtures.

		Sound	Adiabatic	Adiabatic	Sound		Laminar
Hydrogen		speed in	combustion	combustion	speed in	Expansion	flame
concentration	Density	reactants	temperature	pressure	products	ratio	speed
	ρ	$c_{\rm r}$	T_b	P_{icc}	c_p	σ	$S_{ m L}$
$%H_{2}(mol.)$	kg/m ³	m/s	K	bar	m/s	(-)	m/s
4	1.141	350	623	2.47	500	2.08	0.021
6	1.119	354	783	3.13	559	2.59	0.035
8	1.096	357	939	3.75	612	3.08	0.067
10	1.074	361	1093	4.33	659	3.54	0.124
12	1.052	365	1244	4.88	703	3.99	0.213
13	1.041	367	1319	5.14	724	4.21	0.270
15	1.019	371	1466	5.64	765	4.63	0.410
16	1.008	373	1539	5.88	784	4.83	0.493
17	0.997	375	1611	6.11	802	5.03	0.585
20	0.964	381	1824	6.75	855	5.60	0.904
30	0.854	405	2388	8.18	983	7.02	2.201

The gas filling system was used to prepare the test mixtures. It consists of two mass flow rate controllers EL-FLOW® series manufactured by Bronkhorst High-Tech B.V. for air and hydrogen. Both components of the mixture injected, via T-junction, to the mixing chamber. Before the mixture injected to combustion channel it goes from mixing chamber through the bypass line to gas analyser to prove the mixture composition. The accuracy of the mixture composition was better than +/- 0.25 % H2. Then, the test channel was filled through the inlet port with test mixture. Inlet port mounted directly to the upper left part of Plexiglas window and outlet port locates at the right bottom of it (Fig. 4) to provide better replacing of heavier gas (air enriched) to the lighter gas (test mixture). Continuously, within 5 minutes, the air inside the channel fully replaced by the test mixture with a flow rate of 25 dm³/min providing an exchange rate more than 100. The quality of outlet gas is checked using the gas analyser. After proper quality of test mixture is established, both valves (feed and exhaust) are closed. The mixture exposure time takes 1-2 minutes to provide stagnant conditions for test gas to suppress any initial turbulent movement. All experiments were performed at ambient conditions of 1 bar and 293 K. The mixture within the glass plate assembly was ignited in the centerline (Ign. A) or in the corner (Ign. B) using a spark igniter (Fig. 4).

3.0 EXPERIMENTAL RESULTS AND ANALYSIS

More than 100 tests have been processed to extract characteristic velocity and maximum combustion pressure. Figure 5 shows two examples of slow and fast (detonation) flame propagation regimes. Slow subsonic deflagration was more typical for smooth channel (0% congestion). The flame accelerates due to the flame instability and turbulent flow ahead the flame, see Fig. 5 (left). The visible flame velocity never exceeds the speed of sound. Fast flame propagation regimes as sonic flame or detonation mainly occurred in fully obstructed channel (100% congestion). The cellular structure of detonation can be resolved in Fig. 5 (right).

Dynamics of flame propagation were obtained from image processing of high speed movies and maximum combustion pressure for different congestion degree and a layer thickness. The effect of hydrogen concentration on flame propagation velocity for lean hydrogen-air mixtures is shown in Fig. 6, left. The repetitive tests marked with label "b". The local velocity never exceeds the level of 10 m/s for lean hydrogen-air mixtures in a smooth 10-mm thick channel (Fig. 6, right). Maximum combustion pressure is not higher than 0.5 bar. Final pressure reduces to -100 mbar due to the effect of steam condensation.

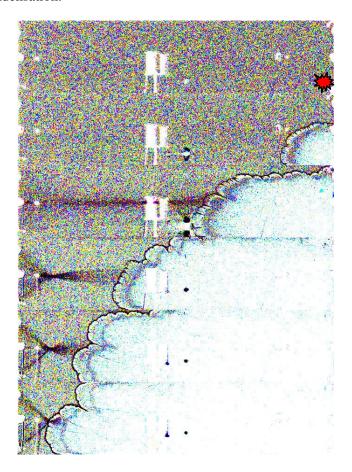




Figure 5. Slow (left, 0% congested, 3000 fps) and fast-detonation (right, 100% congested, 20000 fps, time step 0.05 ms) flame propagation regimes for 30 % H2 in air in a 6-mm channel.

The influence of layer thickness on combustion behavior was investigated for fixed congestion degree of the channel. For smooth channel the velocity never exceeds 80 m/s even for stoichiometric mixture. It increases a bit with a layer thickness increase but never approaches to the speed of sound (Fig. 7). Maximum combustion pressure for smooth channel (0% congestion) changes in the range 1-2 bar and agrees well with characteristic velocity less than speed of sound (Fig. 7).

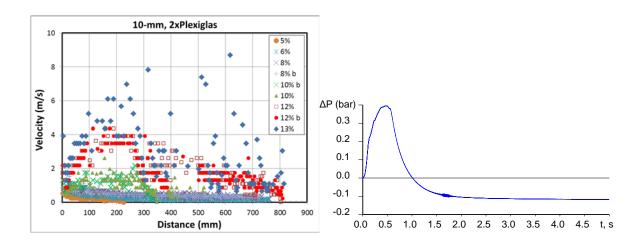


Figure 6. Dependencies of local flame propagation velocity vs. distance for lean H2-air mixtures in 10-mm layer w/o obstacles obtained from high speed image processing (left). Typical pressure record P_1 (x_1 =150 mm) for 12% H2-air in 10-mm layer without obstacles (right).

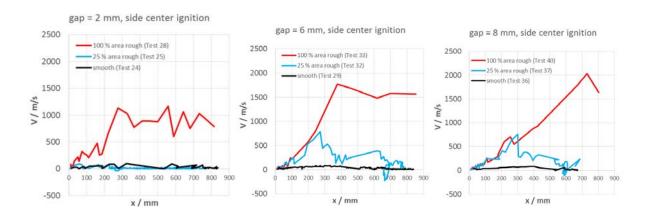


Figure 7. Dependencies of local flame propagation velocity for stoichiometric H2-air mixture in 2, 6 and 8-mm gap vs. distance obtained by image processing for stoichiometric hydrogen-air mixture.

The congestion of 25% of the channel length leads to more efficient initial flame acceleration after ignition. The flame accelerates until the channel is blocked, then velocity decays to the level of less than 80 m/s, typical for smooth channel. For the layer thickness > 4 mm, the local velocity may reach speed of sound or even higher values in the range of 1000-1500 m/s but detonation never occurs for 25% congestion. For thinner layers the velocity never exceeds 80 m/s with 25% of congested space. Maximum combustion pressure agrees well with dynamics of flame propagation. Since the local velocity reaches 1000-1500 m/s, the maximum combustion pressure in these cases also increases to 3-4 bar. For the layers thinner than 4 mm, the maximum combustion pressure never exceeds the level of 0.5 bar (Fig. 8).

The strongest combustion regimes are realized for 100% congestion of the channel length. In all cases with a layer thickness >4 mm the detonation of stoichiometric hydrogen-air mixture with corresponding maximum pressure 12-16 bar is occurred (Fig. 8). For 2 mm layer thickness, sonic flame propagation is usually observed with a velocity ~1000 m/s and corresponding pressure 7-8 bar. For 1 mm layer thickness, only slow subsonic flame or flame extinction occurs due to energy losses and steam condensation (Fig. 9). Due to the strong heat losses in a narrow channel and an additional influence of metal grid the global flame extinction occurred near the center of the channel. As longer the flame propagates, the more heat losses leading to steam condensation and loss of overpressure. Near mid of the channel, the flame finally stops propagating due to global extinction.

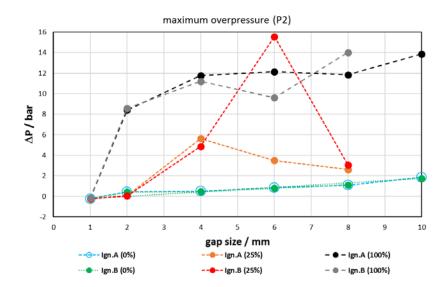


Figure 8. Dependencies of maximum combustion overpressure vs. gap size for stoichiometric H2-air mixture in a channel with different congestion degree.

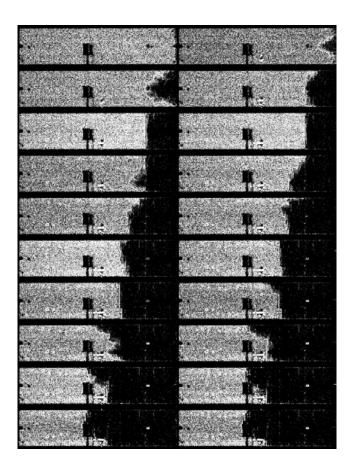


Figure 7. Global flame extinction: stoichiometric H2-air, 1-mm layer, 100% congestion, Ign. A, 3000 fps (every 25th frame).

Six categories of flame propagation were classified, based on experimental data analysis:

- a) Non-ignition;
- b) Ignition & local extinction;
- c) Ignition & slow deflagration;
- d) Ignition & fast deflagration;
- e) Ignition & detonation;
- f) Ignition & local explosion.

The main criteria to choose the category were the maximum combustion pressure and the maximum flame propagation velocity relatively to speed of sound. Other criteria were the flame instability and heat losses due to steam condensation leading to local extinction in narrow channels, even for stoichiometric hydrogen-air. In some experiments transient regimes may occur. For instance, rather high local pressure of 10 bar and maximum flame velocity of about 700 m/s occurred in the tests with 25% congestion degree within the congested area. Then high velocity decays to 80 m/s since the channel has no roughness generating the turbulence supporting the fast deflagration regime. Another transient regime occurred in 4-mm channel when fast deflagration with local explosion occurred but it doesn't lead to steady detonation because of limited width of the channel compared to the detonation cell size $\lambda = 10$ mm which is the measure of detonability for combustible mixture. All the data on flame propagation regimes based on velocity and pressure measurements were put in Table 4. The safety domain with lower maximum combustion pressure and flame velocity is colored in green and blue.

Layer thickness 6-mm 10-mm 4-mm 1-mm 2-mm 8-mm Geometry BR=0 c c c c c BR=25% d/c d/c d f b e e BR = 100%

Table 4. Diagram of state for combustion regimes in a thin layer.

4.0 SUMMARY

The experiments on different combustion regimes for lean (4-20% H2) and stoichiometric H2/air (30% H2) mixtures were performed in a closed rectangular chamber with dimensions of $20 \times 90 \times h$ cm3, where h is the gap size (h = 1, 2, 4, 6, 8, 10 mm).

Three different layer geometries were used: (1) a smooth channel without obstructions; (2) the channel with a metal grid filled 25% of space and (3) 100% of channel space filled with a metal grid.

Critical conditions for six different flame propagation regimes as function of the layer thickness and a presence of obstructions were evaluated from experimental data analysis.

It was found that in a smooth channel, without obstructions, the flame is not available to accelerate to sonic velocity and to detonation. Local flame velocity does not exceed 10 m/s for lean mixtures with maximum combustion pressure less than 0.5 bar and for stoichiometric mixture less than 80 m/s and the pressure less than 1-2 bar.

The only metal grid may lead to the efficient flame acceleration to speed of sound and DDT in a channel thicker than 4 mm.

In 1-mm channel 100% filled with a metal grid, the strong effect of steam condensation, leading to local and then global flame extinction was registered.

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