

## QUENCHING EFFECTS AT JET IGNITION OF LEAN HYDROGEN- AND METHANE-AIR MIXTURES

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### ABSTRACT

Jet ignition through an orifice of premixed hydrogen- and methane-air flames with the same laminar burning velocity was investigated. The diagnostics were based on optical measurement techniques to get inertialess information about the flame activating and quenching effects without disturbing the physical process itself by the probes. Mainly two effects could be identified at lean hydrogen flames. Before the flame reaches the orifice, small amounts of radicals are separated from the main reaction zone. These radicals start the combustion process within the free jet area at an early stage. As the bulk of radicals passes the orifice, a parallel jet penetrates into the second room for about  $2 \cdot 10^{-4}$  sec, before it breaks up and a fast reaction takes place. At methane flames radicals passing the obstacle are deleted totally after four orifice diameters. Ignition behind the orifice could be observed after about  $1.5 \cdot 10^{-2}$  sec.

### INTRODUCTION

Jet ignition is usually treated in literature [8, 15] to be the first stage of the transition from deflagration to detonation, whereby the proposed transition mechanism is based on the Shock Wave Amplification by Coherent Energy Release (SWACER). Most of the reported observations therefore deal with quite sensitive fuel-air-mixtures. Almost no investigation is available for lean mixtures, which are also worth to look at, as far as building safety considerations are concerned. Already relatively small amounts of natural gas (substituted at our experiments by methane) or hydrogen released, due to an assumed failure of pipes or valves, inside industrial or civil structures, can form together with the ambient air a highly explosive mixture. The combustion of hydrogen within the course of a hypothetical accident in a nuclear power plant is one of the main issues of safety considerations in connection with light water reactors. The deliberate ignition of hydrogen by sparks before concentrations sensitive to violent combustion modes are reached, is a possible approach, as the resulting loads on the structure remains low and a decreasing of the hydrogen and oxygen concentration is achieved [4]. The application of deliberate igniters has to rely on a precise knowledge about the

flame acceleration mechanism in partially obstructed confinements.

If the ignition threshold of a fuel-air mixture is sufficiently weak, the combustion process will start out as a slow deflagration. However, due to the heat release of the exothermal reaction, a highly turbulent expansion flow is created inside confined geometry's, which superimposes with the flame itself. It has been shown [1, 2, 3, 9, 14], that the interaction of chemical kinetics and heat and mass transfer due to turbulent mixing and momentum exchange processes can significantly accelerate flames to limits, where a damage of the building structure is to be expected.

Experiments on jet ignition performed by the authors and Mr. Carcassi at the University of Pisa showed some interesting effects which could not totally be described and explained due to lacks of instrumentation. The used test facility consists of a chamber with the inner dimensions of  $677 \times 677 \times 3200$  mm, which is divided into two rooms. The first room has a length of 1050 mm and is separated from the second room by a wall with a central round orifice of 10 cm diameter, which causes a blockage ratio of 98.3 %. The second room has a weak rupture disk to the ambient with the dimensions of  $300 \times 300$  mm. Two axial fans within the separation wall ensure a homogeneous fuel-air mixture of the same equivalence ratio in both rooms before ignition, which is checked by 3 hydrogen concentration sensors in each room.

The conventional instrumentation consists of 7 high-speed piezo-capacitive pressure transducer and 7 thermocouples. For the visualization of the flame propagation a video camera with a frame rate of 25 Hz is used. The combustion is recorded simultaneously through the front windows and, reflected by a  $45^\circ$  mirror, also through the top windows. For contrast enhancement, aerosols of a NaCl solution are added, which are excited to emit light by the high combustion temperature [7].

With a two component LDV system the flow velocity is measured inertialess and non-intrusive for the horizontal component parallel to the main flow and the vertical component at three different points (#1-#3 in figure 1).

Selected frames of a lean methane air flame (6 vol.-%  $\text{CH}_4$  in  $\text{CH}_4$ -air,  $\Phi = 0.63$ ) at this facility are shown in figure 1. After ignition, a nearly spherical flame propagates preferably in

upward direction, due to the buoyancy effect (figure 1, 480 ms after the first observation of a flame).

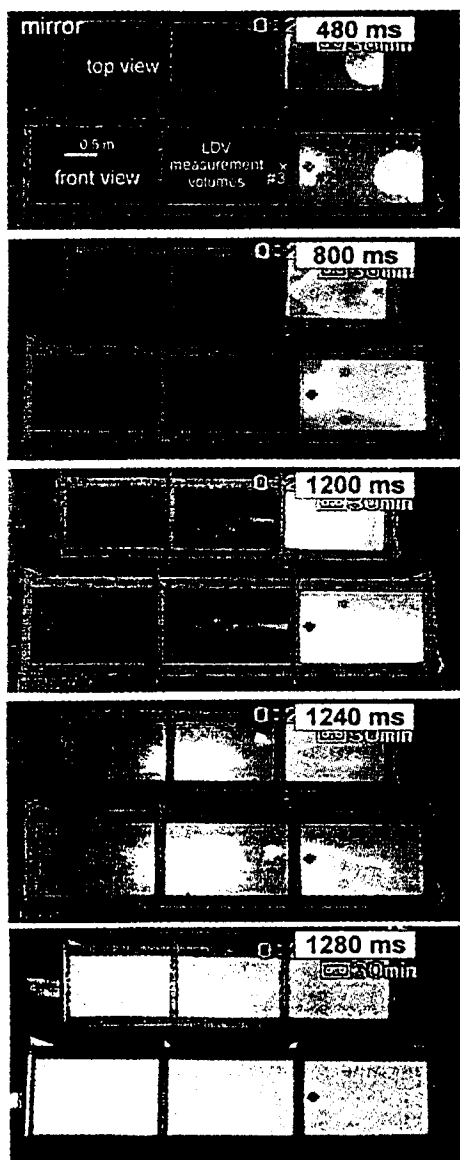


Figure 1: Selected frames of a 6 vol.-%  $CH_4$  in  $CH_4$ -air flame at the L.VIEW facility. The time span between the images is not constant.

As soon as the flame is influenced by the orifice, the flame velocity highly increases, caused by two different effects. Firstly, due to the sudden reduction of the free cross section up to a minimum of 1/58 at the orifice the expansion flow of the hot gas is highly accelerated. Secondly, the combustion itself is accelerated, due to an increase of the turbulence intensity in front of the flame, which is described for example by [5, 14].

After 800 ms, when the flame reaches the orifice, due to the high gas velocity (114 m/s, measured with LDV) a fast ignition of the second room would be expected. Nevertheless, it takes another 400 ms before the first spots of ignition can be detected in the second chamber. The following combustion process is too fast for the resolution of the 25 Hz video camera. One frame later (1240 ms) a flame can be observed at two different positions inside the second volume. The high pressure release in the second room leads to a flow not only through the broken

rupture disk (figure 1, frame 1280 very left) but also back into the first room, where a very fast out-burn takes place.

At higher methane concentrations or with hydrogen as fuel, this effect of single ignition spots inside the second chamber cannot be observed, due to the low repetition rate of the applied camera.

With a methane-air mixture reduced by only 0.1 vol.-%  $CH_4$  compared to figure 1, which is about the order of magnitude for the fuel concentration reproducibility, leads to a totally different, but reproducible burning behavior (figure 2).

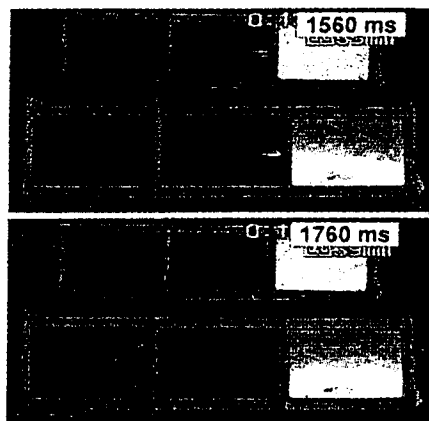


Figure 2: 5.9 vol.-%  $CH_4$  in  $CH_4$ -air, although a flame burns into the second room no ignition takes place.

Although an open jet flame burns into the second room (which is filled with the same mixture as the first room) for about 720 ms, no ignition of the gas in the second room takes place. The lower gas velocity of the leaner mixture in the middle of the jet (103 m/s for 5.9 vol.-% and 114 m/s for 6 vol.-%  $CH_4$ ) can obviously not compensate the much higher minimum ignition energy, needed at a lower methane concentration as shown in figure (3) [13].

Within the following it is explained, how the ignition behind an obstacle with a very high blockage ratio can take place at different positions, after the flame is quenched as observed at figure 1 frame 1200 and 1240. For the investigation of the described physical effects, advanced optical measurement techniques are needed. As the application of these techniques is very difficult at large and medium size facilities, a small scale apparatus was used for the tests.

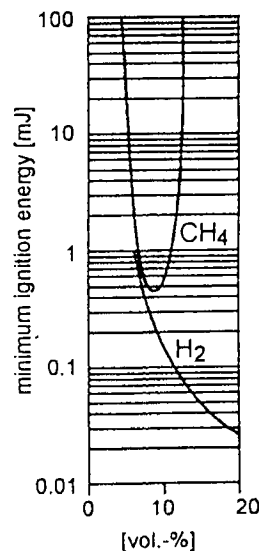


Figure 3: Minimum ignition energies

#### EXPERIMENTAL APPARATUS FOR THE APPLICATION OF ADVANCED OPTICAL TECHNIQUES

The inner area of the optical test section of the facility is 6539 mm<sup>2</sup> (figure 4). An obstacle with a central orifice opening of 16 mm diameter causes a blockage ratio of 97 %. As the two valves (V1 and V2 in figure 4) are closed during the tests, the

total gas volume is about 25 liter in front of the obstacle and about 36 liter after the obstacle. Four quartz windows provide proper optical access from all sides for the optical measurement techniques.

The density gradient of the gas is recorded by means of Schlieren in combination with a high speed video camera working at a frame rate of 9000 frames/sec and a storage capacity of 1000 frames.

As with the Schlieren technique the density gradient of the gas is visualized integrally through the whole depth of the chamber, the flame itself cannot be distinguished from the hot exhaust gas and any structures in depth cannot be resolved. Therefore laser induced predissociation fluorescence (LIPF) is used to visualize thin slices of the flame. OH radicals, indicating the location of the reaction zone, are excited within a lightsheet with a thickness of less than 1 mm. By using an KrF excimer laser, emitting light with a wavelength of 248 nm, the excitation of the OH-radicals  $A^2\Sigma^+, V=3 \leftarrow X^2\Pi, V=0$  is achieved. The laser is tuned for the P1(8) excitation band of OH. The pulse duration of the laser is 17 ns, the lifetime of the OH radicals in the excited state is about  $10^{-10}$  to  $10^{-5}$  sec. The fluorescence appears frequency shifted ( $A^2\Sigma^+, V=3 \rightarrow X^2\Pi, V=2$ ) at a wavelength of 295 – 304 nm. Other fluorescence signals as well as Rayleigh scattering is tuned out with an optical filter.

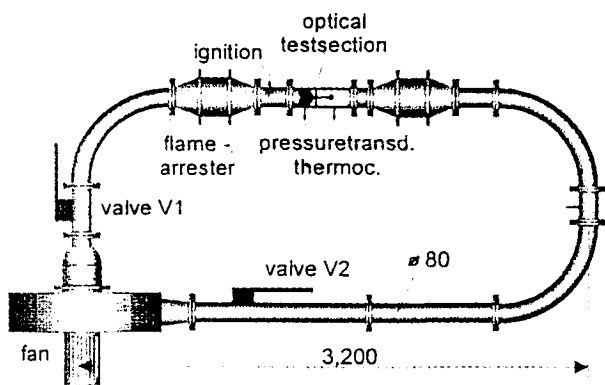


Figure 4: Experimental apparatus

## RESULTS AND DISCUSSION

Figure 5 shows selected frames of the gas propagation during the combustion of a lean hydrogen- (left column) and stoichiometric methane-air (right column) flame, using the Schlieren technique. With this method the combustion process can be resolved in time even during jet ignition. Although both mixtures have about the same laminar burning velocity of 0.4 m/sec [12], the flame behavior is completely different.

As from the beginning of ignition the flame interferes with its own generated pressure waves, the hydrogen flame starts to wrinkle and to form cusps, which does not happen with the methane flame. Basically two transport processes are responsible for this different stability behavior. As soon as a positively curved cusp to the unburned mixture is generated, the enlargement of the preheating zone leads to an increasing heat loss of the reaction zone by thermal diffusion. On the other hand the larger area to the unburned gas enables more molecules of the deficient reactant (fuel in the lean case) to be transported into the reaction zone by mass diffusion. The corresponding negatively curved sections contain accordingly less molecules of the deficient reactant and are therefore locally quenched. These effects are described by the dimensionless Lewis number,

which is the ratio between the thermal diffusion  $a = \lambda / (\rho c_p)$  and the mass diffusion  $D$ . The Lewis number for lean hydrogen air flames is approximately 0.3 – 0.5. For stoichiometric methane flames a Lewis number of about unity can be assumed. At lean hydrogen concentrations positively curved flame sections - caused by acoustic interactions or a turbulent flow - tend to grow, methane flames tend to smooth any interference [1, 2, 11].

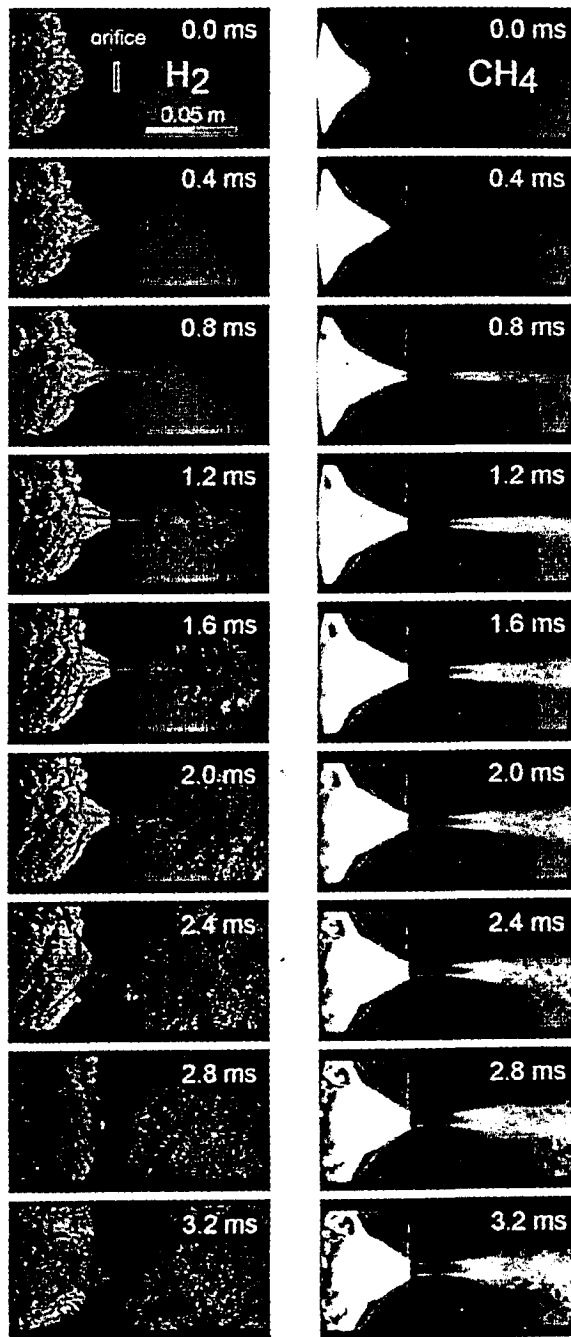


Figure 5: Selected Schlieren images; left: 12 vol.-% hydrogen-air ( $\Phi = 0.41$ ); right: 9.5 vol.-% methane-air ( $\Phi = 1$ ) flame

The propagation velocities can be determined by counting the frames of the high speed record (figure 6). The maximum velocities can be observed right behind the orifice, further downstream, the gas expands also to the walls which leads to a decrease of the axial propagation velocity.

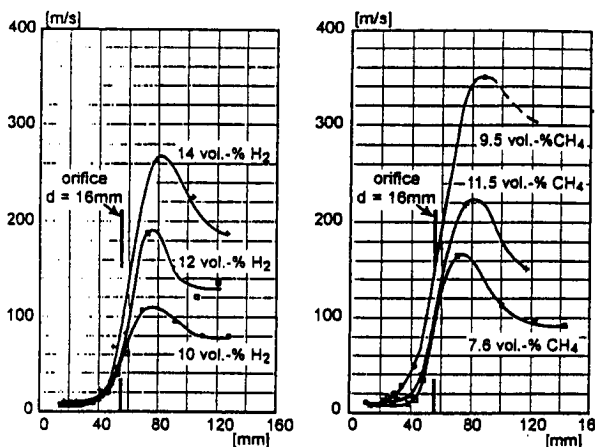


Figure 6: Velocities during the combustion of various fuel-air mixtures

By observing the film<sup>1</sup> of figure 5 with hydrogen, a back flow of the hot gas (figure 5; 2.4 ms), followed by a fast out-burn of the first room (2.8 ms) can be observed, similar to the experiments shown in figure 1.

The flame shreds in the second room, when the flame burns through the orifice as mentioned above (figure 1), cannot be identified at these experiments. As with the Schlieren technique the density gradient of the gas is visualized, it is difficult to distinguish between flame and burned gas. As a matter of fact it is not even possible to indicate the exact time and place of ignition in the second room with this method.

Therefore the corresponding OH-radical distribution to the Schlieren records of figure 5 are visualized by means of planar LIPF (figure 7) [10, 16].

In the first room the above explained theory of local flame quenching at negatively curved cusps at lean hydrogen mixtures can be confirmed. At 12 vol.-% hydrogen in air a high reactivity can be observed at the positive curved flame sections, whereby the flame totally extinguishes at the corresponding negatively curved sections. This effect takes place at two different length-scales – 1/2 tube diameter and 4 mm – simultaneously. The radicals of the stoichiometric methane-air flame (having the same laminar burning velocity) are distributed along the expected smooth contour.

Although, due to the low repetition rate of the LIPF system, only one frame can be exposed per flame, the flame propagation can be reconstructed by evaluating several single shots taken at different flame positions (figure 7). About 1 cm in front of the orifice, both flames are highly influenced by the flow through the orifice. With methane this effect leads to a forced positive curved still very smooth flame with low reactivity, due to the enhanced heat transport. On the contrary, the reactivity at the hydrogen flame at the positive curvatures increases.

At the bottom of figure 7 several examples of tests with identical hydrogen concentrations are zoomed in. It can be seen, that small amounts of radicals are separated from the main reaction area. This effect could be caused by the high pressure gradient during the acceleration process between the unburned gas in front and the burned gas after the orifice. On the other hand, the high velocity of the unburned gas at the edge of the

flame, which also flows through the orifice, leads to an enhanced traverse strain. In combination with the large surface due to the un-regular structure of the hydrogen flame this is a further explanation for the radical separation.

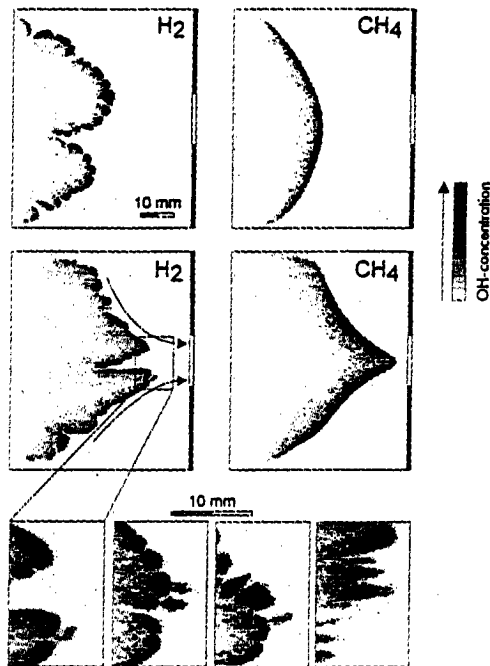


Figure 7: Flame shapes in front of the orifice. Left: hydrogen-air ( $\Phi = 0.41$ ); right: methane-air ( $\Phi = 1$ ) flame. Bottom: several examples showing the separation of the radicals from the main flame

As with LIPF not the flame itself but the OH radicals are visualized, the separated radicals do not necessarily indicate a reaction zone, they also can be thermally quenched, due to the high acceleration. Further downstream, when the gas velocity decreases after the maximum of about 200 m/s (ref. to figure 6) these few separated radicals in front of the main reaction zone stimulate the combustion process again without having any visible connection to the flame in front of the orifice itself (figure 8). The observed quenched time of the radicals before re-ignition is in most cases of the order of magnitude of  $10^{-4}$  sec.

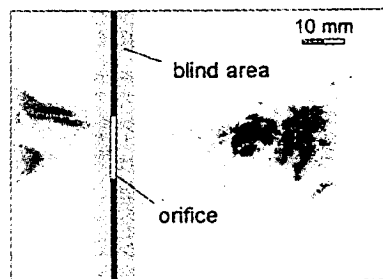


Figure 8: Ignition in the second room without any visible connection to the first room ( $H_2$ -air  $\Phi = 0.41$ ).

<sup>1</sup> the whole film can be downloaded from the internet homepage of the chair: <http://www.thermo-a.mw.tu-muenchen.de/Lehrstuhl/Projekte/puflag>

effect of the ignition in the second room before the main reaction zone passes the orifice cannot take place.

As soon as the hydrogen flame itself reaches the orifice, a free jet penetrates into the second room (figure 9). Over a length of two to three diameters of the orifice (which is about  $2 \times 10^{-4}$  sec), this jet has a very straight, parallel orientation, due to the high penetration velocity. The superposition of the free jet and the increasing volume of the burned gas leads to a formation of a complex turbulent structure and macro scale eddies in the shear layer of the jet, which cause a very fast reaction.

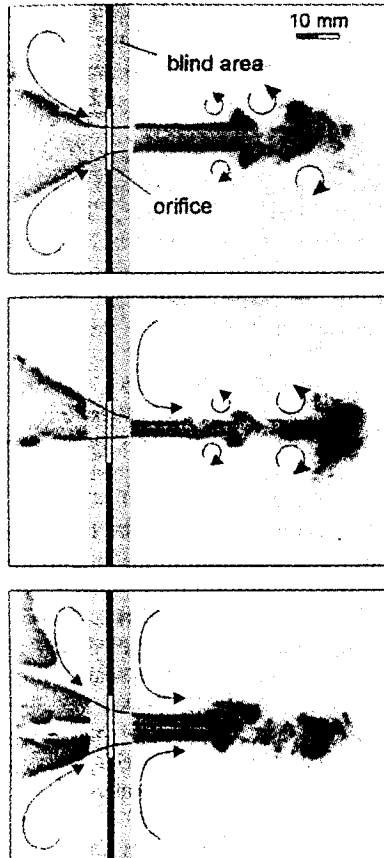


Figure 9: Examples of the OH-radical concentration and ignition in the second room ( $H_2$ -air  $\Phi = 0.41$ )

With methane a quite similar parallel radical free-jet penetrates into the second room, being accelerated up to the order of magnitude of sonic speed. After about  $1 \times 10^{-4}$  sec the parallel radical jet breaks up but no ignition in the second room takes place so far, due to the high minimum ignition energy needed for methane-air mixtures (figure 10). It takes about  $1.5 \times 10^{-2}$  sec, before the main ignition in the second chamber can be observed. Hence the Schlieren images of figure 5 (right) only indicate the path of the exhaust gas and not the flame itself. The ignition within the second chamber using a much leaner methane mixture as seen in figure 1, can be explained by a lower gas velocity, due to the different geometrical configuration and burning volumes. The ignition mechanism of the observed flame shreds can be compared to the small scale tests run with hydrogen, where local flame quenching and re-ignition could be observed behind the orifice, by visualizing the radicals.

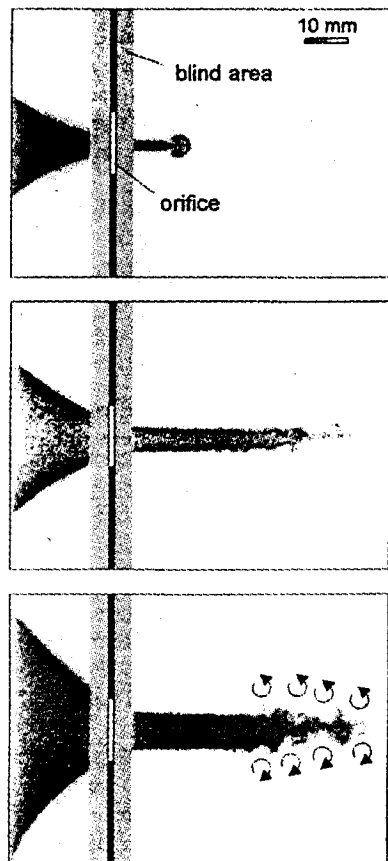


Figure 10: Examples of the OH-radical concentration in the second room ( $CH_4$ -air  $\Phi = 1$ )

Approximately 2.5 ms after the images shown in figure 9 the out-burn of the first room can be observed using the LIPF technique. Figure 11 shows two examples, both with 12 vol.-% of hydrogen. The corresponding Schlieren image is about 2.4 ms in figure 5. Just in front of the orifice, the very high reactivity is caused by the back flow of the burning gas from the second room which induces a high turbulence intensity and a good mixture between burned and unburned gas. This area of high reactivity is still enclosed by quite laminar flamelets [6, 17] burning to the direction of totally unburned gas (compare figure 5).

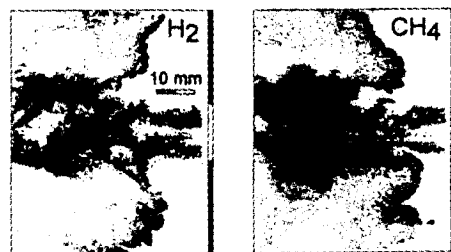


Figure 11: Two examples of the out-burn of the first room at the same conditions of 12 vol.-%  $H_2$ -air

## CONCLUSION

Jet ignition of stoichiometric methane-air and lean hydrogen-air flames with about the same laminar burning velocity was investigated. Applying an orifice plate in a closed confinement flame propagation velocities were observed 1000

times higher than the laminar burning velocity of the mixture. Local flame quenching leads to an accumulation of free radicals behind the orifice, which has to be taken into account for the calculation of the turbulent burning velocity. After ignition, these free radicals lead to a very fast reaction in the second room which cannot only be explained by an enhancement of the turbulent burning velocity due to the high turbulence intensity. The pressure release behind the orifice leads to a back flow upstream of the orifice and a out-burn of the first room, resulting in a alternating pressure stress of the orifice plate itself.

#### ACKNOWLEDGEMENTS

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