EFFECT OF TURBULENT TRANSPORT AND MIXING ON FLAME ACCELERATION THROUGH HIGHLY BLOCKING OBSTACLES

Martin Jordan, Nikolai Ardey, Franz Mayinger
Lehrstuhl A für Thermodynamik
Technische Universität München
85747 Garching
Germany

Marco Carcassi
Dipartimento di Costruzioni Meccaniche e Nucleari
Università degli Studi di Pisa
Via Diotisalvi 2
56126 Pisa
Italy

ABSTRACT

The propagation of gaseous explosions under premixed conditions is governed by both molecular heat and mass transport, and the interaction of the flame front with the turbulent expansion flow generated by the combustion process itself. The molecular transport controls the flame stability and its sensitivity to flame stretching and quenching. Therefore it determines the response of a flame front to flow vorticity, resulting in either flame acceleration or turbulent quenching. The present paper reports on experimental investigations of transport phenomena during flame propagation with highly blocking flow obstacles by means of non-intrusive, optical techniques. It is shown, that turbulent flame quenching processes lead to an acceleration of free radicals behind the obstacle. The mixture of these radicals with the unburned gas leads to an enhanced chemical reaction and an increase of the turbulent burning velocity.

A comparison of two different fuels (hydrogen and methane) is presented, strongly differing in molecular behavior due to their different Lewis numbers. The experiments have been performed in two explosion tubes of different scale (PuFlaG facility: ∅ 80 mm and L.VIEW facility: 0.7×0.7 m) in order to address scaling effects. Blocking ratios between 95% and 99.7% have been investigated.

A high-speed video-camera operated at repetition rates of up to 9000 images/sec is used to record the self-fluorescence of the flame and to visualize the flame propagation. The path of the hot gas is recorded via Schlieren photography with the high-speed video-camera. The OH-radicals in the cross section of the flame are visualized with laser induced predissociation fluorescence with an exposure time of several nano-seconds.

INTRODUCTION

Due to a possible failure of any gas infrastructures at industrial or civil sites, gaseous fuel, such as hydrogen or natural gas could be released. In combination with the ambient air, a highly explosive mixture can be formed, which endangers the building integrity. The ignition of relatively small amounts of fuel in air, having a low laminar burning velocity, will start out as a slow deflagration. However, due to the heat release of this exothermal reaction, a highly turbulent expansion flow is created inside confined geometries, which superimposes with the flame itself.

It has been shown by various authors (Beauvais 1994; Ardey et al. 1995,1997; Phylaktou et al. 1991) that the interaction of chemical kinetics and heat and mass transfer processes due to turbulent mixing in combination with momentum exchange processes can significantly accelerate flames to limits, where the damage of a building structure is to be expected. The influence of obstacles inside the structure on the flame acceleration, the turbulent burning velocity, and the resulting pressure loads have been investigated in detail by /Ardey, Durst, Mayinger/.

However, it will be shown, that at highly blocking obstacles (> 95%) the flame acceleration can not only be explained by the increase of turbulence, when the expansion flow passes the obstacle.

EXPERIMENTS AT THE L.VIEW FACILITY

First experiments in the scope of this work on jet ignition were performed in Pisa at the L.VIEW facility. The apparatus consists of a rectangular test section with the inner dimensions of 677 x 677 x 3200 mm, which is divided into two chambers. The first chamber has a length of 1050 mm and is separated from the second chamber by a wall with a central round orifice with a

diameter of 10 cm, resulting in a blocking ratio of BR = 98.3 %. The second chamber has a weak rupture disk to the ambient with the dimensions of 300 x 300 mm. Two axial fans inside the dividing wall ensure a homogeneous fuel-air mixture of the same equivalence ratio in both chambers before ignition, which is monitored by 6 hydrogen concentration sensors.
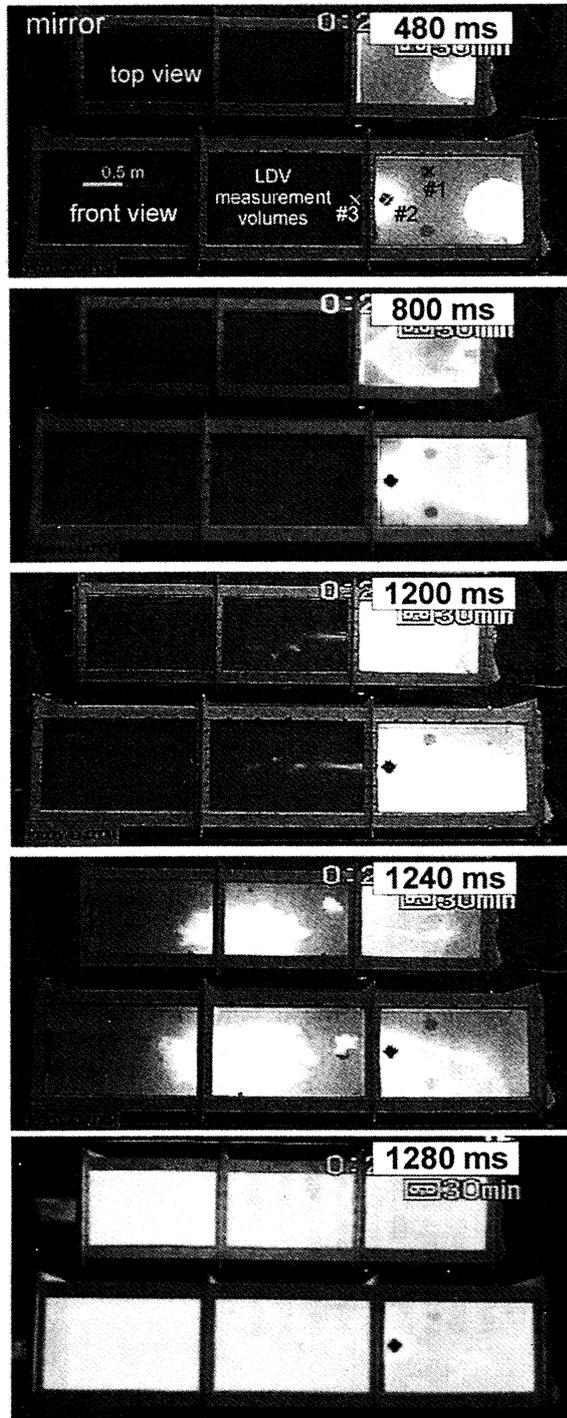

Figure 1: Selected frames of a 6 vol.-% CH_4 in CH_4-air flame at the L.VIEW facility with an obstacle with a blocking ratio of BR = 98.3%.

The conventional instrumentation consists of 7 high-speed piezo-capacitive pressure transducer and 7 thermo-couples. To visualize 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° mirror, also through the top windows. To enhance the contrast, aerosols of a NaCl solution are added, which are stimulated to emit light by the high combustion temperature. The flow velocity is measured inertialess and non-intrusively for the horizontal component parallel to the main flow and the vertical component at three different positions (#1-#3 in figure 1) with a two component LDV system.

In figure 1 selected frames of a lean methane-air flame (6 vol.-% CH_4 in CH_4-air Φ = 0.63), which was ignited at the lower right corner, are shown. Three remarkable peculiarities can be observed during the combustion process:

1. 800ms after ignition the flame reaches the orifice for the first time. Due to the high penetration velocity of the gas through the orifice (in this case 140 m/s, measured with LDV), a sudden ignition in the second chamber would be expected. However, another 400ms are needed before the first spots of ignition can be observed in the second chamber.

2. The ignition in the second chamber does not occur in the vicinity of the orifice as it would be expected. In fact several spots of ignition can be detected separately from each other.

3. Once the second chamber is ignited, a high combustion rate is observed.

Caused by the high pressure release in the second chamber not only a flow through the broken rupture disk (figure 1, frame 1280 very left), but also back into the first chamber occurs. This leads to a very fast final combustion of the unburned gas in the first chamber, resulting in the highest pressure of the whole process.

Due to the relatively low repetition rate of the camera used at these tests it is not possible to resolve the ignition process in the second chamber at higher methane concentrations or with hydrogen as fuel. A concentration of 6 vol.-% methane is in fact the leanest mixture where ignition in the second chamber could be observed. With 5.9 vol.-% CH_4 in CH_4-air the flame behaviour in the first chamber is very similar to the example shown in figure 1, though no ignition takes place in the second chamber.

EXPERIMENTS AT THE PUFLAG FACILITY

Due to the relatively big dimensions of L.VIEW it is difficult to apply sophisticated optical measurement techniques to analyse the processes in this facility. Therefore, small scale tests have been performed at the PuFlaG apparatus, which is very good equipped with optical measurement techniques. The inner area of the optical test section is 6539 mm^2. An obstacle with a central orifice of varying diameters causes blocking ratios between 95% and 99.7%. The total gas volume is about 25 liters in front of the obstacle and about 36 liters behind the obstacle. Four quartz windows provide a proper optical access from all directions for optical measurement techniques. Four high-speed piezo-capacitive pressure transducers are used to record the pressure history during the combustion process.

The gas propagation through the orifice is recorded by means of Schlieren records. The images are recorded with a high-speed video-camera, driven at frame rates of up to 9000 frames/sec. With the Schlieren technique the density gradient of the gas is visualized, therefore the flame itself cannot be distinguished from the hot exhaust gas. For that reason, the self fluorescence of the OH-respectively the CH-bands is recorded, to determine the flame itself. The light is amplified by a combination of two intensifiers in front of the high-speed camera.

Nevertheless, both methods deliver integral images through the whole depth of the chamber, so any structure in the depth cannot be clearly resolved.

Therefore, thin layers of the flame are visualized with the laser-induced predissociation-fluorescence (LIPF). OH radicals are excited within a lightsheet with a thickness of less than 1 mm. By using an excimer laser run with KrF, emitting light with a wavelength of 248 nm, the excitation of the OH-radicals $A^2\Sigma^+,V = 3 \leftarrow X^2\Pi,V = 0$ appears. A special optic allows to tune the laser for the P1(8) excitation of the OH-radical. The pulse duration of the laser is 17 ns, the lifetime of the OH radicals in the excited state ranges between 10^{-10} and 10^{-5} sec (Eckbreth 1988). The fluorescence appears frequency shifted ($A^2\Sigma^+,V=3 \rightarrow X^2\Pi,V=2$) at a wavelength of 295 – 304 nm. Additional fluorescence signals as well as Rayleigh scattering is tuned out by means of a filter.

Most of the PuFlaG tests presented in this paper were performed with a gas mixture of either 12 vol.-% H_2 in H_2-air ($\Phi = 0.41$) or 9.5 vol.-% CH_4 in CH_4-air ($\Phi = 1$). Both mixtures have approximately the same laminar burning velocity of 0.4 m/s. The minimum ignition energy is about 0.45 mJ for the stoichiometric methane-air mixture and about 0.08 mJ for the $\Phi = 0.41$ hydrogen-air mixture (Lewis et al. 1951).
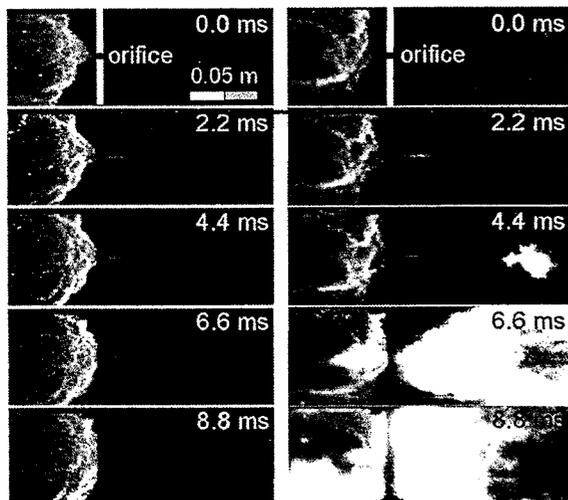

Figure 2: Selected frames of the self fluorescence of the flame; left: 12 vol.-%; right: 12.2 vol.-% H_2 in H_2-air flame at the PuFlaG facility with a blocking ratio of BR = 99.7%.

Corresponding to the L.VIEW tests, a limiting orifice diameter for each fuel and each concentration can be determined, where the flame is not quenched completely and ignition in the second chamber occures first. For the

12 vol.-% hydrogen flame, the limiting orifice diameter at this geometrical configuration was found to be 5 mm (BR = 99.7%) for total flame quenching. In figure 2, selected images of a 12 vol.-% H_2 flame compared to a 12.2 vol.-% H_2 flame are shown. Both flames have approximately the same velocity in front of the orifice (approx. 3.3 m/s) but as the richer flame reignites after a certain time (2 ms after the flame reaches the orifice first), the leaner mixture is quenched completely. As it has been measured at the L.VIEW facility, the ignition of the second chamber does not appear in the vicinity of the obstacle but approximately 20 orifice diameters behind the obstacle.
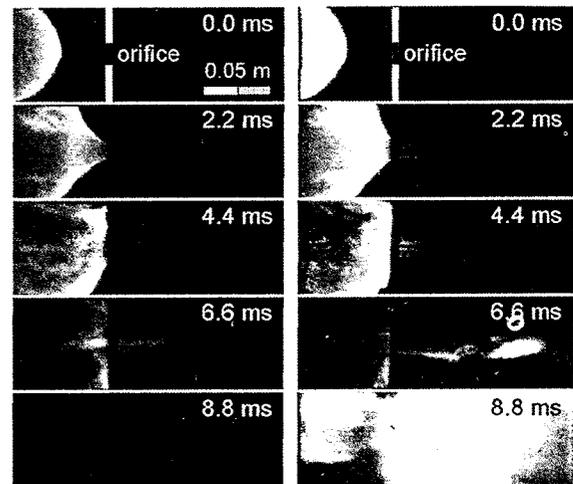

Figure 3: Selected frames of the self fluorescence of the flame; left: 9.4 vol.-%; right: 9.5 vol.-% CH_4 in CH_4-air flame at the PuFlaG facility with a blocking ratio of BR = 95.2%.

For the methane air mixture the limiting orifice diameter was found to be 20 mm (BR = 95.2%), as shown in figure 3. A stoichiometric mixture leads to a combustion process behind the orifice, a mixture of 0.1 vol.-% less than that does not ignite inside the second chamber.

A more detailed analysis of the beginning of the combustion in the second chamber, taken with the Schlieren technique, points out, that also with hydrogen as fuel, several spots of ignition can occur, as shown exemplarily in figure 4 at 1.2 ms. In this example a 16 mm orifice (BR = 97%) has been used, therefore the location of the ignition is much closer to the orifice as it has been in the test with the 5 mm orifice.

The above mentioned reverse flow of the hot gas into the first chamber, followed by an enhanced reactivity occurs also in the tests with the 16 mm orifice with hydrogen after 2.4 ms (figure 4).

The gas propagation velocities through the orifice can be determined by measuring the location of the gas jet and the time passed between two frames of the high-speed records. In figure 5, these velocities are plotted for different hydrogen- and methane-air mixtures for the tests with the 16 mm orifice. The maximum velocities for both mixtures can be observed immediately behind the orifice. Further downstream, the gas expands to the walls yielding to a decrease of the axial propagation velocity.
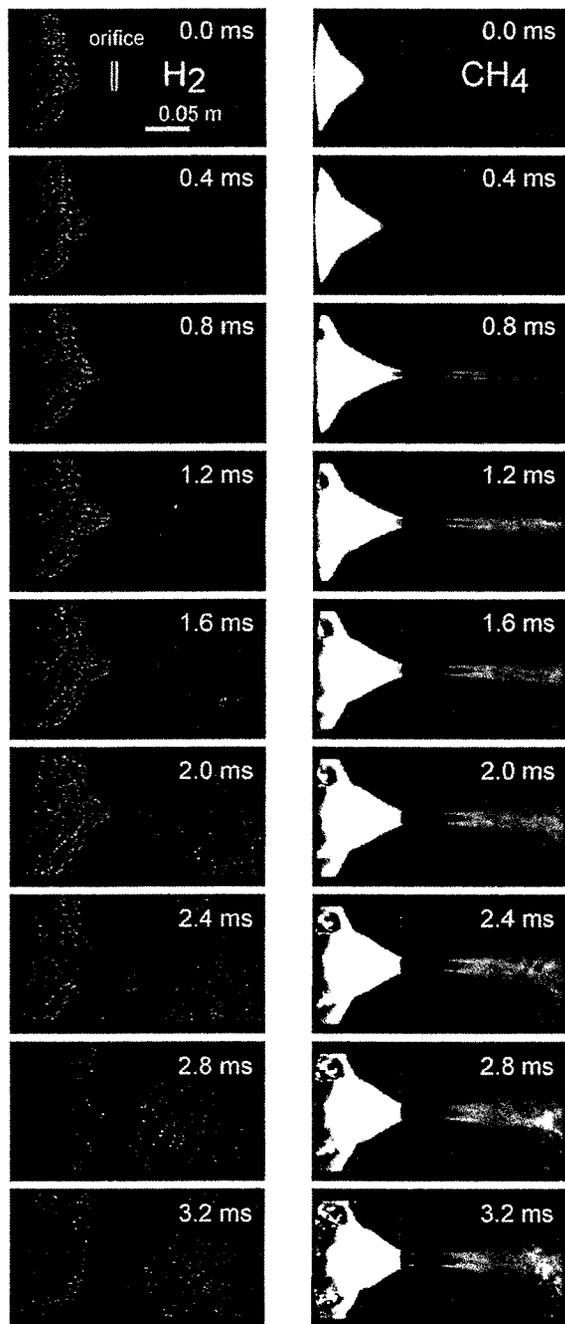
Figure 4: Selected Schlieren images; left: 12 vol.-% H2 in H2-air; right: 9.5 vol.-% CH4 in CH4-air; BR = 97%

Although the laminar burning velocity of both mixtures is approximately the same, the hydrogen- and the methane-air flame show a completely different behaviour. Since from the beginning of ignition the flame interferes with its own generated pressure waves, the hydrogen flame starts to wrinkle and to form cusps. This does not happen with the methane flame. Two transport processes are basically responsible for this different stability behavior. As soon as a positive curvature 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. In addition to that, 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. Corresponding negative

curved sections contain accordingly less molecules of the deficient reactant and are therefore locally quenched. These effects can be described by the dimensionless Lewis number, which is the ratio of 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. Contrarily, methane flames tend to smooth any interference (Ardey et al. 1995, Goix et al. 1993, Mayinger 1996).
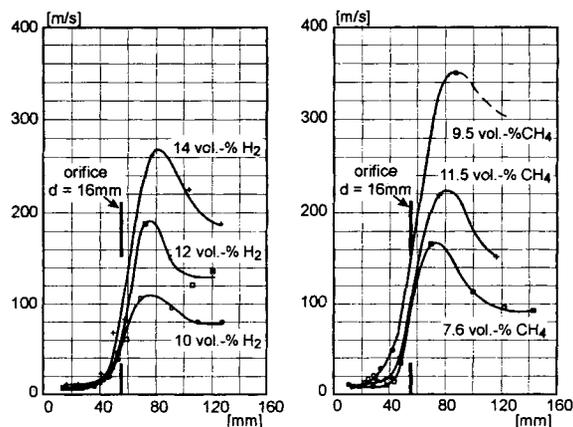

Figure 5: Propagation velocities during the combustion of various fuel-air mixtures through an orifice; BR = 97%

This effect becomes even more evident at the corresponding LIPF measurements of both mixtures. 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 6). 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 but still very smooth flame, with less reactivity, due to the enhanced heat transport. In contrast to that, the reactivity of the hydrogen flame at the positive curvatures increases.

As soon as the hydrogen flame itself reaches the orifice, a free jet penetrates into the second chamber (figure 7). Due to the high penetration velocity of up to almost 200 m/s, this jet has a very straight, parallel orientation for a length of two to three orifice diameters. The radicals in this area can be considered to be quenched since any combustion would lead to a propagation direction perpendicular to the jet surface, which has not been observed. The reignition of the flame can be determined due to the sudden increase of the radical concentration, most of the times simultaneously at several different positions. The superposition of the free jet and the increase 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 can be regarded as one reason for the fast combustion in this area.
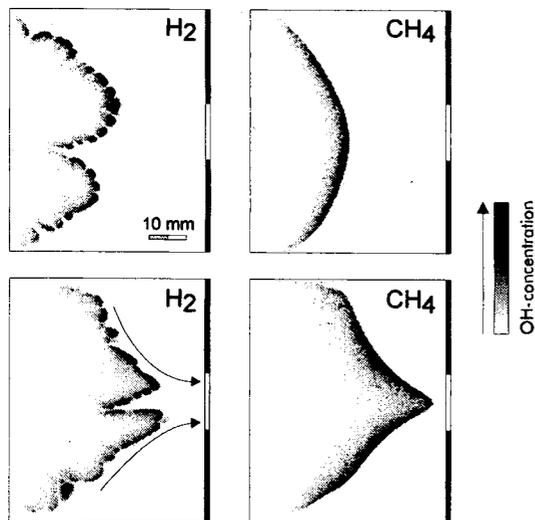
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Figure 6: OH-radical distribution in front of the orifice at different times taken with LIPF; left: 12 vol.-% H_2 in H_2-air; right: 9.5 vol.-% CH_4 in CH_4-air; BR = 97%
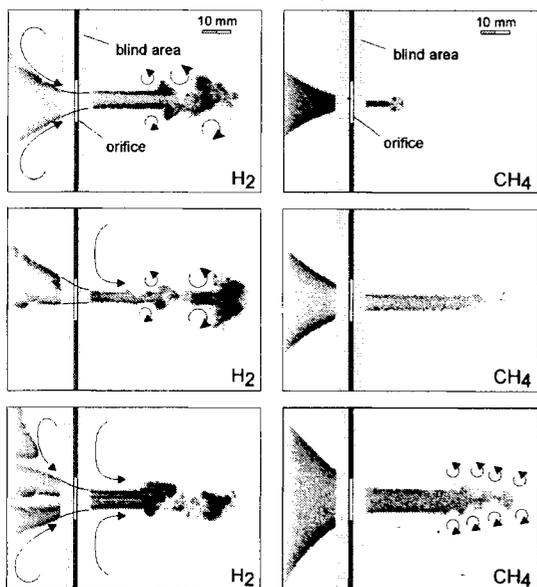

Figure 7: OH-radical distribution through the orifice; left: 12 vol.-% H_2 in H_2-air; right: 9.5 vol.-% CH_4 in CH_4-air; BR = 97%

As the limiting orifice diameter for the stoichiometric methane air mixture was shown to be 20 mm, no ignition of this mixture occurs using the 16 mm orifice. In figure 7 (right side) a quite similar parallel radical free jet, compared to the hydrogen flame, penetrates into the second chamber, being accelerated up to the order of magnitude of sonic speed. After about 10^{-4} sec, the parallel jet breaks up but no ignition in the second chamber takes place, due to the high minimum ignition energy, necessary for methane–air mixtures.

COMPARISON WITH CHEMKIN-II CALCULATIONS

Calculations with the Chemkin-II code from Sandia National Laboratories were performed, to get information about the time scale of the reaction process, the ignition delay, and the life time of important radicals. This chemical kinetics code considers four elements (H, C, N and O), 51 species and 235 chemical reactions. In figure 8 the normalised species concentrations for the zero-dimensional adiabatic calculation of a 12 vol.-% hydrogen-air flame is plotted. The initial temperature of 950K is approximately the minimum ignition temperature for this mixtures.

After a certain ignition delay the radical concentration increases and decreases again before the temperature reaches its adiabatic maximum. But as long as the process is not cooled down, the OH, O, and H radicals are in a thermal equilibrium with the generated H_2O and left O_2 molecules. Therefore, the detected OH radicals in figure 7 between the orifice and the spots of ignition not necessarily indicate a reacting zone. In this case they indicate the path of the compressed hot exhaust gas.

Similar calculations of a stoichiometric methane-air mixture have been performed. Before the temperature trace has its point of infection, any reactions containing C-atoms (except CO and CO_2) are completed. The O, H and OH radicals show a similar behaviour compared to the calculations with hydrogen as fuel.
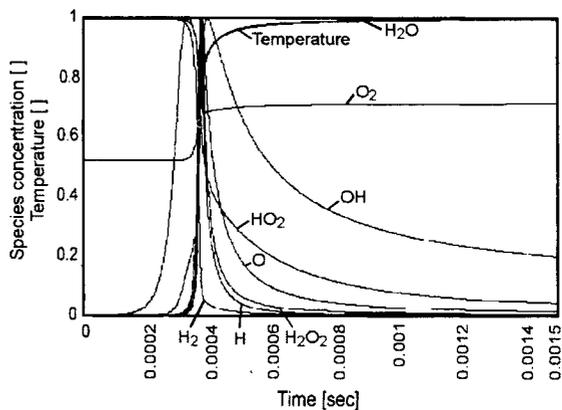

Figure 8: Chemkin calculations of a 12 vol.-% H_2 in H_2-air mixture; initial temperature T = 950 K

The previous presented tests at the L.VIEW and the PuFlaG facilities have shown, that burned gas is blown through the orifice for a long time (up to 800 ms in figure 1), before ignition in the second chamber occurs. Hence, some of the radicals transported into the second chamber are mixed with unburned gas before an ignition occurs. In figure 9 the time-series of a similar calculation as in figure 8 is plotted, where the initial unburned gas is mixed with 10 vol.-% burned gas (H_2O, N_2, O_2) together with the OH-, HO_2-, O-, H_2O_2-, and H-radicals. This yields to a decrease of the ignition delay, which explains the high reactivity of the gas mixture in the second chamber. In the case of methane the calculations with the initial fuel-air, mixed with exhaust gas inside the second chamber shows the same tendency. 10 vol.-% burned gas (CO_2, CO, H_2O, N_2, O_2, OH-, HO_2-, O-, H_2O_2-, and H-

radicals) mixed with stoichiometric methane-air leads to a decrease of the ignition delay of approximately 50%.
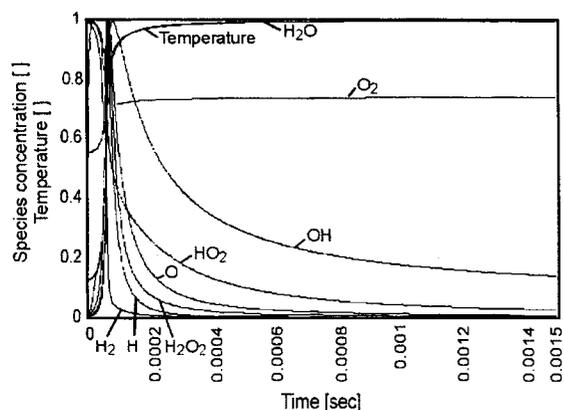

Figure 9: Chemkin calculations of 12 vol.-% H_2 in H_2-air mixed with 10 vol.-% of the burned gas from the calculation of figure 8; initial temperature T = 950 K

CONCLUSION

The influence of highly blocking obstacles on free propagating methane- and lean hydrogen-air flames has been investigated. It has been shown, that flame quenching effects, due to the high acceleration of the gas, lead to an accumulation of free radicals behind the obstacle. The mixing of these radicals with the unburned gas leads to a highly explosive mixture, which endangers building integrity.

The mechanism of hot-jet ignition, taking place at more sensitive mixtures can directly lead to a detonation of the fuel-air mixture. For a successful numerical simulation of these processes a more detailed quantification of these processes has to be made.

ACKNOWLEDGEMENT

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