

# SPONTANEOUS IGNITION PROCESSES DUE TO HIGH-PRESSURE HYDROGEN RELEASE IN AIR

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## ABSTRACT

Spontaneous ignition processes due to the high-pressure hydrogen releases into air were investigated both experimentally and theoretically. Such processes reproduce accident scenarios of sudden expansion of pressurized hydrogen into the ambient atmosphere in cases of tube or valve rupture. High-pressure hydrogen releases in the range of initial pressures from 20 to 275 bar and with nozzle diameters of 0.5 – 4 mm have been investigated. Glass tubes and high-speed CCD camera were used for experimental study of self-ignition process. The problem was theoretically considered in terms of contact discontinuity for the case when spontaneous ignition of pressurized hydrogen due to the contact with hot pressurized air occurs. The effects of boundary layer and material properties are discussed in order to explain the minimum initial pressure of 25 bar leading to the self-ignition of hydrogen with air.

## 1.0 INTRODUCTION

Hydrogen is successfully used as energy carrier in many different applications. Accidental hydrogen release from pipe systems are one of the main hazards that occur in the handling of pressurised hydrogen. The accidental H<sub>2</sub> release from the system should be detected fast and as safety consequence the main supply tank should be closed. So the released amount of hydrogen is limited and the release conditions are unsteady. But the generated hydrogen cloud can be ignited subsequently by an external ignition source. For low initial storage pressure, up to 16 bar, it was showed experimentally [1] that for a given amount of hydrogen a distinct ignition time and ignition position exists for the generation of a maximum pressure wave due to a "local explosion" in the free jet. In case of a sudden hydrogen release from a high pressure initial state the possibility of a self ignited jet fire is present [2] [3] [4]. Currently no systematic studies are available on the hazard potential of transient releases of hydrogen from high pressures with a spontaneous or forced subsequent ignition. In this study, the free hydrogen jet release from pipes with residual overpressure is experimentally simulated by using two different start up release conditions. The first procedure is a fast valve opening to produce a hydrogen free jet. The parameters in these experiments with a forced spark ignition are: circular release opening with inner diameters of 3 mm, 4 mm and 10 mm, initial reservoir pressures of up to 200 bar, and a reservoir volume of 0.37 dm<sup>3</sup>. In the second release configuration an additional rupture disc is installed in the exhaust pipe, which leads to a sudden hydrogen release in combination with shock wave generation from the ruptured disc [5]. The parameters in the experiments with self ignited jet fire are: circular release opening with an inner diameter of 4 mm with different extension pipe lengths after the rupture disc, different break points of the disc, initial reservoir pressures of up to 200 bar, and a reservoir volume of 0.37 dm<sup>3</sup>. The ignited hydrogen jets were recorded with a high speed camera, and the resulting pressure and thermal loads to the environment were investigated under variation of ignition position, ignition point in time and initial vessel pressure. Goal of this work is to

quantify the possible hazards arising from external ignited or auto ignited free transient hydrogen jets.

## 2. Experimental set-up

The facility for the generation of a defined and transient horizontal hydrogen jet from a high pressure reservoir is shown in Figure 1. Via a feed line valve (a) the cylindrical storage vessel (d) with a volume of  $0.37 \text{ dm}^3$  is filled with highly pressurized  $\text{H}_2$ . A helium driven needle valve DN 4 mm (c) with a valve opening time  $< 2 \text{ ms}$  works as leak opening tool in all experiments. The initiation time of this valve is  $16 \pm 0.3 \text{ ms}$ . Cylindrical tubes connected to the valve were used as release nozzle (d). Optionally cylindrical tubes with a rupture disc (aluminum foil) inside were installed in the experiments with auto ignition initiation, see inserted picture of the rupture disc holder in Figure 1 right. The  $\text{H}_2$ -jets released into the free environment were ignited after a distinct time in different positions along the jet axis via a high frequency electric arc (20 kHz,  $\sim 60 \text{ kV}$ ,  $\sim 10 \text{ W}$ ) (f). The dynamic overpressure resulting from the combustion of the released hydrogen was detected by five dynamic pressure sensors (PCB Type 113A31) in special adapters that were positioned in a line with a distance of 40 cm to each other. The pressure sensor line (e) is placed parallel in a distance of 50 cm to the jet axis (h). Two heat flux sensors (g) were positioned via lances along the jet axis (h). The hydrogen flow and combustion were observed by high speed shadow-schlieren-methods in different scales and techniques. The experiments were performed in a specific test chamber with a volume of approx.  $160 \text{ m}^3$ . The experimental facility was mounted on a rack in a way that the axis of the  $\text{H}_2$ -jet (h) had an unobstructed length in space of 4 m and a distance to the floor and the nearest wall of 1.6 m.

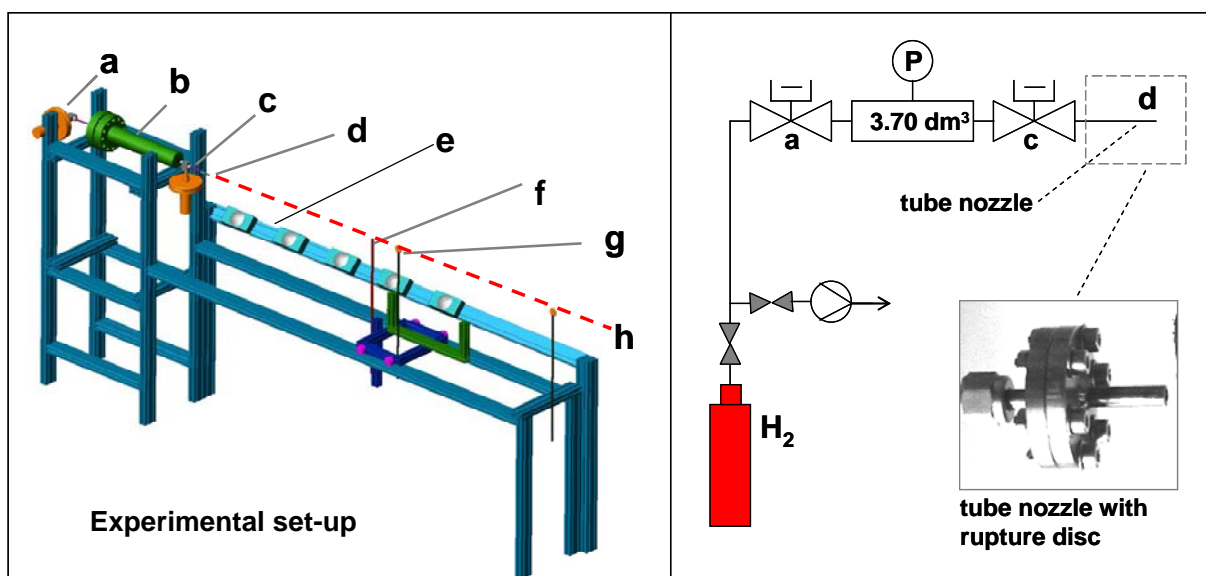


Figure 1: CAD-drawing of the experimental set-up and sketch and schematic of the experimental set-up.

In Table 1 the main experimental variables are listed, experiments with forced ignition via spark (no rupture disc in the nozzle pipe) were performed with 3 and 4 mm nozzles and initial overpressures of 100 and 200 bar. Additionally a 10 mm end nozzle was installed (this nozzle area is larger than the area in the leak valve DN 4 mm) to study the influence of an increasing release area. Experiments with rupture discs in the 4 mm nozzle pipe were performed with different initial overpressures of up to 225 bar and different extension pipe lengths between the disc and the nozzle exit. In these experiments the break pressure of the used aluminum rupture disc was clearly lower than the overpressures in the vessel and the small volume

between the rupture disc and the leak valve was filled with H<sub>2</sub> as recipient at atmospheric pressure. The extension pipe (nozzle) downstream of the rupture disc is open to the atmosphere. The nozzle exit was scanned with a photodiode to identify the luminescence of reacting hydrogen. A more detailed description to the rupture disc behavior and the experimental set up is given in [5], where the spontaneous ignition processes due to high-pressure hydrogen releases in air were studied.

Table 1: Overview of the main experimental variables.

| Nozzle / mm | Pressure / bar | Rupture disc | Extension pipe behind the rupture disc / mm | Spark ignition | Ignition distance/ cm |
|-------------|----------------|--------------|---|----------------|-----------------------|
| 3           | 100 / 200      | no           | --  | yes            | 25 to 100             |
| 4           | 100 / 200      | no           | --  | yes            | 25 to 150             |
| 4           | up to 225      | yes          | 5 to 120                                    | no             | --                    |
| 10          | 200            | no           | --  | yes            | 40 to 100             |

### 3. Release conditions

To compare the hazard potential of ignited and auto ignited free transient hydrogen jets in air the release conditions in both cases are important. The effluent formula of SAINT-VENANT and WANTZELL in combination with a linear nozzle form factor [1] can describe the pressure decay in the vessel during the H<sub>2</sub> release. In Figure 2 the comparison of measured and calculated pressure decay for the 4 mm nozzle and initial pressure of 200 bar in the vessel is shown for the start up of the H<sub>2</sub> release. In the pressure history of the vessel for a rupture disc experiment the stagnation point before the failure of the disc (burst pressure ~ 160 bar) is not detected, only a refraction shock wave that propagates against the flow in the vessel and is reflected there from wall to wall. All three plotted pressure decay curves are in good agreement and the average mass flow rate in the first 10 ms is 66 g /s. After 64 ms the storage tank is half empty (100 bar) and the complete effusion time is roughly 500 ms, see small diagram in Figure 2.

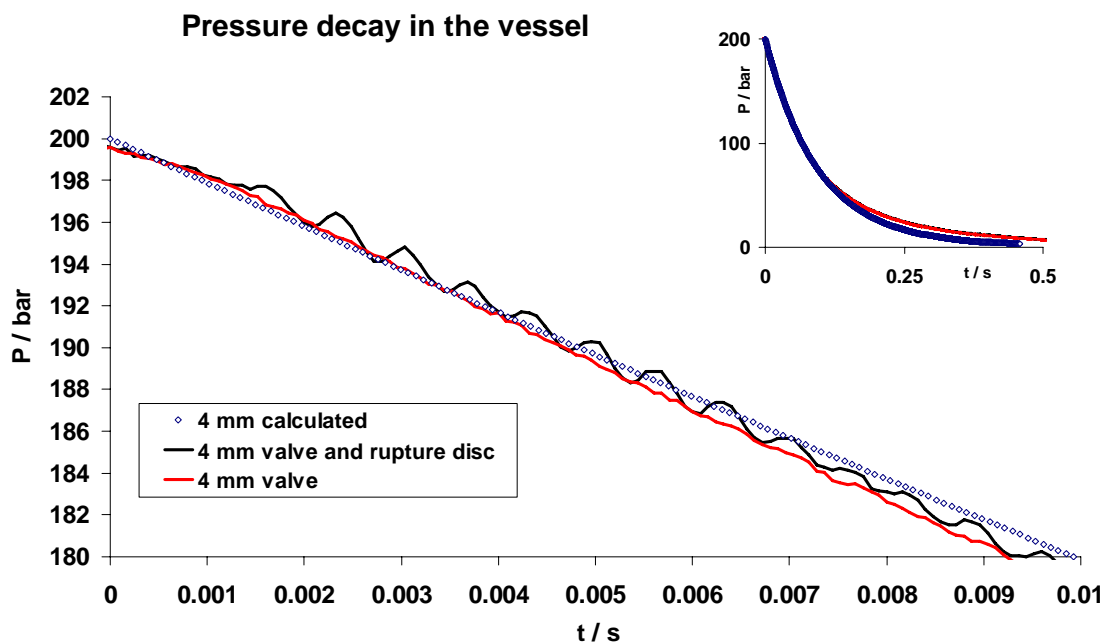


Figure 2: Comparison of measured and calculated pressure decay in the vessel (4 mm nozzle, 200 bar initial pressure).

In Figure 3 the development of the H<sub>2</sub>-jet on the nozzle exit is shown for the same examples shown in Figure 2. As scale for the shadow-schlieren picture sequences the caliper gauge in the first picture on the left is opened 50 mm. The left side shows the H<sub>2</sub>-release without a ruptured diaphragm between the valve and the nozzle exit, the frame rate is 10000 f/s. In the beginning of the flow the head of the horizontal axis symmetrical jet propagates with super sonic velocity, visible due to the weak mach cone, later only weak pulsating shock waves from the nozzle exit are visible. The picture sequence on the right demonstrates the influence of a rupture disc in the flow path from the leak valve to the nozzle exit, the frame rate is 50000 f/s. Initially a relatively strong shock wave propagates spherically from the nozzle exit, behind which the released H<sub>2</sub> also spread spherically at first. Later the shock wave propagates faster than the released gas jet. In this example the released H<sub>2</sub> is ignited spontaneously inside the 42 mm long extension pipe behind the rupture disc which leads to a jet fire. Due to the expansion of the reacted gas the released gas cloud on the right side looks larger in volume compared to the left side at the same time.

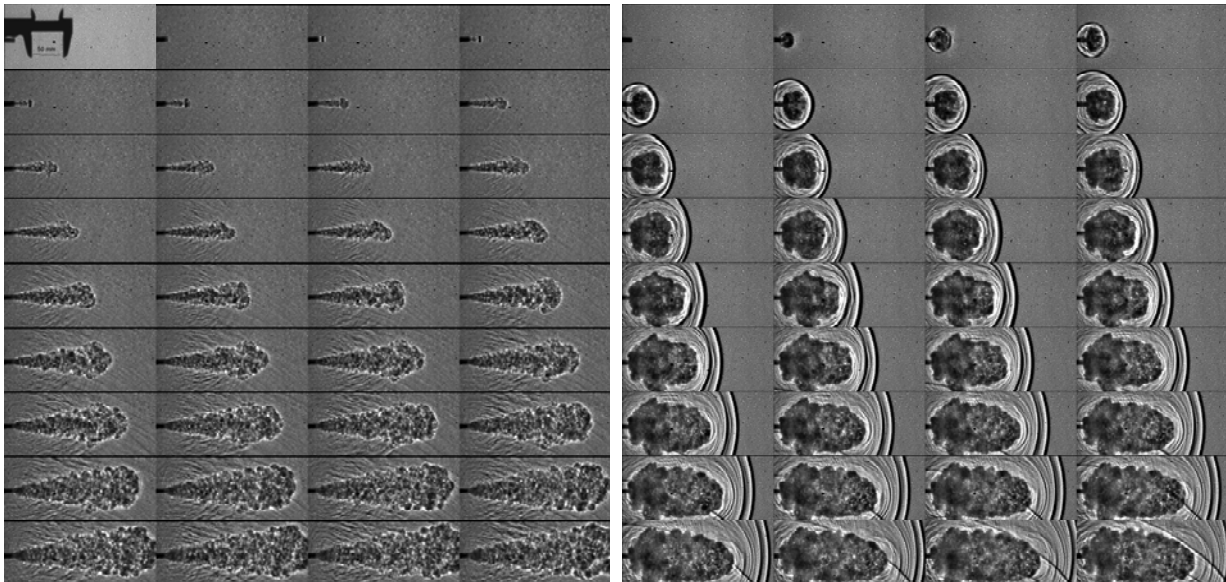


Figure 3: Development of the H<sub>2</sub> jet on the nozzle exit (4 mm nozzle, 200 bar initial pressure) in experiments with (right) and without (left) rupture disc in the flow path from the leak valve to the nozzle exit.

The influence of the rupture disc in the flow path between the leak valve and the nozzle on the mass flow rate and the release time is negligible. The main difference in the release conditions between the two release scenarios investigated is the shock wave which is transmitted from the breaking rupture disc. This shock wave propagates in front of the H<sub>2</sub>-flow in the nozzle tube.

### 3. Experimental results for ignited unsteady free jets

#### 3.1. Conditions for spontaneous ignited transient free jets

A detailed overview of experimental observations for controlled and unexpected spontaneous ignition of hydrogen releases is presented in [6], A strong influence of the local peculiarities and the test procedure and facilities on the experimental results is described there. The initial

pressure conditions for spontaneous ignited transient free jets observed in the presented facility are plotted for different lengths of the extension tubes in Figure 4. For this configuration with an equal diameter of 4 mm for the nozzle tube and the rupture disc all spontaneous ignition events lead to a jet fire. The spontaneous ignition takes place inside the extension tube downstream of the rupture disc. The minimum initial pressure for a spontaneous ignition is 25 bar using a 42 mm long extension tube [5].

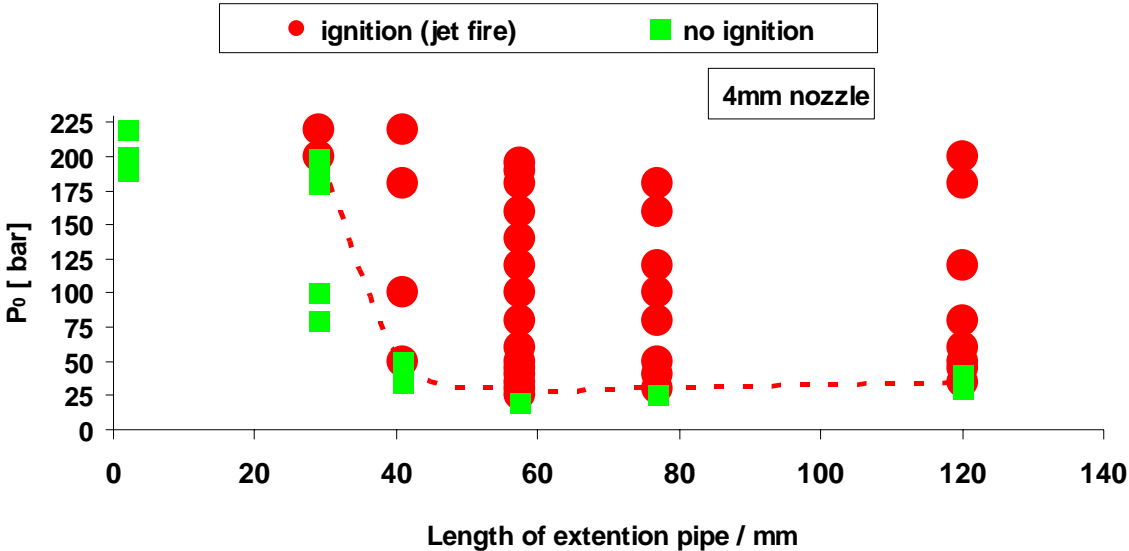


Figure 4: Critical conditions for spontaneous ignitions inside the extension tube downstream the rupture disc.

The location inside the extension tube downstream the rupture disc is the earliest possible position for an ignition of the released hydrogen. As can be seen in Figure 3, the shock wave from the ruptured disc leaves the nozzle exit in front of the hydrogen flow. This pressure wave was detected by the pressure sensors P1 to P5 on the sensor line positioned parallel to the jet axis. Later the complete released hydrogen burns, this combustion generates a second overpressure wave which is stronger than the first one in all cases.

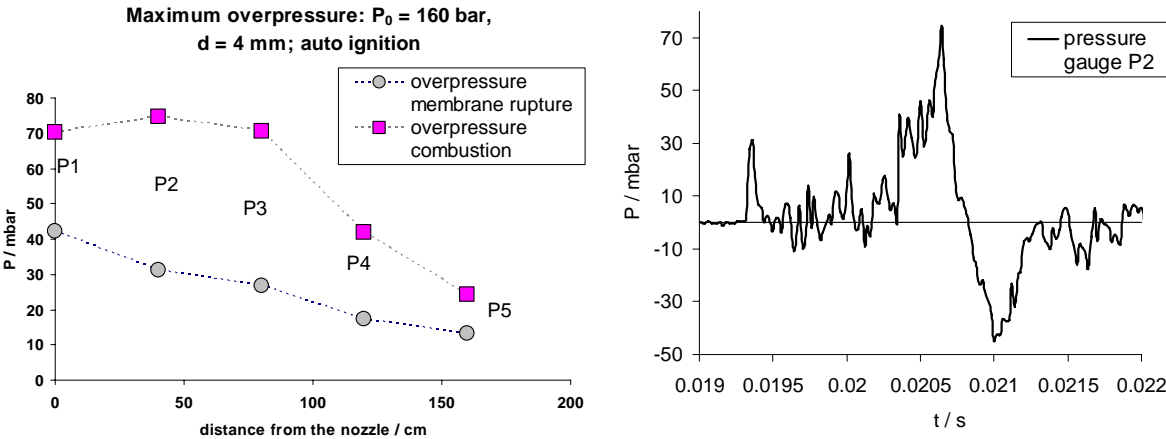


Figure 5: Example for the combustion pressure amplitude of a spontaneous ignition test. Left: Maximum amplitudes measured on the gauge line P1 to P5. Right: Pressure history of gauge P2.

In Figure 5 a typical measured pressure curve for the disc rupture and the combustion of the hydrogen jet is shown for an example with auto ignition. The highest amplitude from the

ruptured disc is detected by gauge P1, which is the closest to the nozzle exit, while the highest overpressure resulting from the burned jet is detected by gauge P2.

### 3.2. Conditions for forced ignited transient free jets

In this test the released unsteady jets were intentionally ignited by a spark. The ignition position lies on the jet axis in all experiments but the distance to the nozzle was varied. The ignition time was set variable with an accuracy of 0.33 ms and the minimum duration of the high frequency spark was 5 ms. Optical high speed observation of the H<sub>2</sub>-flow and the ignition process of the clouds was used to identify the real ignition time and the position of the jet head in the flow. For every given amount of hydrogen a distinct ignition time and ignition position exists for the generation of a maximum pressure wave due to a "local explosion" in the free jet [1]. The plot on the left side in Figure 6 shows as an example for constant ignition distance of 50 cm, 4 mm nozzle and 200 bar initial storage pressure the measured combustion overpressure recorded by center gauge P3. The scale for the ignition delay time includes the 16 ms induction time of the leak valve. The earliest possible ignition time in 50 cm distance is 6.5 ms (22.5 ms) after the beginning of the release. The time span for an ignition under generation of a strong pressure wave due to a "local explosion" in the free jet lies in this example between 8.9 and 10.5 ms. In further ignition distance the typical time range to produce the maximum pressure load is wider and a clear maximum is detectable. The plot on the right side in Figure 6 shows the maximum measured overpressures for different ignition positions and nozzle diameters for 100 and 200 bar of initial pressure. For each nozzle diameter and initial pressure state an ignition distance exists in which the ignition leads to the maximum combustion overpressure.

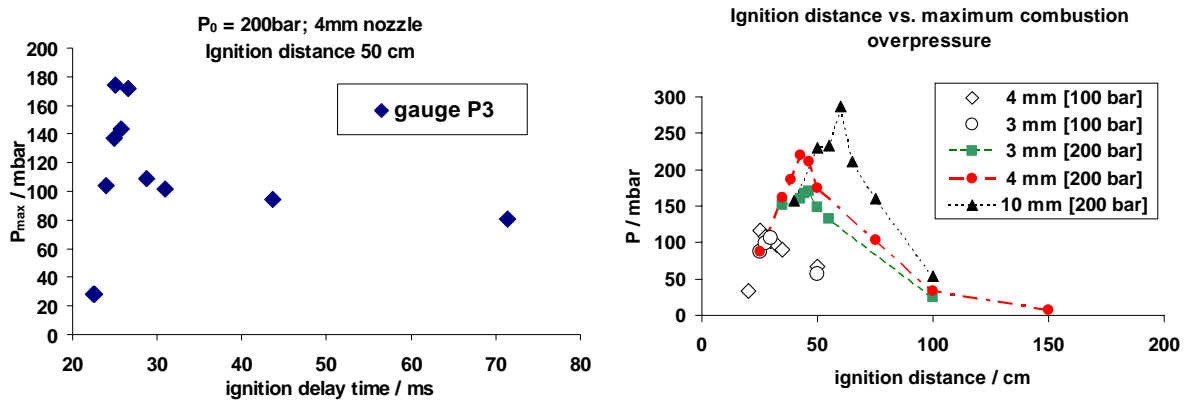


Figure 6: Left: dependence of the ignition delay time on the generated combustion overpressure. Right: maximum measured combustion overpressures for different ignition distances.

The time span for a possible forced ignition on the jet axis of the released hydrogen jet is in the range of the release time. Close to the release nozzle the leading head of the flow is not ignitable by a spark, in such a narrow ignition position the jet only ignites late in time when the release flow decreases significantly and the hydrogen concentration becomes ignitable. In a far ignition position the spark ignites only a lean mixture and the burning velocity is in a range of the flow velocity of the jet in this position, when the flow decreases the jet burns in counter-flow direction up to the nozzle. Due to the start-up process of the head of the flow in combination with the transient release conditions a highly reactive hydrogen / air cloud inside the turbulent jet is formed. The volumetric size and the position of this highly reactive



hydrogen / air cloud are changing during the transient release. The shadow picture sequence of Figure 7 shows the ignition process inside this highly reactive hydrogen / air cloud. The first combustion phenomena is visible 8 cm behind the ignition position, up to this position the measured burning velocity is in the range of the sound speed in air, including the flow velocity in co-flow and counter-flow direction.

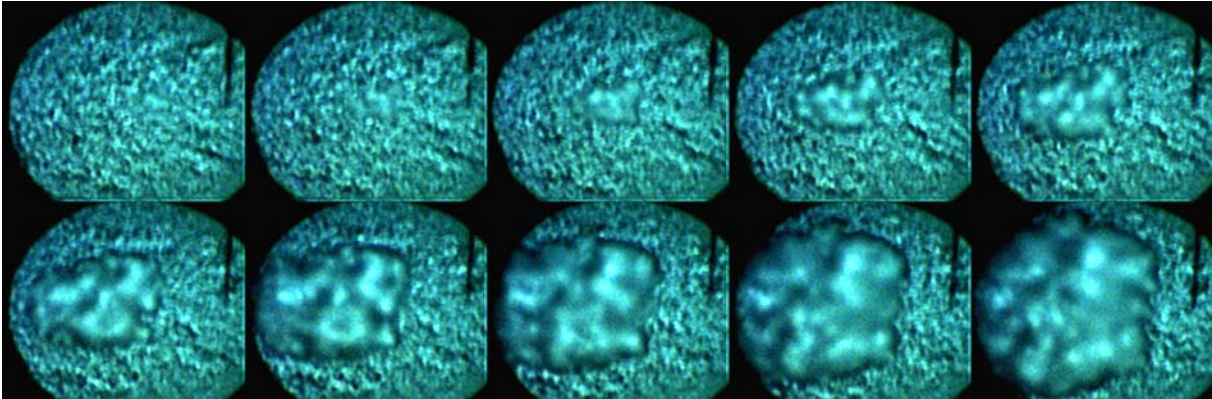


Figure 7: High speed shadow images of the ignition process inside the turbulent transient H<sub>2</sub> free jet.

**3.3. Thermal energies from ignited transient free jets**

For measurements of the transmitted thermal energy of the ignited transient H<sub>2</sub>-free jet copper disks were positioned as heat flux sensors at different positions along the jet axis in the experiments. The integral heat flux sensors consist of a copper disk with thermocouples in the center, this device was already described in [1]. Due to the fast fluctuation of the mass flow rate, the instationary flow and combustion conditions only integral thermal loads in the burning jet were measured. In [1] it was also shown for lower initial release overpressure that an early and close to the nozzle ignited free jet generates the maximum thermal load for its ambience. The combustion duration of the released hydrogen lies in the range of the effusion times. The maximum integral heat flux measured on the jet axis decreases almost linearly with increasing distance to the nozzle. The spontaneous auto ignition of the first released H<sub>2</sub> inside the tube downstream of the rupture disc is absolutely the earliest ignition position in the released jet.

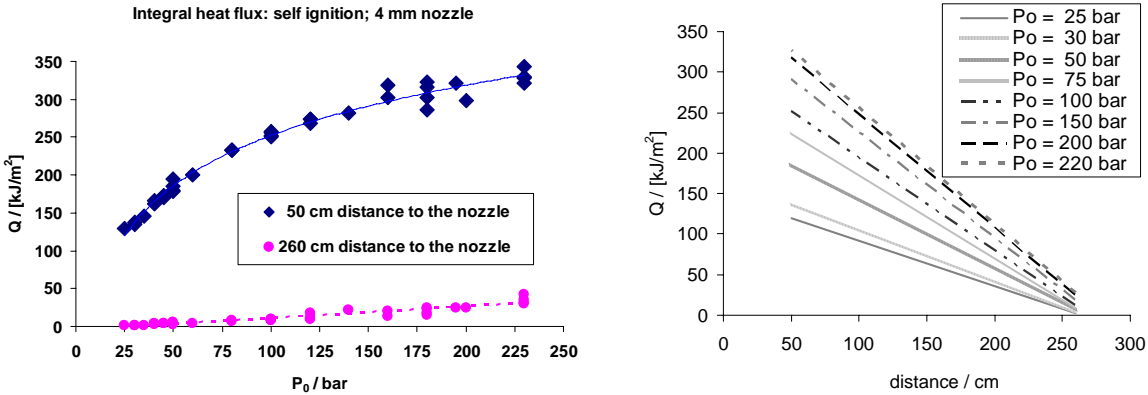


Figure 8: Left: measured integral thermal loads versus initial release overpressure from experiments with spontaneous ignition in the 4 mm nozzle. Right: linearization of the maximum thermal loads along the jet axis.

Figure 8 left shows measured integral thermal loads versus initial release overpressures from experiments with spontaneous ignition in the 4 mm nozzle. In a distance of 2.60 m from the nozzle exit the measured integral thermal loads have a quasi linear dependency on the initial overpressure in contrast to a position in 0.5 m distance to the nozzle. Under the assumptions that the maximum integral heat flux measured on the jet axis decreases almost linearly with increasing distance to the nozzle [1] a conservative linearization of the maximum thermal loads along the jet axis is feasible, see Figure 8 right. The conservative linearization of the possible maximum thermal loads on the jet axis in Figure 9 is not valid for the region close to the nozzle. In this region no measurements were performed. The size (50 mm in diameter) of the heat flux sensors used is not completely covered by the jet, and the sensor width of 2 mm is not negligible compared to the small nozzle exit. In Figure 9 the measured thermal loads along the jet axis of experiments with forced ignition and initial release pressures of 100 bar and 200 bar are plotted. The measured thermal loads decrease almost linearly with increasing distance to the nozzle. The values for the 3 mm nozzle lie partially below the data of the 4 mm nozzle. The values from the 10 mm nozzle, which has a comparable  $H_2$  mass release to the 4 mm nozzle, shows in 50 cm nozzle distance 100  $\text{kJ/m}^2$  less thermal load than the 4 mm nozzle. For 100 bar and 200 bar of initial release pressure the measured thermal loads in the tests with auto ignition are in good agreement with the values for forced ignition, and the conservative linearization covers all data points.

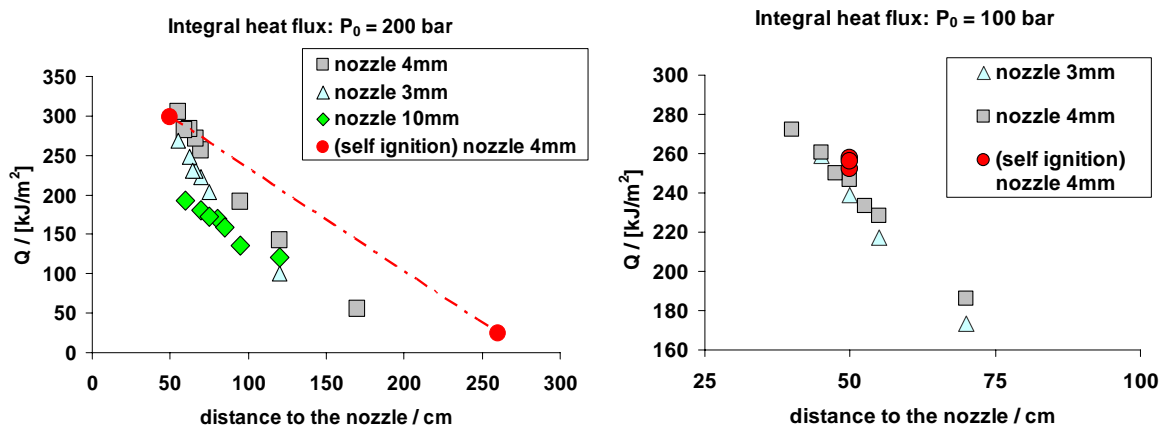


Figure 9: Measured thermal loads along the jet axis of experiments with forced ignition.

## 4. Hazard potentials

### 4.1. Hazard potential of pressure loads

The combustion experiments with forced spark ignition have shown that hydrogen that is effusing as free jet is quickly diluted with air. The time span in which the gas cloud is ignitable is almost identical with the effusion time of the gas from the vessel. For many ignition times and ignition positions the ignition of the unsteady free jet produced only a local combustion with no detectable pressure wave. In contrast to this, an ignition of the free hydrogen jet shortly after it has reached its maximum effusion rate can lead to a local explosion with detectable pressure waves. In the experiments with a rupture disc in the nozzle pipe and a spontaneous ignition inside the nozzle it was shown that the combustion of the released  $H_2$  leads also to a local explosion with a detectable pressure wave. To analyze the hazard potential of these pressure loads the orientation of the gauge to the incoming wave is important. All pressure waves presented in this work were measured in a reflected orientation on a pressure gauge line parallel to the jet axis in a distance of 50 cm. In Figure 10 the



maximum measured overpressure amplitudes for the experiments with the 4 mm nozzle, forced spark ignition and the two initial release overpressures investigated are plotted. Additionally the measured pressure amplitudes resulting from the self ignition tests, according to Figure 4, are presented. The plotted maximum amplitude is the highest value measured by one of the five gauges of the pressure sensor line, in the most cases P2 or P3. If no ignition occurred only the pressure wave from the ruptured disc is used as maximum, see Figure 5.

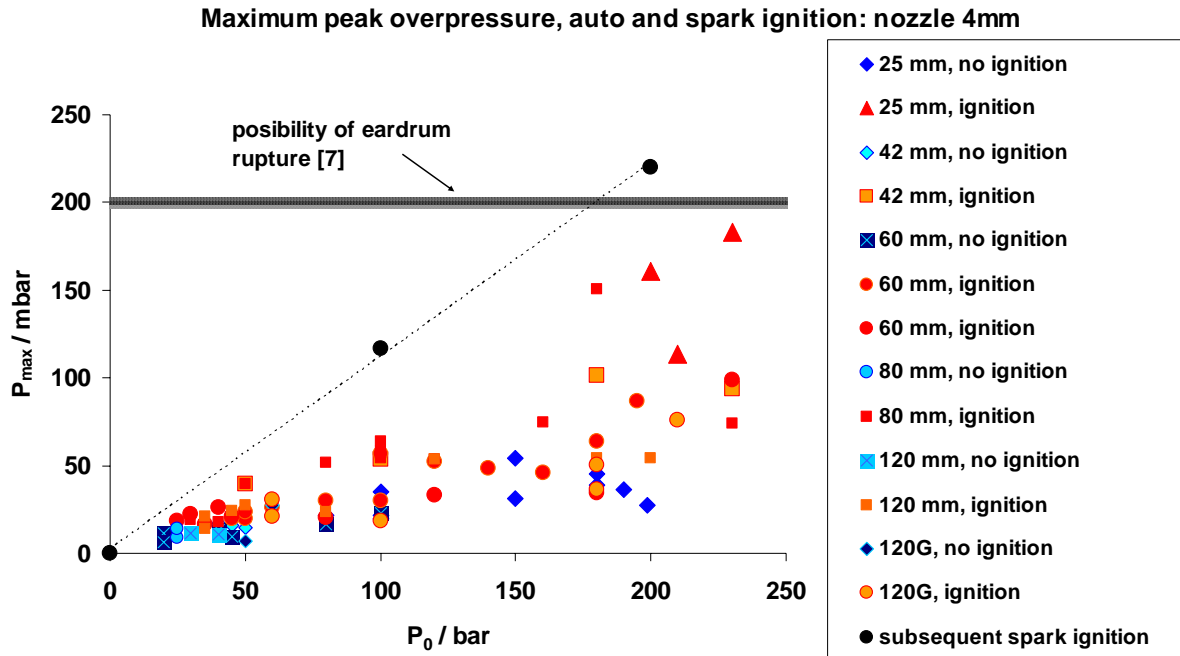


Figure 10: Maximum measured overpressure amplitudes from the 4 mm nozzle experiments, measured in reflected orientation in 50 cm distance to the jet axis.

In Figure 10 the lower threshold for a possible damage to the ear-drum at exposure durations of the positive peak overpressure of 1.5 ms, according to [7], is depicted for comparison. The mean duration of the positive pressure phase was determined to about 0.5 and 1.5 ms for all initial pressures investigated in the experiments. Only the value for a release overpressure of 200 bar with an optimized ignition distance and ignition delay time lie in the region for possible irreparable ear injuries. The data also shows the possibility for a relatively strong overpressure generation if the ignition of the transient  $\text{H}_2$  jet takes place inside the release nozzle, especially for high initial release pressures. In a further distance to the jet axis the overpressure amplitude decays as a function of the reciprocal distance ( $1/r$ ) and the hazard potential of pressure loads decreases rapidly with increasing distance to the source [1].

#### 4.2. Hazard potential of thermal loads

An early ignition of the released hydrogen close to the nozzle produces the highest thermal loads [1]. The case of the spontaneous ignition of the first released hydrogen inside the nozzle pipe is the earliest possible ignition time, since almost the complete effusing hydrogen is burned and the combustion duration extends over the whole effusion time. Considering the effusion time of the hydrogen from the vessel as maximum combustion duration of the released hydrogen, and thereby as maximum exposure time of the thermal energy, from the extrapolated data [8] for different initial release overpressures the thermal load which may lead to second degree burns can be estimated. The intersection of these specific thermal load energies for injuries with the linear maximum possible thermal loads on the jet axis, depicted

in Figure 8, results in a specific critical distance diagram. In Figure 11 the critical distances to the nozzle for possible second degree burns of human skin, gained with this method, are plotted versus the initial release overpressures from the vessel with a volume of  $0.37 \text{ dm}^3$ . For an initial release overpressure of 25 bar the critical distance for possible second degree burns of human skin is 2 m, for 200 bar of initial release overpressure the critical distance is 2.5 m. The point of intersection of the straight lines in Figure 9 with the abscissa can be considered as the maximum thermal range of the ignited free jets, which is quite considerably less than 3 m.

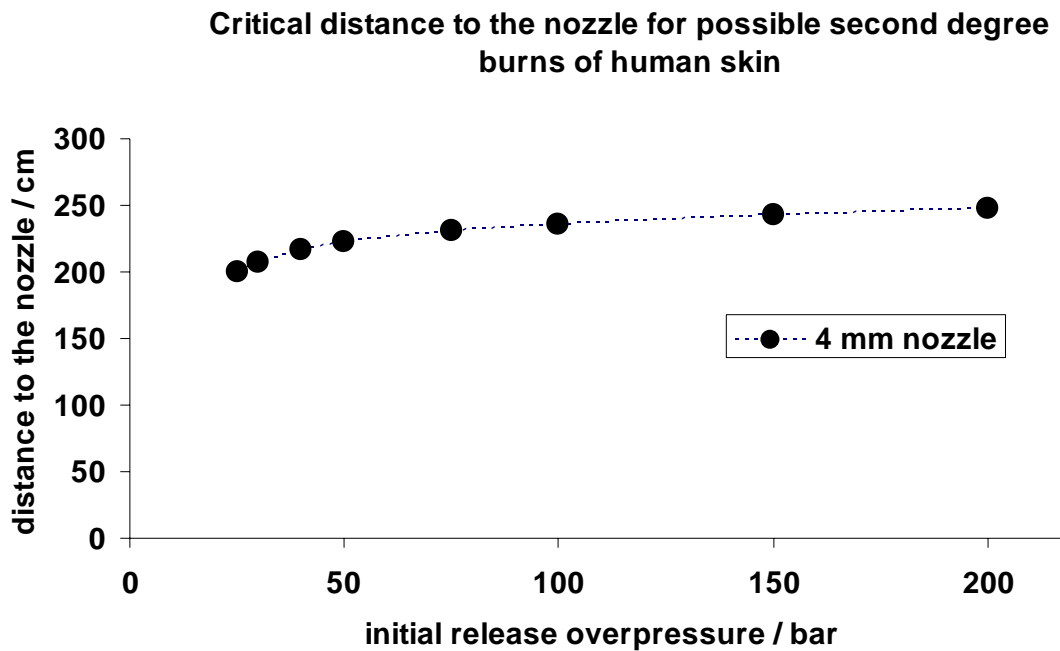


Figure 11: Critical distance to the nozzle for possible second degree burns of human skin.

## 5. Conclusions

In this study an accidental  $\text{H}_2$  release from a small high pressurized reservoir of  $0.37 \text{ dm}^3$  was investigated experimentally. Two different starts-up release conditions were simulated. In the first procedure a fast valve opens and different tubular release nozzles with diameters of 3, 4, and 10 mm produce a hydrogen free jet in air. In the second release application an additional rupture disc was installed in the 4 mm exhaust pipe. To study the combustion behaviours and the resulting hazard potential, the released  $\text{H}_2$  were intentionally ignited by a spark or a sudden release condition initiated a spontaneous auto ignition of the jet.

The rupture of an alumina disc inside the exhausts pipe can lead to a spontaneous auto ignition of the release  $\text{H}_2$  inside the extension pipe between the disc and the nozzle exit with a resulting jet fire. A minimum initial reservoir overpressure of 25 bar for a spontaneous ignition event was observed for an extension pipe length of 42 mm. The experiments show that the transition from the ignition inside the exhaust nozzle pipe to the full jet fire in the ambience generates a detectable overpressure blast wave due to a "local explosion" in the free jet. For 200 bar of initial release overpressure reflected amplitudes of 160 mbar in a distance of 50 cm to the jet axis were measured. Without rupture disc inside the exhausts pipe no spontaneous auto ignition was observed and the released  $\text{H}_2$  were than ignited by a spark. These experiments demonstrate that for an initial hydrogen reservoir overpressure a distinct

ignition time and ignition position exists for the generation of a maximum pressure wave due to a "local explosion" in the free jet. For 200 bar of initial release overpressure reflected amplitudes of 220 mbar in a distance of 50 cm to the jet axis were measured, this value reaches the upper limit for possible irreparable ear injuries.

The generated thermal loads were investigated systematically for both investigated ignition scenarios. A free jet that is ignited early and close to the nozzle generates the maximum thermal load for its ambience. The spontaneous auto ignition of the first released H<sub>2</sub> inside the tube behind the rupture disc is the earliest ignition situation of the released jet. The maximum thermal range of the ignited free jets investigated here is less than 3m. The possible hazard from burning of transient H<sub>2</sub> free jets concerning their thermal loads was evaluated for the example of second degree burns of the human skin. In this case the critical distances can reach up to 2.5 m, depending on the initial release overpressure.

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