NUMERICAL AND EXPERIMENTAL INVESTIGATIONS ON SELF-IGNITION PROCESS OF HYDROGEN GAS RELEASE

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Abstract

This paper describes hydrogen gas self-ignition as a result of the release from high-pressure chamber through the tube into the ambient air. Hydrogen is a very flammable gas with high energy density, small minimum ignition energy and wide flammability limits. Moreover, because of its high diffusivity in metals and small molecules it may cause leakages from storage systems. Pressurized hydrogen releases were found to ignite with no particular ignition source and it was the reason for this work. The condition for self-ignition of hydrogen gas discharge through the tube is a proper combination of the burst disk rapture, the tube length and diameter. This work consists of experimental and computational parts. Numerical simulation of hydrogen gas discharge through the tube is investigated in the first part. Calculations were performed using a commercial CFD code AVL FIRE. Investigated domain was composed of two cylindrical high-pressure (compressed hydrogen) and low-pressure (ambient air) vessels connected by the tube. The release tube had diameter D of 10 mm and length L of 10-100 mm. The experiments were carried out in a closed rectangular channel filled with ambient air where the high-pressure (2-17 MPa) hydrogen was injected through tubes with various diameters (d = 6, 10 and 14 mm) and lengths (L = 10-100 mm). The equipment used included: pressure sensors, photodiodes, ionization probes and high-frequency data acquisition system. Both numerical and experimental investigations showed the general tendency that the possibility of self-ignition increased with increasing initial pressure and the tube length. As a conclusion, ignition appears due to the rise of temperature of the mixture, which is caused by the diffusion of hydrogen on the contact surface with the air, heated by leading shock wave.

Keywords: *hydrogen release, hydrogen safety, hydrogen self-ignition, AVL Fire*

1. Introduction

Transience of crude oil prices, increasing fuel consumptions and decreasing oil reserves affect economy of many countries. New energy carrier, which can be independently produced, can solve this problem. One of the possibilities is hydrogen, known as fuel of the future. The advancements of hydrogen internal combustion engines and hydrogen fuel cells is fast. Hydrogen-fuelled vehicles are available for usual, untrained customers. Nevertheless, in comparison with other fuels hydrogen is one of the most dangerous. It is a very flammable gas with high energy density (LHV: 120 MJ/kg), small minimum ignition energy (0.02 mJ) and wide flammability limits (4-75% in air). Moreover, because of its high diffusivity in metals and small molecules it may cause leakages from storage systems or bursting installation elements as a result of hydrogen embrittlement. Suwa et al. in [11] described piping systems as one of the most possible reason of accidents. Pressurised hydrogen releases were found to ignite with no particular ignition source, even if initial temperature was under the auto-ignition one. Astbury and Hawksworth [1] established that Joule-Thomson expansion, hot surface ignition, compression ignition, diffusion ignition considered separately are not sufficient cause of the ignition. There is possibility that during hydrogen release two or more mechanisms act simultaneously.

In order to ensure safety, phenomenon of the spontaneous ignition during release from high-pressure chamber through the tube and its characteristics must be investigated. The first research concerned this incident were carried out by Wójcicki and Wolański [12]. They established the reason of ignition as a result of heating oxidiser (air or pure oxygen) by shock wave in adiabatic compression and mixing hydrogen with hot oxygen in contact surface. It was called diffusion ignition. Later, the critical pressure was found out to depend from geometric configurations – tube length, tube diameter, presence of obstacles, diaphragm. Xu et al. [14] made numerical simulations of diaphragm rapture and proved that maximal temperature is decreasing while time of disk rapture is increasing. In paper [4] Golovastov and Bocharnikov demonstrated the lack of self-ignition due to insufficient intensity of shock wave, as a result of too long opening time. It can be concluded that diaphragm rapture damps the shock wave. Dryer in [3] found out that the reason of ignition in this geometric configuration are shock waves reflecting from the walls of the tube and their interference. The most possible place for ignition kernel is the axis and the wall of tube. Xu and Wen [13] studied the influence of local contraction. When shock wave leaves the contraction it diffracts and its shape becomes spherical. It creates partially premixed layer between hydrogen and air due to the highly turbulent stream. As the result, the contraction of tube can facilitate the occurrence of ignition.

2. Numerical investigations

The aim of these investigations is to define the characteristic of hydrogen gas self-ignition and check the mechanisms mentioned above. Simulations of the cases where large pressure gradients, high velocities with shock waves and combustion appear are a great challenge. Results are compromise between computation time and precision. Calculations were performed using a commercial CFD code AVL FIRE, which enables the simulations of various types of kinetic problems with internal chemistry interpreter based on the same theory, which is implemented in CHEMKIN. Solver is based on the Finite Volume approach and uses the Reynolds-averaged energy, concentration and Navier-Stokes equations. Turbulence was modelled by means of k-epsilon model, which yields enough realistic predictions of flow, including heat transfer, combustion and can be widely adopted. According to FIRE tutorial [2] for y_+ higher than 11.63, the standard wall function was applied for wall treatment and heat transfer wall model.

The FIRE General Gas Phase Reactions Module was applied to enable the simulations of kinetic problem. The kinetic scheme of 23 chemical reactions with 11 chemical species (plus M – the chaperon) was used for simulations of air – hydrogen mixture combustion [7]. The General Species Transport Module was used to provide the required transport equation for chemical species. Compressible flow was considered.

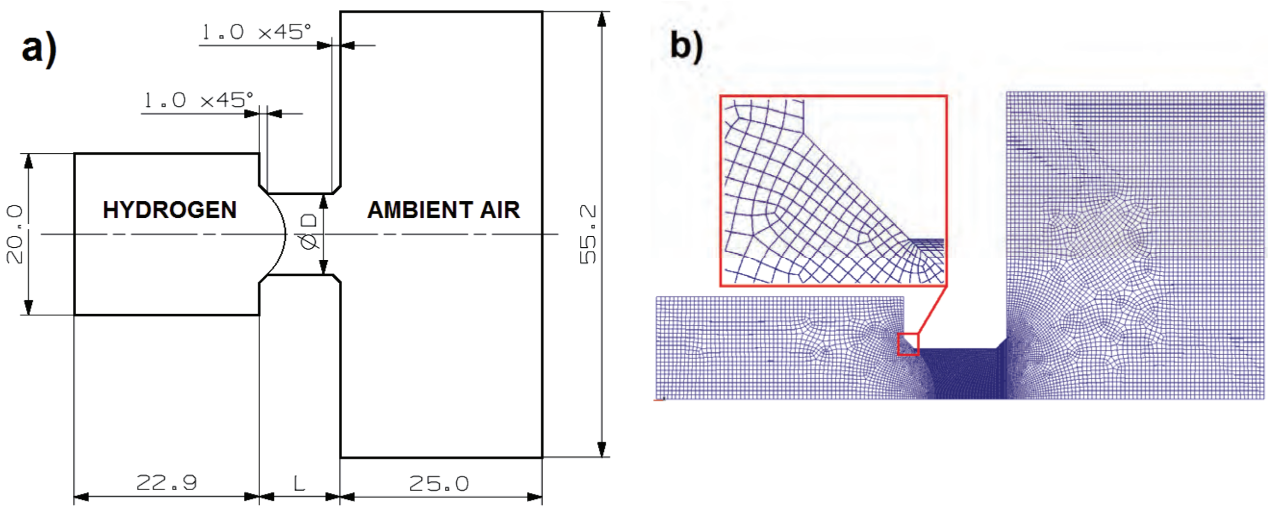


Fig. 1. Investigated geometry (a) and the example of the mesh model, L = 10 mm (b)

The computational domain were composed of two cylindrical vessels (high pressure with hydrogen and low pressure with ambient air) connected by the channel, see Fig. 1a. The release tube had the constant diameter D of 10 mm and length L of 10-100 mm. The influence of the rapture disk was not taken into account. Simulation geometry corresponded to the geometry investigated experimentally. In order to simplify calculations, the 2-degree section of cylinders was considered. Mesh was hexahedral with cell size from 0.08 mm to 0.4 mm, see Fig. 1b. Meshes consisted of 31 896 to 166 020 cells depending on the channel diameter and length.

The following boundary conditions were applied: stationary wall with the temperature of 300 K at all of the outer surfaces, symmetry at the axis and side surfaces. Calculations were started when the high-pressure vessel was filled with pure hydrogen (mole fraction of H_2 = 1 with temperature of 300 K and pressure according to simulated scenario) and low-pressure vessel with air (mole fraction of O_2 = 0.21, mole fraction of N_2 = 0.79 with initial temperature of 300 K and pressure of 1 bar).

2.1. Experimental facility

The experiments were carried out in a closed rectangular channel (0.1×0.1×1 m) filled with ambient air where the hydrogen (at 1–17.5 MPa and ambient temperature) were depressurized through different tubes: diameter d = 6, 10 and 14 mm, extension tube length: L = 10, 25, 40, 50, 75 and 100 mm. Fig. 2 presents the experimental facility scheme and photo. The equipment used in the facility included: pressure sensors (PCB), photodiodes, ion probes, data acquisition system (DAS) and fast camera (Photron SA1.1). Activation of the measurement system was coupled with electromagnetic valve activation and triggered by the staff. The mixture depressurized to the volume separated from the air-section with a diaphragm. The burst pressure (P_{burst}) was measured by PS1 pressure sensor placed near the diaphragm. The diaphragm was metal sheet of copper, brass or aluminium.

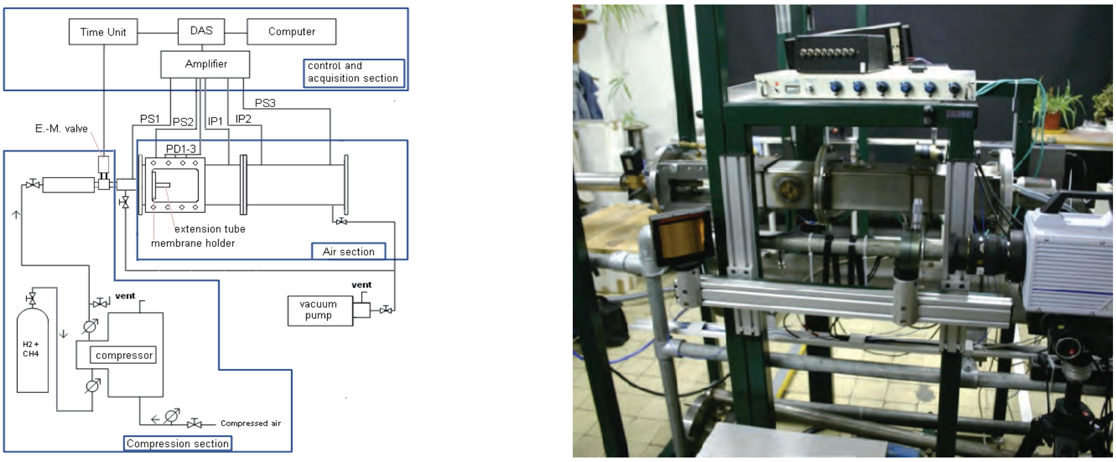
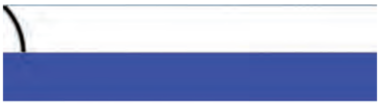
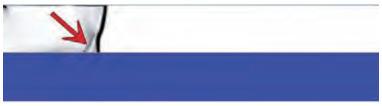
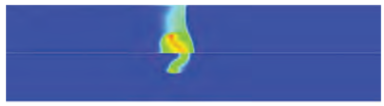
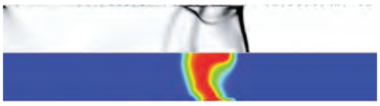
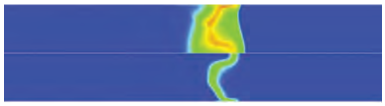


Fig. 2. Experimental facility scheme (left) and real view (right): PS – pressure sensors, IP – ionization probes, PD – photodiodes

3. Results

Numerically, thirty cases were investigated. According to [6] it was assumed that the indicated temperature above 1500 K and mass fraction of H_2O higher than 10^{-4} indicated ignition. However, in all investigated cases with the ignition the temperature rise to 2200-3500 K with H_2O mass fraction over 0.25 were observable. Density gradient, temperature, OH and H_2O mass fraction for the tube of length of 40 mm are representatively shown in Tab. 1. In all investigated cases, the ignition kernel appeared in the axis of the tube. On the contact, surface diffusive mass transfer occurred involving hydrogen and air, which was heated up by the leading shock wave. Ignition was caused by overlapped shock waves, which were reflected off the wall, and this is similar to the processes described in [3]. This phenomenon is presented in Tab. 1. by the density gradient contours for 5 and 8 µs (wave is marked by red arrow). Spherical shock wave generated by the diaphragm curvature reflecting from the tube wall diagonally spreads out to the axis, where it focuses. This collision causes temperature and pressure rising and, when the rise is sufficiently high the ignition. Combustion was not expected far from the outlet of the tube, because the shock wave diffracts, and loses strength.

Tab. 1. Example results for the tube of length 40 mm, pressure 9.5 MPa: temperature (200-3200 K), OH mass fraction (0-0.025), density gradient (200-10000 kg/m^4), H_2O mass fraction (0-0.25)

Time	Density gradient and H_2O mass fraction	Temperature and OH mass fraction
0 µs		
5 µs		
8 µs		
12 µs		
14 µs		
17 µs		
20 µs		
23 µs		

The characteristic of hydrogen gas self-ignition as a function of the tube length and the initial pressure of hydrogen was determined and it is presented in Fig. 3. The calculations were carried out with step of hydrogen initial pressure equal to 0.5 MPa and if ignition case occurred next to no ignition case, the mid pressure was assumed as the ignition limit. In conclusion, initial pressure necessary to ignite hydrogen decreases when the tube length increases. The approximated ignition limit asymptotically tends to the *Y*-axis. Lower pressure necessary for ignition in longer tubes is a result of longer mixing process of hydrogen with air and more complicated reflected shock waves configuration, which creates more possible ignition spots. In all investigated cases, self-ignition of hydrogen is always possible for initial pressure of at least 15 MPa. This fact is probably caused by the chemical reaction mechanism used. The further analysis need to be done.

Figure 4 presents temperature in the cases with ignition near to the diffusion limit for all investigated tube lengths. As far as in these cases ignition was assumed to occur at the end of the tube, the ignition delay time rises, when the channel length increases. It is the result of smaller initial pressure of hydrogen. For lengths: 20-40 mm and 80-100 mm, this order is changed and it may be

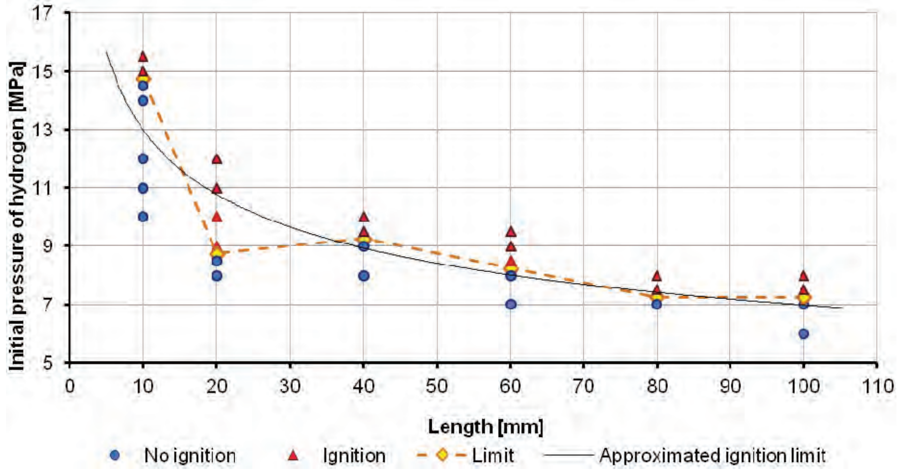


Fig. 3. Ignition limit of hydrogen

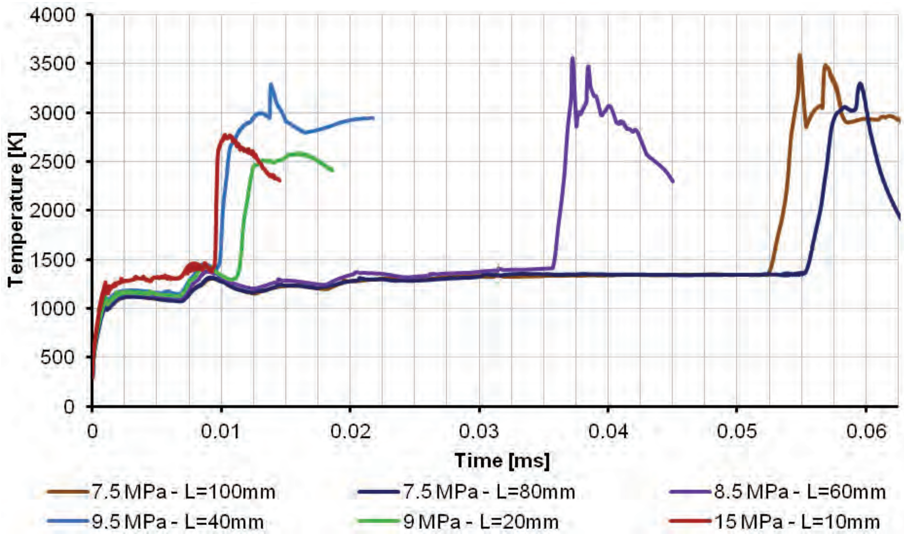


Fig. 4. Maximum temperature vs. time

the result of too small pressure accuracy. Fig. 5 shows the temperature in subsequent cases with ignition compared to no ignition case for tube L = 20 mm. For initial pressure of 12 MPa ignition occurred after 8 μs with maximum temperature of 3030 K, and for initial pressure of 9 MPa after 11.5 μs with maximum temperature of 2650 K. The ignition delay time decreases and maximum temperature increases with the initial pressure rise. For lower pressures, the temperature graph is more flat.

Totally, more than 150 experiments were conducted. The ignition, if occurred, was clearly indicated by the photodiodes just after the gas left the extension tube. Example sensors indications for cases with and without ignition of pure hydrogen flow are presented in Fig. 6. Pressure sensors PS2 and PS3 placed near the ends of the tube indicated oscillations for both cases with and without ignition. Oscillations correspond to successive shock wave reflections from the tube ends. For cases with ignition, both photodiodes and ionisation probes indicated distinct signal.

The cumulative diagrams obtained for cases with hydrogen flow through all the diameters are presented in Fig. 7. The experiments showed that the ignition of pure hydrogen is possible (or not) only above certain pressure. The possible reason for this stochastic ignition feature is different membrane breaking process, which affects the initial turbulence conditions of the flow. Similar behaviour has already been recorded by several researchers [5, 6, 8-10]. It was also observed that for burst pressures up to 17.0 MPa and 25 mm tube length ignition did not occur for any investigated diameter. For the tube length of 40 mm the burst pressure required for ignition seems to increase

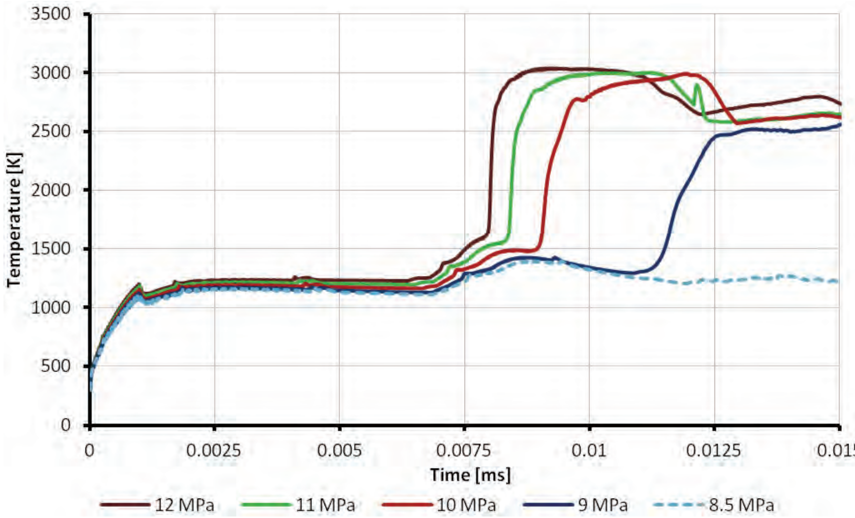


Fig. 5. Calculated temperature in subsequent cases with ignition compared to no ignition case, L = 20 mm

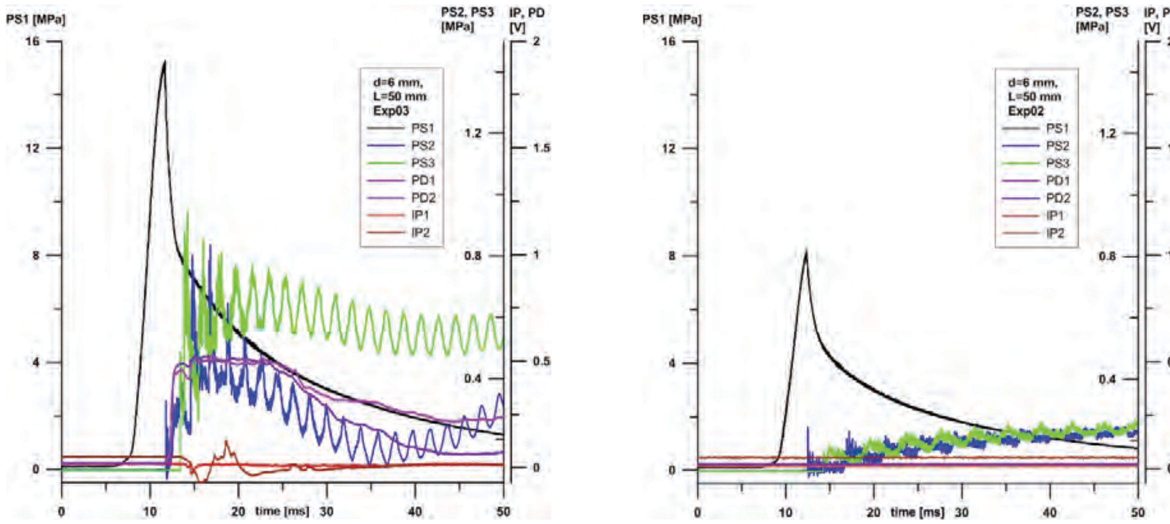


Fig. 6. Sensors indications for cases d = 6 mm, L = 50 mm with ignition (left, P_{burst} = 15.3 MPa) and without ignition (right, P_{burst} = 8.4 MPa)

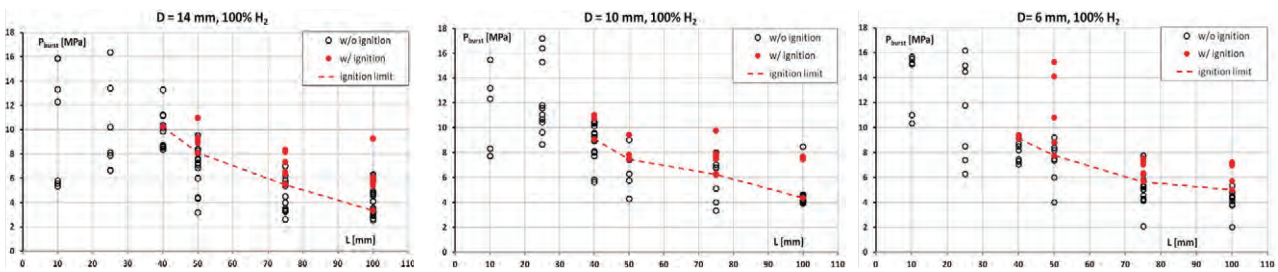


Fig. 7. Cumulative diagrams for pure hydrogen flow through tube with d = 14, 10 and 6 mm and various tube lengths

faster than for longer tubes. It allows concluding that for this specific experimental setup and geometry used there is some kind of limiting tube length that will not enable the ignition. Based on Fig. 8 the tube diameter seems to have undetermined influence on the ignition limit for pure hydrogen flow ignition. The comparison between numerically and experimentally obtained ignition limits are presented in Fig. 8. Additionally, during the experiments fast camera and optical schlieren system were used. Fig. 9 presents the images obtained for the case with and without ignition. The images clearly show leading shock wave, mixing front and turbulence intensity. Generally, it is very difficult to distinguish the images with and without ignition and the differences are very slight in turbulence, intensity just after the hydrogen leaves the tube. This feature is noticeable in Fig. 9 for t = 0.0740 ms.

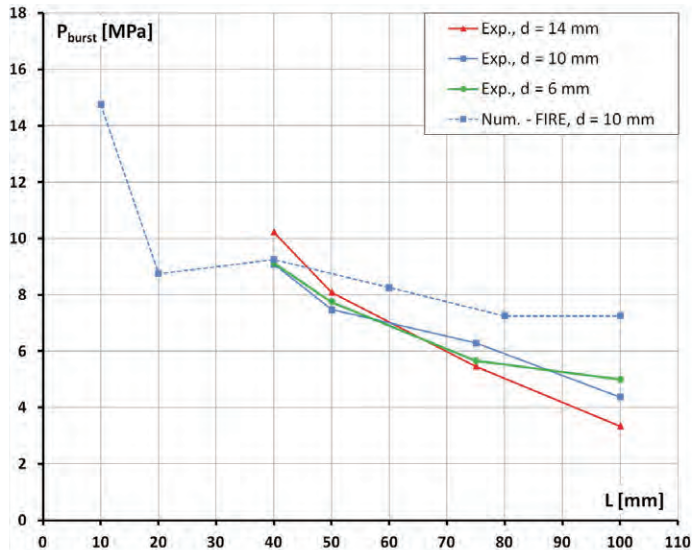


Fig. 8. Cumulative diagram of experimental self-ignition limits of hydrogen flow through tubes with various lengths and diameters and comparison between experimental and numerical results

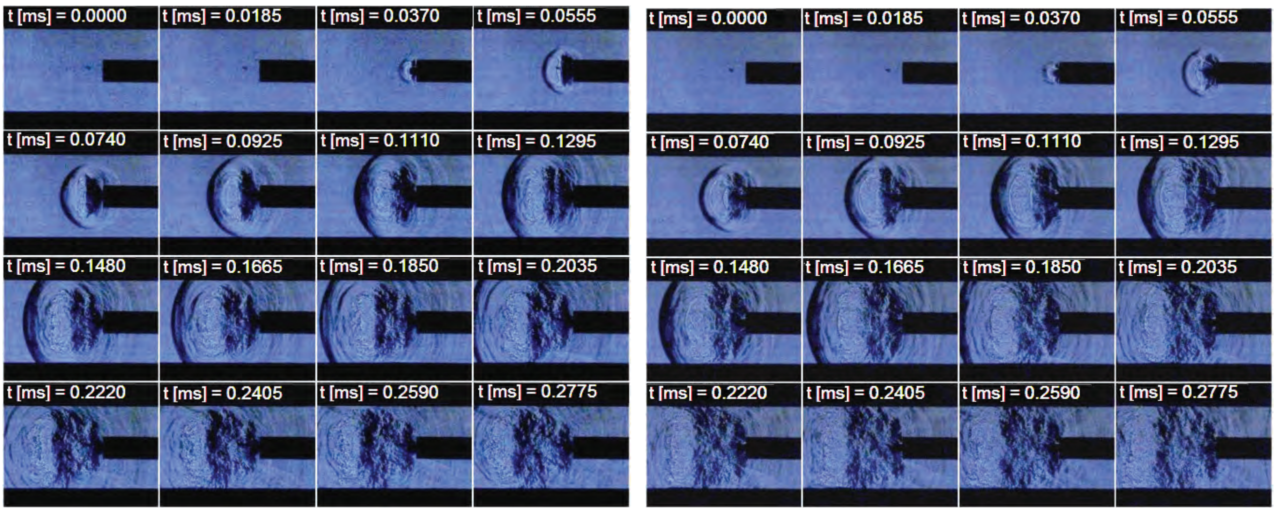


Fig. 9. Schlieren images (recording speed 54 000 fps, 1/297000 s shutter) for cases d = 6 mm, L = 75 mm, with ignition (left, P_{burst} = 7.15 MPa) and without ignition (right, P_{burst} = 4.28 MPa), time t = 0 s specified arbitrarily

4. Conclusions

Self-ignition of hydrogen gas during release through the tube has been investigated over a wide range of tube length. Calculations with detailed chemistry were carried out. Mechanism of this phenomenon postulated by [12] and [3] has been confirmed numerically. Initial pressure needed for ignition is smaller for longer tubes due to more complicated mixing and shock interaction processes. For very short tubes, initial pressure necessary for ignition rises rapidly. This conclusion was confirmed numerically and experimentally (see Fig. 8). The differences in results between calculations and experiments are caused by presence of the real diaphragm, which may intensify the turbulence and affect the ignition process. The next possible reasons are differences in initial conditions in the high-pressure chamber in the moment of membrane rupture. Numerically initial condition of velocity and turbulence intensity is equal to zero; experimentally there is some initial turbulence present.

Further investigations both numerical and experimental are required in order to assess precisely the influence of the tube diameter, length and roughness of the downstream channel walls.

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