EXPERIMENTAL AND NUMERICAL STUDY ON SPONTANEOUS IGNITION OF HYDROGEN-METHANE JETS IN AIR

Rudy, W., Dabkowski, A., and Teodorczyk, A.
Institute of Heat Engineering, Warsaw University of Technology, Nowowiejska Street 21/25, Warsaw, 00-665, Poland, wrudy@itc.pw.edu.pl

ABSTRACT

This paper is aimed at investigation of the spontaneous ignition process of high-pressure hydrogen and hydrogen-methane mixtures injected into the air. The experiments were conducted in a closed channel filled with air where the hydrogen or hydrogen-methane mixture was injected through different geometries (various diameters and extension tube lengths). The methane addition to the mixture was 5% and 10% [v/v]. The results showed that only 5% methane addition may 2.75 times increase the pressure at which the mixture may ignite in comparison to the pressure of the pure hydrogen flow. The 10% of methane addition did not provide an ignition for burst pressures up to 15.0 MPa in the geometrical configuration with the longest tube (100 mm). Additionally, the simulations of the experimental configuration with pure hydrogen were performed with the use of KIVA numerical code with full kinetic reaction mechanism.

1.0 INTRODUCTION

Due to the unique properties (low ignition energy, very high calorific value, wide flammability range) hydrogen is a possible future energy carrier. These unique properties may also be considered as disadvantages, especially from a safety point of view. One of hydrogen features already observed in the 1920's [1] was the self-ignition of high-pressure hydrogen release despite the lack of any known ignition source. Due to the hydrogen's high diffusion coefficient, "diffusion ignition" model has been proposed and investigated experimentally in 1972 [2]. Recently, the spontaneous ignition of hydrogen has been investigated experimentally [2-12] and numerically [4,14-16]. The studies were mainly aimed at understanding the ignition process of depressurized hydrogen released through the extension tube with various length, cross section area and shape. The main conclusion regarding these works was that the ignition probability increases when there is an increase in the tube length [3-10]. One paper of Kim et al. [12] was devoted to flow visualisation in the rectangular (10 x 10 mm) channel utilizing combination of the shadowgraph, high-speed direct photography as well as pressure and lightness sensors. The initial ignitions were always observed in the boundary layer of the mixing zone and the flame spreads to the whole mixing zone. It was also observed that ignition is possible if the static wall pressure exceeded 2.3 MPa. Similar value of 2.5 MPa was provided by Golub et al. [4] in their numerical investigations. The conclusion regarding the observed ignition spot position in the vicinity of the wall [12] is also consistent with the previously performed numerical research [15]. The conclusions regarding tube diameter influence did not determine clearly its impact on the ignition (see Fig. 1.). Nevertheless, as the referenced work showed [4], ignition in tubes with rectangular cross-section area is more probable than in the circular ones. However, none of the latest papers tried to investigate the influence of any gas addition on the ignition occurrence. This paper is aimed at investigation of the methane addition to the hydrogen on the self-ignition occurrence. The chemical and physical properties of hydrogen and methane differ significantly. Methane has narrower than hydrogen flammability and detonability limits, higher minimum ignition energy and lower laminar burning velocity which should have an effect on safety of unintentional high-pressure release of the mixture. The purpose of this research was to provide quantitative data of methane addition to hydrogen and its influence on self-ignition phenomenon.

Previous studies report different hydrogen pressure ranges necessary for the ignition occurrence and the probable reason for that is that different experimental facilities and particular experiment procedures were used. Therefore, the first step of present work was to conduct the experiments and find the ignition range of pure hydrogen flow as a function of initial pressure and tube geometry (diameter and length) as the reference. The next step included experiments with 5% and 10% of methane addition performed with the use of the same experimental stand and experimental procedure.

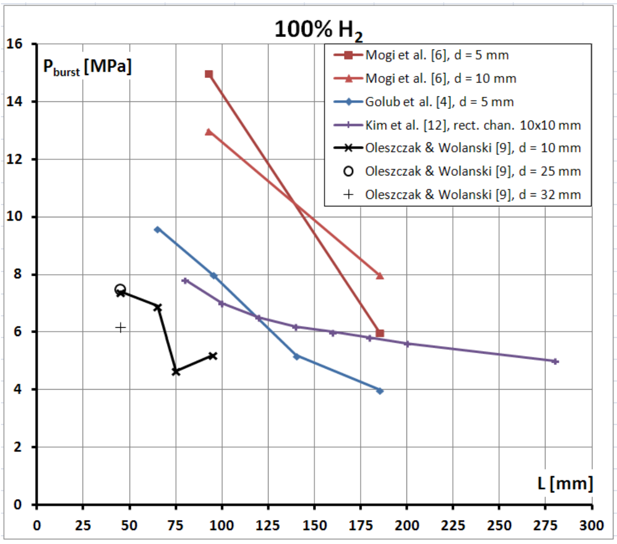

Figure 1. Extension tube length and tube diameter influence on the initial hydrogen pressure necessary for the ignition occurrence [4,6,9,12].

2.0 EXPERIMENTAL FACILITY

The experiments were conducted in Gaseous Detonation Laboratory in Institute of Heat Engineering at Warsaw University of Technology. The experiments were carried out in a closed rectangular channel (0.1 x 0.1 x 1 m) filled with ambient air where the hydrogen and hydrogen-methane mixture (at 1-17.5 MPa and ambient temperature) were depressurized through different geometries: diameter d = 6, 10 and 14 mm, extension tube length: L = 10, 25, 40, 50, 75 and 100 mm. The methane addition to the mixture was 5% and 10% [v/v]. Figure 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 (FASTCAM SA1.1) with schlieren optical system.

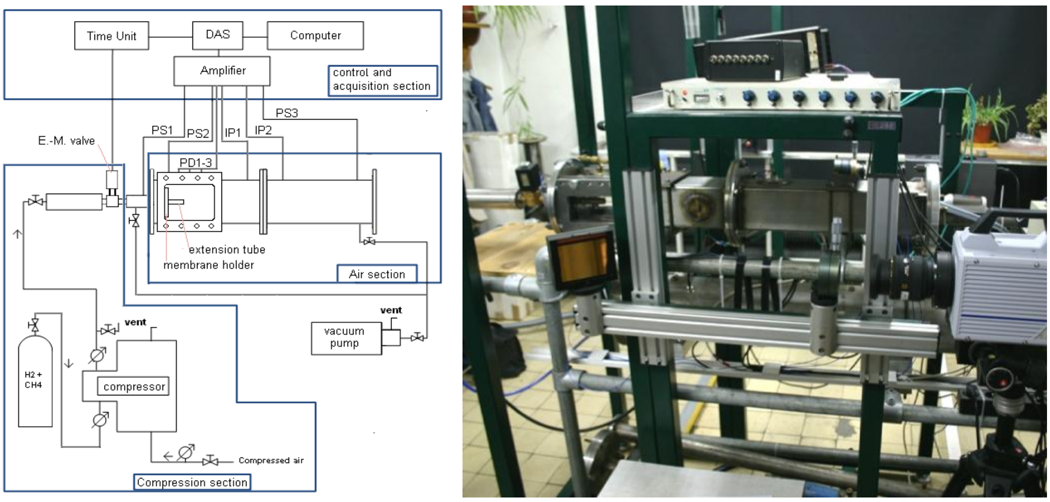

Figure 2. Experimental facility scheme (left) and real view (right). PS - pressure sensors, IP - ionization probes, PD - photodiodes.

The mixture was prepared by means of partial pressures method and compressed with an air-driven gas booster (Haskel AG-75C) to a defined pressure. Activation of the measurement system (with the use of time unit) 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.

3.0 NUMERICAL SIMULATIONS

The numerical simulations were performed with the use of KIVA3V code [17]. The turbulence model used was rng k-epsilon. The solved equations of mass, momentum and energy balance and numerical scheme description are described in details in Los Alamos National Laboratory reports [17-18]. The geometry investigated corresponds to simplified experimental geometry. Left border of the computational domain was defined as pressure inlet boundary condition with pressure equal to P_{burst}. On the right, bottom and top border of the volume filled with air a pressure outlet boundary condition was implemented. Due to the high numerical cost of full chemical reaction mechanism of hydrogen-methane mixtures, only cases with pure hydrogen flow were investigated numerically with use of 23-reaction hydrogen-air mechanism provided by Konnov [19]. The mesh used was 2D, axisymmetrical and included 45 to 105 kcells depending on the tube length and diameter. Additionally, initial simulations showed that ignition, if occurred, takes place in the tube and is independent of the mesh cell size up to 0.25 mm. The extension tube volume was meshed with orthogonal 0.15 mm cell size mesh. The cells in the air-filled volume were no larger than 0.25 x 0.25 mm. The general geometry is presented in figure. 3. It was assumed that the ignition took place if the temperature exceeded 1500 K and OH mole fraction was larger than 0.001. It was also assumed for ignition criteria that these two parameters cannot decrease below the mentioned limiting values up to the end of simulation (50 - 80 μs depending on the tube length).

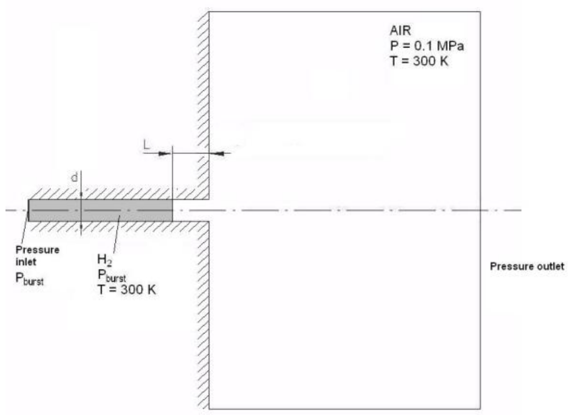

Figure 3. The geometry used in KIVA numerical simulations.

4.0 RESULTS AND CONCLUSIONS

The first step of the experimental work was to find the ignition limit of pure hydrogen flow as a function of initial pressure and tube geometry. The limiting values were then used as a reference for cases with methane addition to the mixture. Totally, more than 250 experiments were conducted for pure hydrogen and hydrogen-methane mixtures. 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. 4. 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. For mixtures with methane addition ionisation probes indicated slightly lower signal due to the fact they were calibrated for hydrogen flame only.

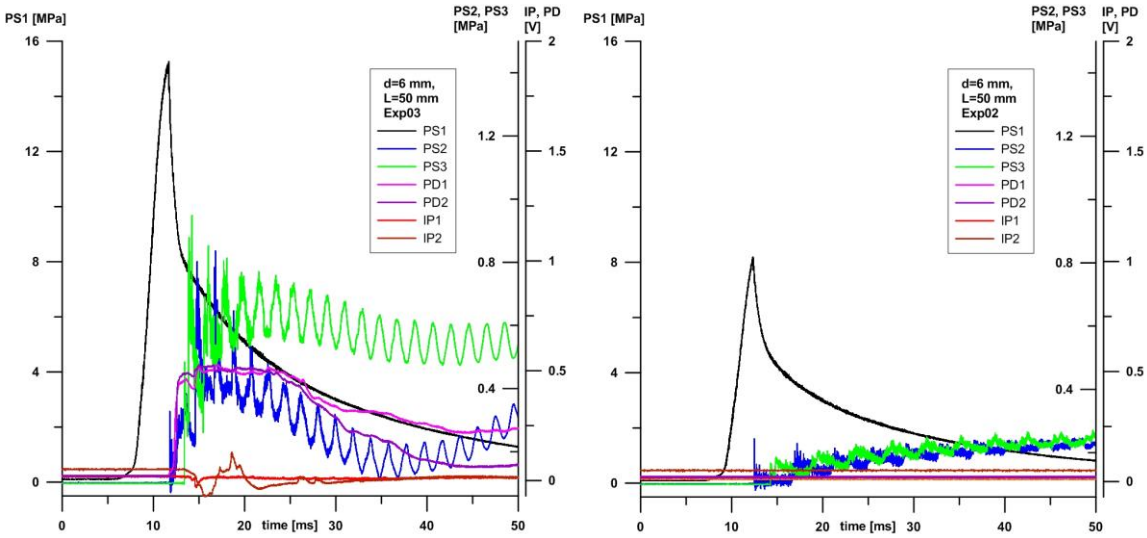

Figure 4. Sensors indications for cases d = 6 mm, L = 50 mm with ignition (left, P_{burst} = 15.3 MPa) and without ignition (right, P_{burst} = 8.4MPa).

The cumulative diagrams obtained for cases with pure hydrogen flow through all the diameters are presented in Fig. 5-7. 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 [3-4,6,8-9]. 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 faster than for longer tubes. It allows to conclude that for this specific experimental setup and geometry used there is some kind of limiting tube length that will not enable the ignition. Similar ignition limiting curves were prepared for 5% and 10% of methane addition to the mixture. The 10% of methane addition to the mixture did not give ignition for any orifice diameter and tube length L = 100 mm for burst pressures up to 15.0 MPa. Therefore, further experiments with shorter tubes and 10% of methane were not conducted. Diagram presenting the ignition limiting curves for 5% of methane in the mixture and its comparison to the pure hydrogen flow ignition limits are presented in Fig. 8. Based on Fig. 8 the tube diameter seems to have undetermined influence on the ignition limit for pure hydrogen flow ignition. For 5% methane addition the diameter influence is more evident. The 5% of methane addition for geometry d = 6 mm and L = 75 mm has the strongest effect on the ignition limit shifting limiting pressure from 5.5 MPa (pure hydrogen case) to 15.1 MPa. These values correspond to pressure ratio 15.1/5.5 = 2.75. The similar pressure ratios for 10 mm and 14 mm diameter and L = 75 mm are equal to 1.5 and 2.4 respectively. Taking into account relatively small methane fraction in the mixture it has a very strong effect. From the safety point of view it is a very important conclusion and needs additional research especially related to the chemical reaction mechanism. Additionally, during the experiments fast camera and optical schlieren system were used. Figure 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.

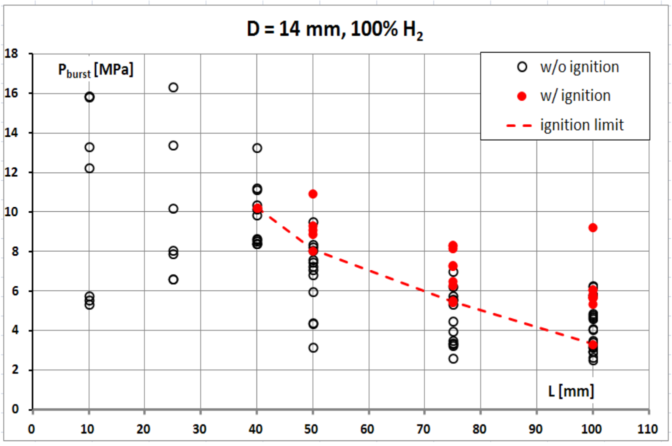

Figure 5. Cumulative diagram for pure hydrogen flow through tube with d = 14 mm and various tube lengths.

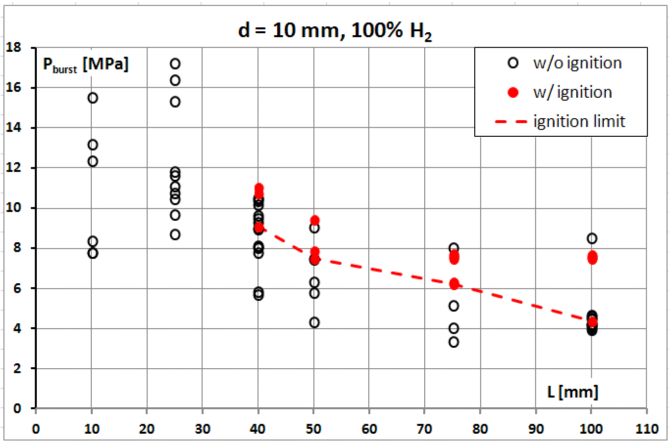

Figure 6. Cumulative diagram for pure hydrogen flow through tube with d = 10 mm and various tube lengths.

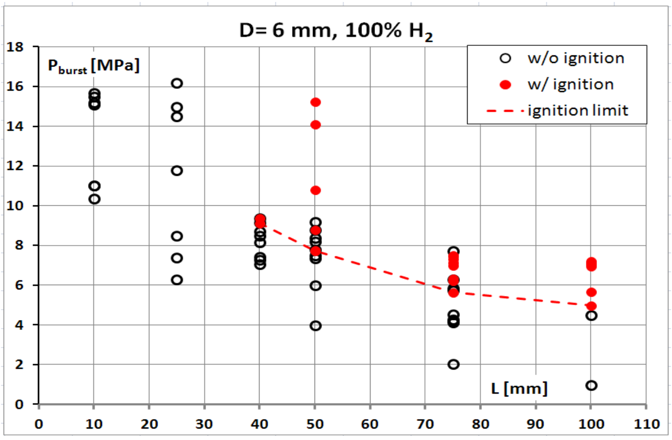

Figure 7. Cumulative diagram for pure hydrogen flow through tube with d = 6 mm and various tube lengths.

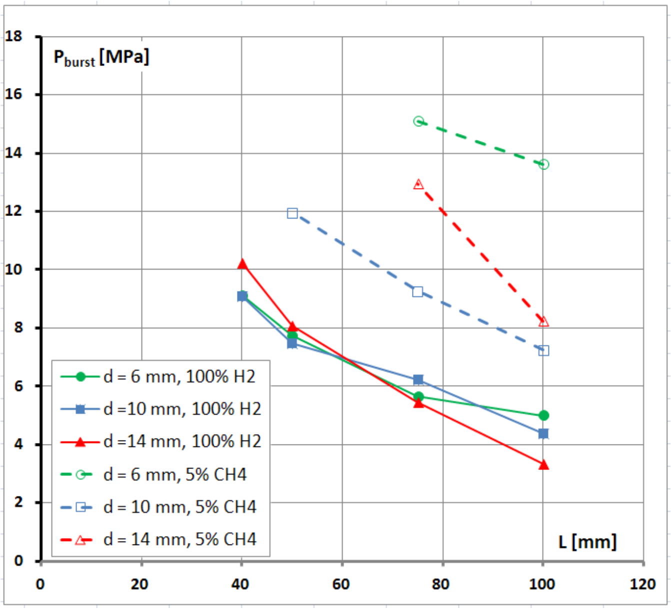

Figure 8. Ignition limits for pure hydrogen and 5% methane addition to hydrogen. Flow through tubes with diameters d = 6, 10 and 14 mm and extension tube lengths L = 40, 50, 75 and 100 mm.

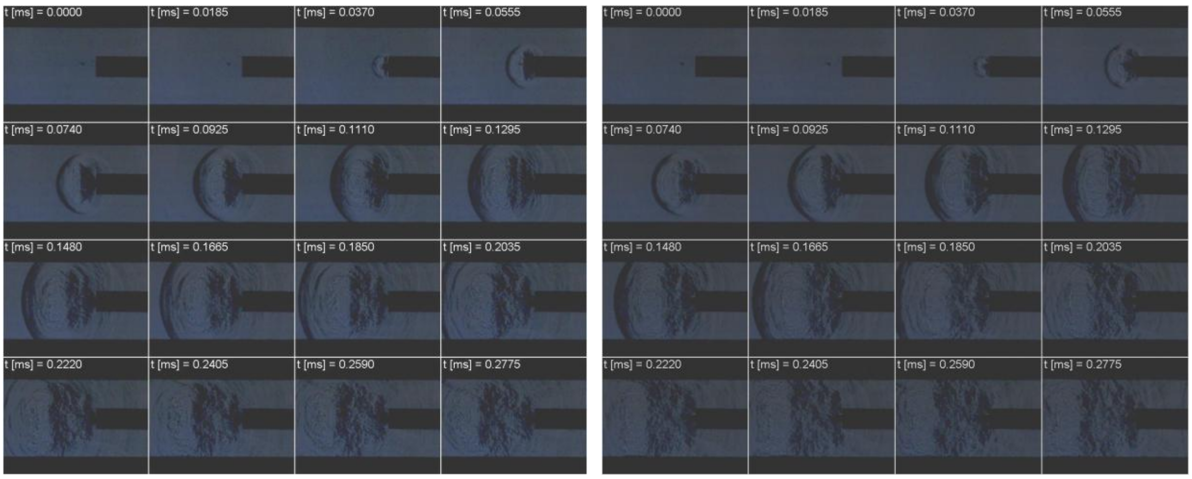

Figure 9. Schlieren images (recording speed 54 000 fps, 1/297000 s shutter) for cases d = 6 mm, L = 75 mm, with ignition (left, P_{burst} = 7.15 MPa) and without ignition (right, P_{burst} = 4.28 MPa). Time t = 0 s specified arbitrarily.

The comparison between the present and previously performed works [4,6,9,12] for pure hydrogen flow ignition is presented in Fig. 10. Generally, all the results are qualitatively similar. The ignition probability increases as the extension tube length increases and as present work shows this relationship seems to be non-linear, especially for shorter tubes. The results obtained by Mogi et al. [6] for different tube diameters are placed in the close vicinity to each other, which is also true for the results described in the present work. It allows to conclude that the results obtained strongly depend on the specific experimental setup and experimental procedure. It is a very important conclusion because of the high quantitative discrepancy between the results presented in Fig. 10 where the differences in limiting pressures for similar geometrical parameters may differ by 400 %.

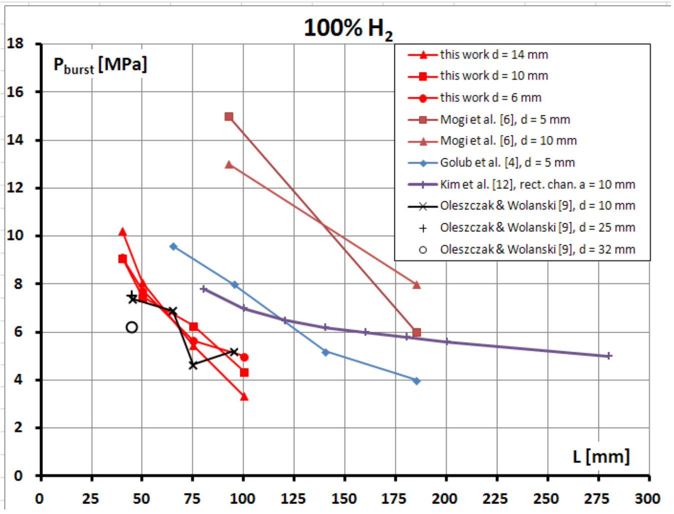

Figure 10. Ignition limits of 100% hydrogen flow. Comparison between present and referenced works [4,6,9,12].

Numerical simulations performed within the 2-20 MPa pressure range and extension tube length 10-100 mm. The ignition, if occurred, started from the area in the close vicinity of the wall in the channel where the complex shock-wall and shock-shock interactions are present increasing the temperature and hydrogen-air mixing process. This process may be observed in the Fig. 11, which presents the example contours of temperature and OH mole fraction during the hydrogen discharge process. Ignition starts at the wall (Fig. 11, t = 20 μs) and the reaction propagates across the hydrogen-air mixing zone. When the gas flows out of the tube, the reaction zone is being stretched due to the gases decompression and two main reactions zones are produced. First zone, in the vicinity of the orifice edge and the vortex produced by the edge and the second one, in the channel axis (Fig.11, t = 60 μs). If the conditions are favourable (sufficient initial pressure and reacting area) these two reaction zones merge. For the cases with shorter tubes (tube length approximately lower than 25 mm) the ignition took place at first in the channel axis, near its end or just after the hydrogen leaves the channel. Then, the reaction zone is being stretched and propagates along the mixing zone. In this case the ignition is a result of leading shock influence, which increases the temperature of the downstream air. For this type of ignition higher initial pressures are necessary. This type of ignition is similar to "diffusive ignition" firstly described by Wolanski and Wojcicki in 1972 [2]. In spite of the fact that the real flow is 3-dimensional and the simulations are 2-dimensional the results obtained with KIVA seem to be qualitatively correct. The other parameter which may have influence on the numerical results is the diaphragm. In our simulations the diaphragm was a flat virtual border between two volumes with different initial conditions. In real experiments the bulging of a diaphragm appeared before bursting. The diaphragm bulging effect was numerically investigated in paper of Golub et al. [4] indicating meaningful effect on combustion intensity.

The cumulative diagram of the results obtained from the experimental and numerical part of this paper is presented in Fig.12. As it has already been observed in the experiments and referenced works, if the tube length increases the ignition possibility also increases. In numerical results this possibility is non-linear for shorter tubes (L < 25 mm) and decreases rapidly as the length decreases. For 10 mm tube length only for d = 14 mm case the ignition was observed with burst pressure of 17 MPa. Cases with L = 10 mm, d = 6 and 10 mm did not produce conditions for ignition to occur for pressures up to 20 MPa. The channel diameter influence may also be noticeable. For longer tubes (L > 25 mm) the possibility of ignition increases as the diameter decreases. For example, the initial pressure necessary to ignite the flow in 100 mm tube is twice lower for d = 6 mm than for d = 14 mm case.

In spite of the research done the successive investigation should be done both experimental and numerical. The field of that research should include: wider range of the extension tube length, wider range of the burst pressures and numerical simulations of hydrogen-methane mixtures flow with full kinetics reaction mechanism and 3D geometry of the experimental facility.

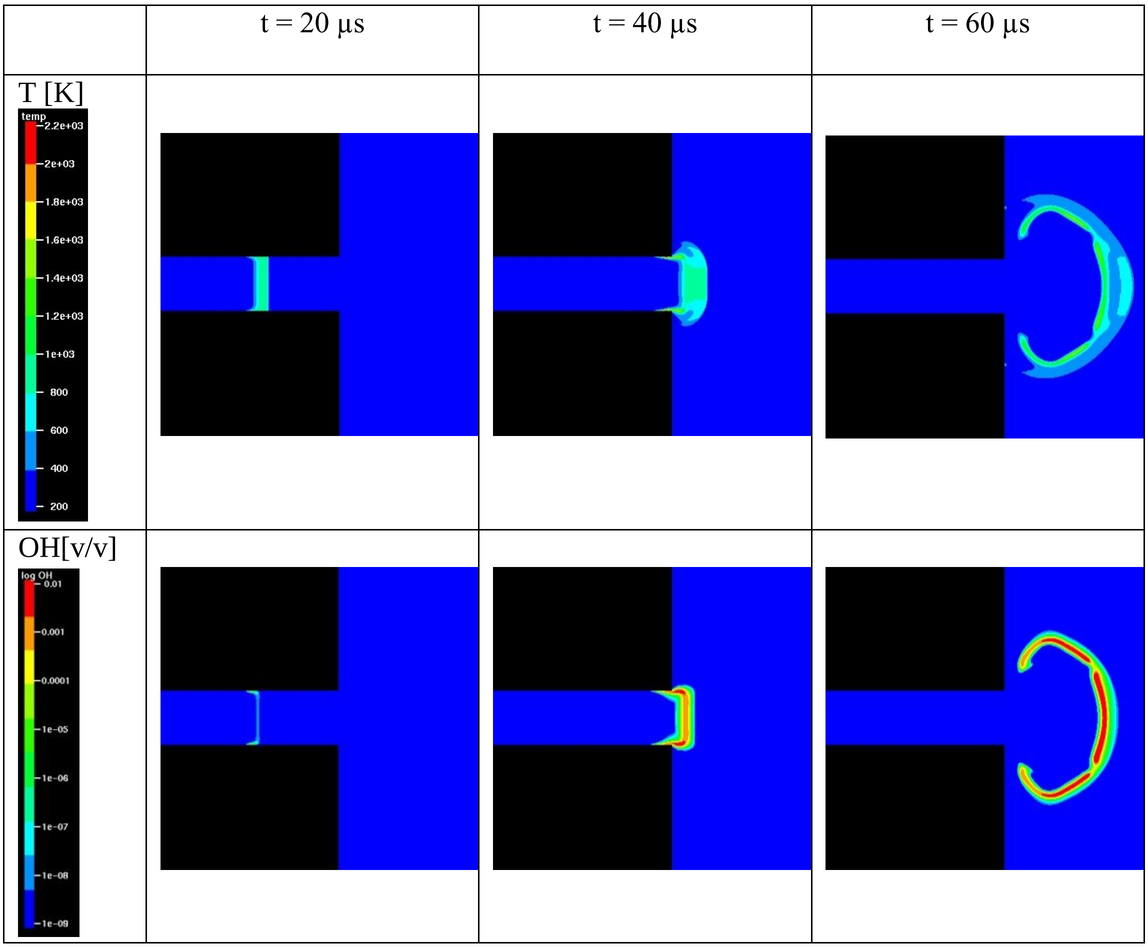

Figure 11. Temperature and OH fraction distribution during hydrogen discharge. Case: d = 14 mm, L = 50 mm, P_{burst} = 8 MPa.

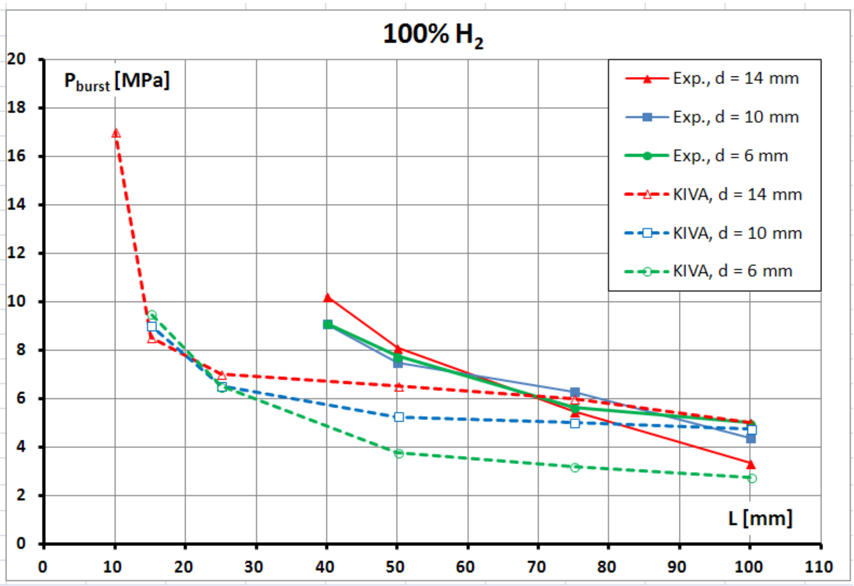

Figure 12. Comparison between ignition limits obtained experimentally (solid lines) and numerically (dashed lines) for different diameters and extension tube lengths.

5.0 ACKNOWLEDGEMENTS

This paper was supported by the Perennial Program – II stage: *"Work Conditions and Safety Improvement"* financed during 2011-2013 within the frame of scientific and development research by the *Ministry of Science and Higher Education/The National Centre for Research and Development.* Program coordinator: *Central Institute for Labour Protection – National Research Institute.*

6.0 REFERENCES

[1] Anon, Spontaneous Ignition of Hydrogen, *Engineering,* **113**, 1922.

[2] Wolanski, P., Wojcicki, S., Investigation into the mechanism of the diffusion ignitron of a combustible gas flowing into an oxidizing atmosphere, *Proceedings of the Combustion Institute,* **14**, 1972, pp. 1217-1223.

[3] Golub, V.V., Baklanov, D.I., Bazhenova, T.V., Bragin, M.V., Golovastov, S.V., Ivanov, M.F., Volodin, V.V., Shock-induced ignitron of hydrogen gas Turing accidental or technical opening of high-pressure tanks, *Journal of Loss Prevention in the Process Industries,***20**, 2007, pp. 439-446.

[4] Golub, V.V., Baklanov, D.I., Golovastov, S.V., Ivanov, M.F., Laskin, I.N., Savaliev, A.S., Semin, N.V., Volodin, V.V., Mechanisms of high-pressure hydrogen gas self-ignition in tubes, *Journal of Loss Prevention in the Process Industries,* **21**, 2008, pp. 185-198.

[5] Dryer, F.L., Chaos, M., Zhao, Z., Stein, J.N., Alpert, J.Y., Homer, C.J., Spontaneous ignition of pressurized releases of hydrogen and natural gas into air, *Combustion Science and Technology,* **179**, No. 4, 2007, pp. 663-694.

[6] Mogi, T., Kim, D., Shiina, H., Horiguchi, S., Self-ignition and explosion during discharge of high-pressure hydrogen, *Journal of Loss Prevention in the Process Industries,* **21**, 2008, pp. 199-204.

[7] Bazhenova, T. V., Golub, V. V., Laskin, I. N., Semin, N. V., Prevention of hydrogen self-ignition at technical opening via replacement of one orifice by several smaller ones, 22nd International Combustion Dynamics of Explosions and Reactive System, Minsk, Belarus, July 27-31, 2009.

[8] Mogi, T., Wada, Y., Ogata, Y., Hayashi, A.K., Self-ignition and flame propagation of high-pressure hydrogen jet during sudden discharge from a pipe, *International Journal of Hydrogen Energy,* **34**, 2009, pp. 5810-5816

[9] Oleszczak, P., Wolanski, P., Ignition during hydrogen release from high pressure into the atmosphere, *Shock Waves,* **20**, 2010, pp. 539-550.

[10] Lee, H.J., Kim, Y.R., Kim, S.H., Jeung, I.S., Experimental investigation on the self-ignition of pressurized hydrogen released by the failure of a rupture disk through tubes, *Proceedings of the Combustion Institute,* **33**, 2011, pp. 2351-2358.

[11] Grune, J., Kuznetsov, M., Lelyyakin, A., Jordan, T., Spontaneous ignition process due to high-pressure hydrogen release in air, Proceedings to the *4th International Conference of Hydrogen Safety*, San Francisco, 2011, pp.132-7A3, USA.

[12] Kim, Y. R., Lee, H.J., Kim, S.H., Jeung, I.S., A flow visualization study on self-ignition of high pressure hydrogen gas released into a tube, *Proceedings of the Combustion Institute,* **34**, 2013, pp. 2057-2064

[13] Rudy, W., Dabkowski, A., Teodorczyk, A., Investigation into the Ignition Process of Hydrogen Jet in Air, *Archivum Combustionis,* **30**, No.1-2, 2010.

[14] Wen, J.X., Xu, B.P., Tam, V.H.Y., Numerical study on spontaneous ignition of pressurized hydrogen release through a length of tube, *Combustion and Flame,* **156**, No.11, 2009, pp. 2173-2189

[15] Bragin, M.V., Molkov, V.V., Physics of spontaneous ignition of high-pressure hydrogen release and transition to jet fire, *International Journal of Hydrogen Energy,* **36**, 2011, pp. 2589-2596.

[16] Yamada, E., Kitabayashi, N., Koichi Hayashi, A., Tsuboi, N., Mechanism of high-pressure hydrogen auto-ignition when spouting into air, *International Journal of Hydrogen Energy,* **36**, 2011, pp. 2560-2566.

[17] Amsden, A. A.: *KIVA-3: A KIVA Program with Block-Structured Mesh for Complex Geometries*, Los Alamos National Laboratory Report LA-12503-MS, New Mexico 87545, March 1993.

[18] Amsden, A. A., O'Rourke P.J., Butler T.D.: *KIVA-II: A Computer Program for Chemically Reactive Flows with Sprays*, Los Alamos National Laboratory Report LA-11560-MS, New Mexico 87545, May 1989

[19] Konnov, A.A., *Detailed reaction mechanism for small hydrocarbons combustion Release 0.5,* 2000.