Experimental and Numerical Study of Spontaneous Ignition of Hydrogen-Methane Jets in Air

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Presentation plan

1. Introduction
2. Experimental facility
3. Numerical simulations
4. Results
5. Conclusions
6. Future research
7. Acknowledgements
8. References

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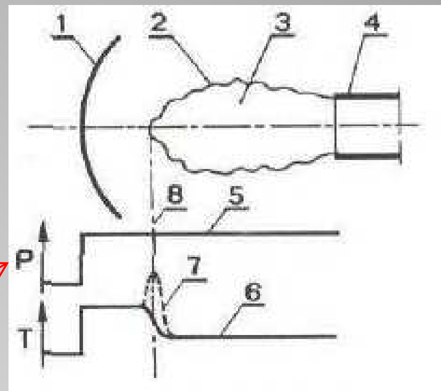
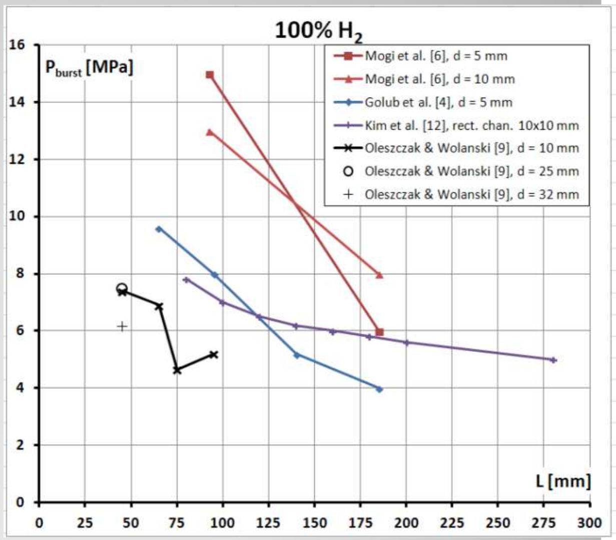
1. Introduction

- Unique properties (wide flamm. range, LHV, MIE etc.) indicate hydrogen as a future energy carrier
- Unintended high pressure release with possible self-ignition firstly obseved in 1920's [Anon]
- „Diffusion ignition" model proposed in 1972 [Wolański & Wójcicki] – synthesis gas ($3H_2+N_2$)@ 300^0C and 20 MPa



- **Recent experimental works** - geometrical configuration influence on the self-ignition occurence:
  - tube diameter [Mogi et al. 2008,2009; Golub et al. 2007, 2008; Oleszczak 2009…]
  - cross-section shape [Golub et al.…]
  - tube length [Oleszczak, Mogi et al, Golub et al…].
- General tendency is observed but high quantitative discrepancy between researchers – different experimental stands and procedures



- **Recent numerical works:**
  - longer tubes: ignition in the boundary layer and in the axis [Lee&Jeung 2009]
  - shorter tubes: self-ignition in axis - thin diffusion layer [Xu et al. 2009]

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1. Introduction cont.

- In general we may claim that main geometrical factors responsible for hydrogen self-ignition are well recognised but phenomenon is highly **boundary and initial conditions dependent.**

How to make hydrogen safer without significant changes in the other properties?
There is no research aimed at doping influence on the self-ignition occurence

The aim of this research is to find the influence of the methane addition to hydrogen on the self-ignition.

Property	H_2	CH_4
Molecular weight [kg/kmol]	2.016	16.043
Diffusive coefficient in air @ NTP [cm^2/s]	0.61	0.16
Viscosity @ NTP [g/cm-s x 10^{-5}]	89	11,7
Density @ NTP [kg/m^3]	0.0838	0.6512
LFL - UFL	4-75	5.3-15
MIE [mJ]	0.02	0.29
Maximum burning velocity [m/s]	42.5	10.2

Source: Alcock et al. 2001

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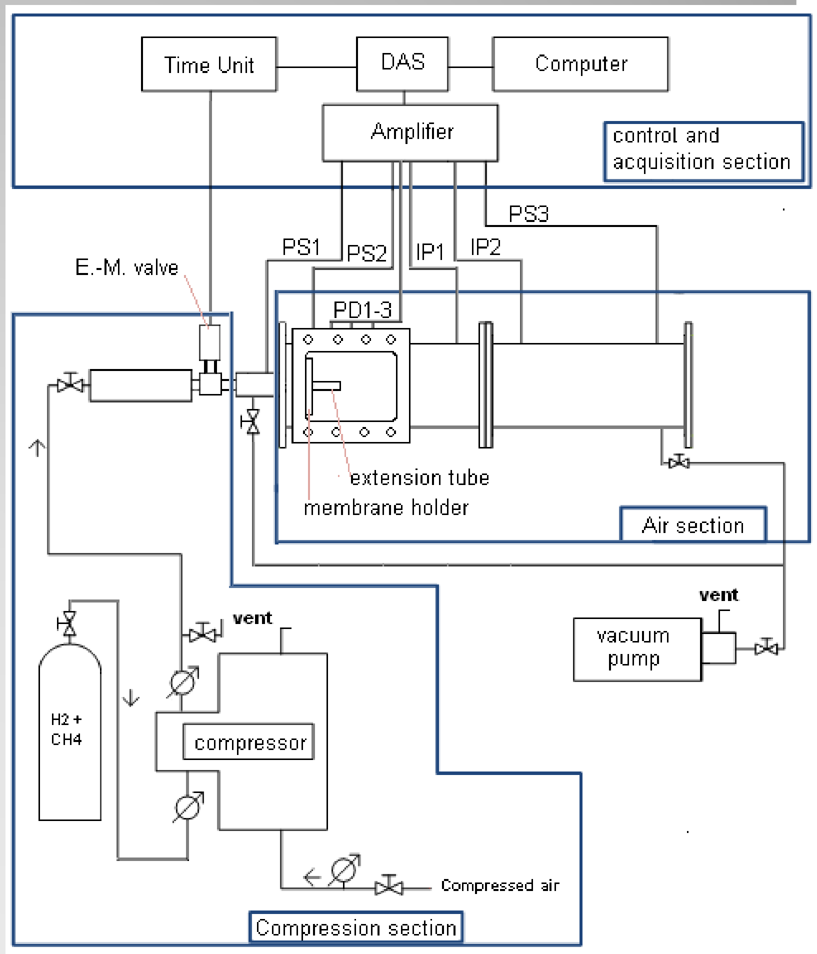
2. Experimental facility

- 3 sections:
- Compression section (cylinder, compressor, feeding line, E-M valve, pressure accumulator)
- Control and acquisition section (computer, time unit, amplifier, data acquisition system)
- Air section (1x0.11x0.11 m rectangular tube, membrane, membrane holder)
- Equipment: pressure sensors (PCB), photodiodes, ion probes, camera Photron SA.1.1 & IS-1M
- Mixtures:
  Hydrogen+Methane (0%, 5% and 10 %)
- Extension tubes:
  d = 6, 10, 14 mm
  L = 10, 25, 40, 50, 75 and 100 mm



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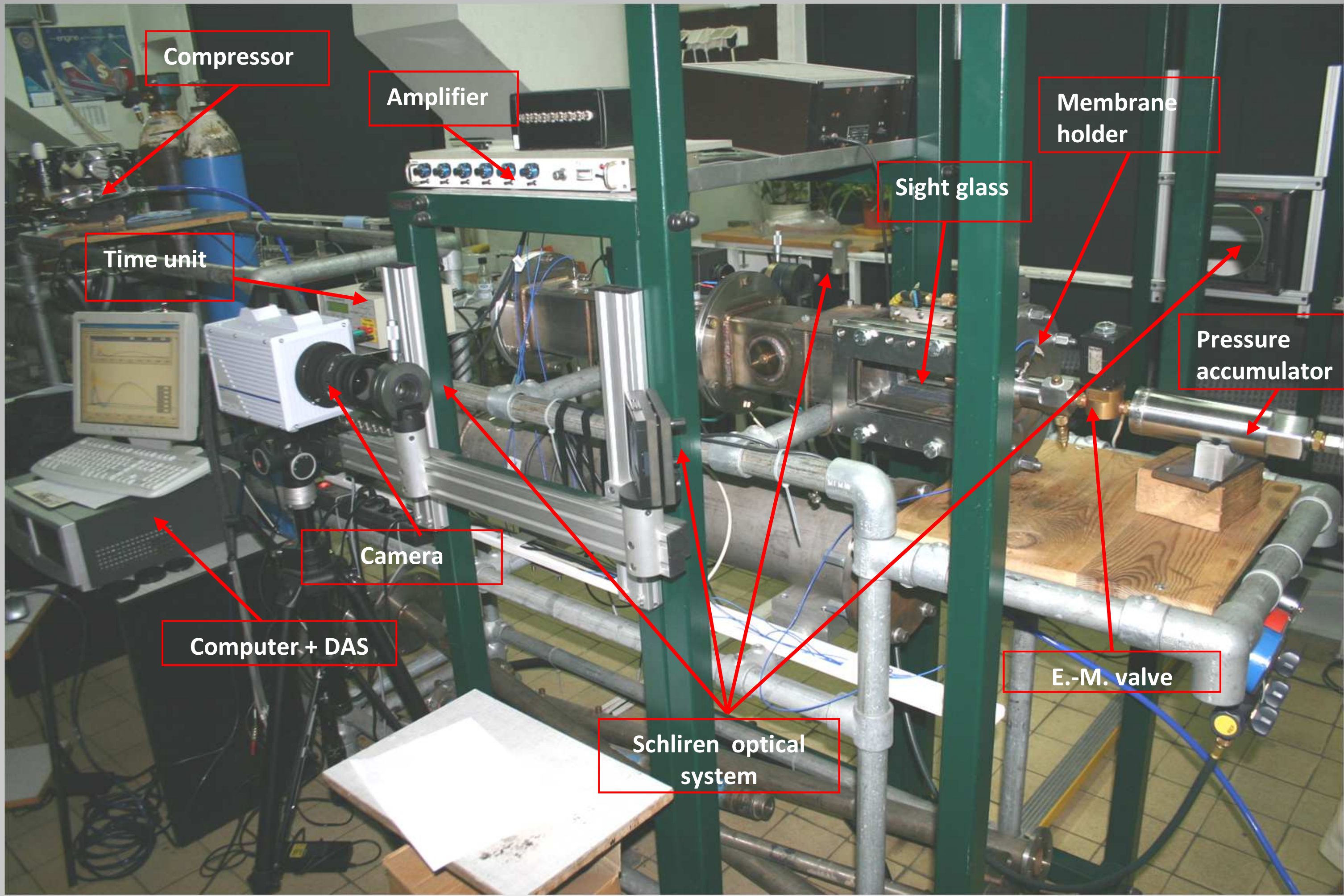
2. Experimental facility cont.



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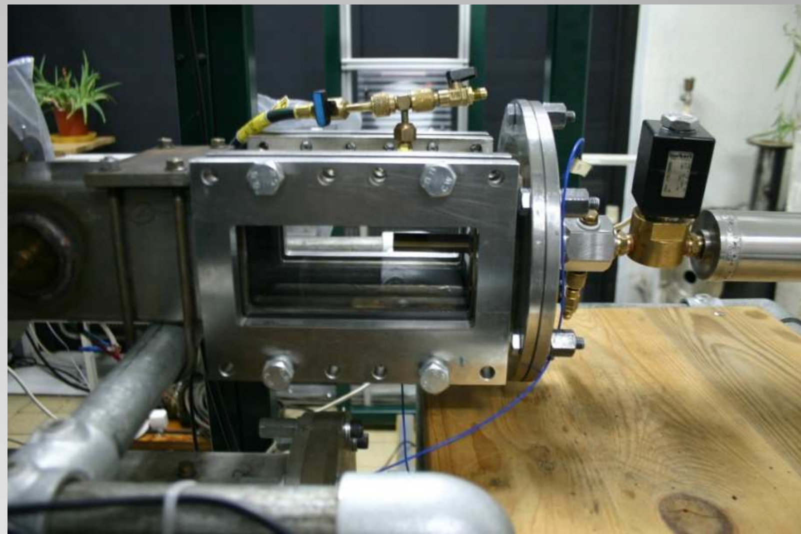
2. Experimental facility cont.



d = 6 mm d = 10 mm d = 14 mm

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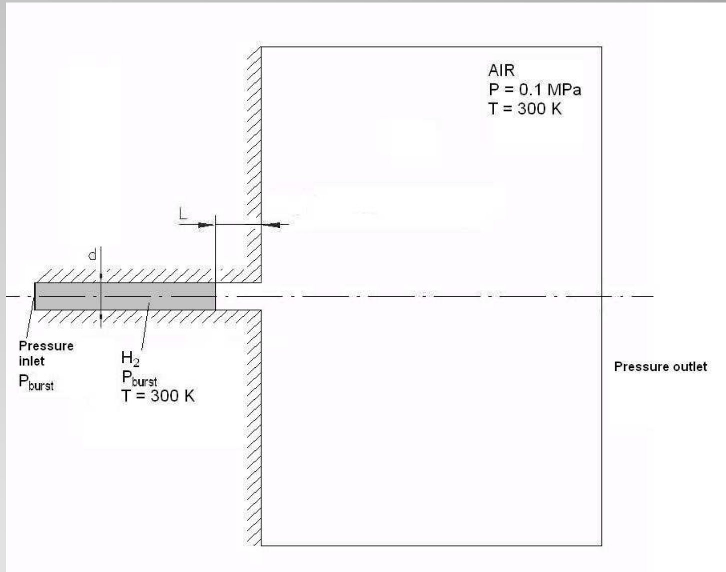
3. Numerical simulations

- 2D axisymmetrical geometry – representing simplified experimental geometry
- KIVA-3V code
- Hydrogen-air reaction mechanism (23 reactions) [Konnov]
- 45 to 105 kcells depending on geometry
- Structural mesh in the tube 0.15x0.15 mm
- Maximum cell dimension 0.25x0.25 mm
- 2 volumes:
  - High pressure hydrogen section
    P = 2-18 MPa, T= 300 K
  - Low pressure air section
    P = 0.1 MPa, T=300 K
- Extension tubes:
  d = 6, 10, 14 mm, L = 10, 15, 25, 50, 75, 100 mm



Ignition criterion:
Temperature >1500 K
and
OH mass fraction > 0.001

4. Results

Totally more than 400 experiments were conducted,
Parameters changed:

- Methane concentration: 0%, 5%, 10% v/v,
- Tube diameter: 6, 10 and 14 mm,
- Tube length: 10, 25, 40, 50, 75 and 100 mm,
- Burst pressures: 2-18 MPa

Totally more than 75 numerical simulations were performed:

- Pure hydrogen flow,
- Tube diameter: 6, 10 and 14 mm,
- Tube length: 10, 15, 25, 50, 75 and 100 mm
- Burst pressures: 2-20 MPa

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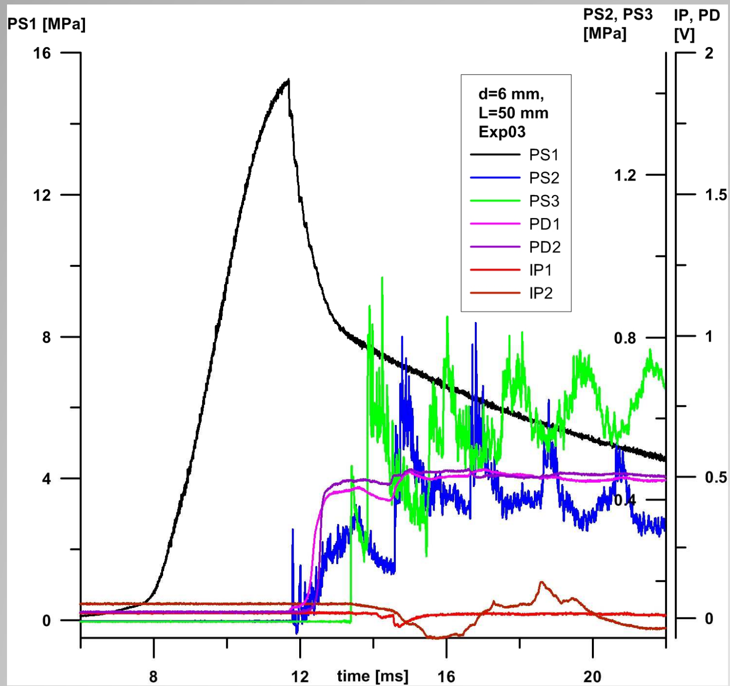
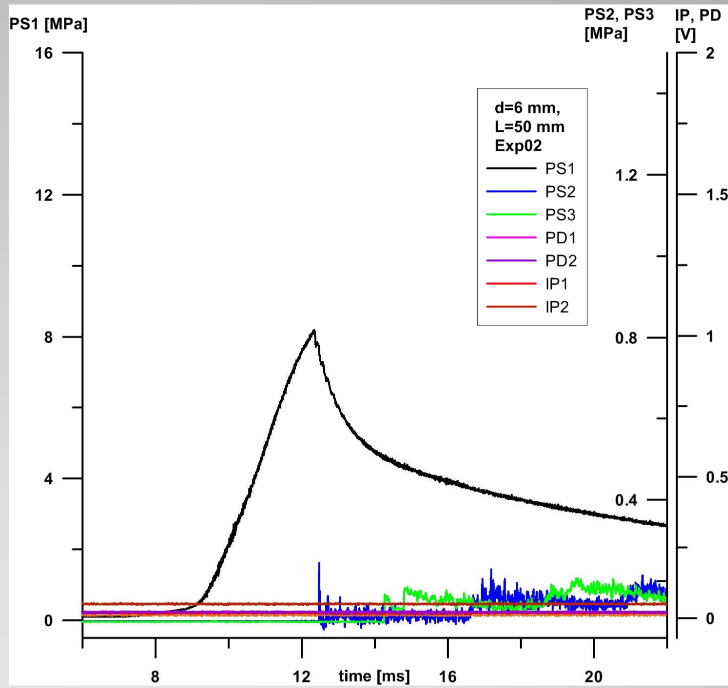
4. Results cont.





Example sensor indications for cases w/ (left) and w/o ignition (right)

P_{burst} = 15.3 MPa P_{burst} = 8.4 MPa

- Clear PD signal → ignition in a tube
- Clear IP signal → flame sustained
- PS2 and PS3 → clear increase in pressure caused by the combustion, oscillations - shock reflections

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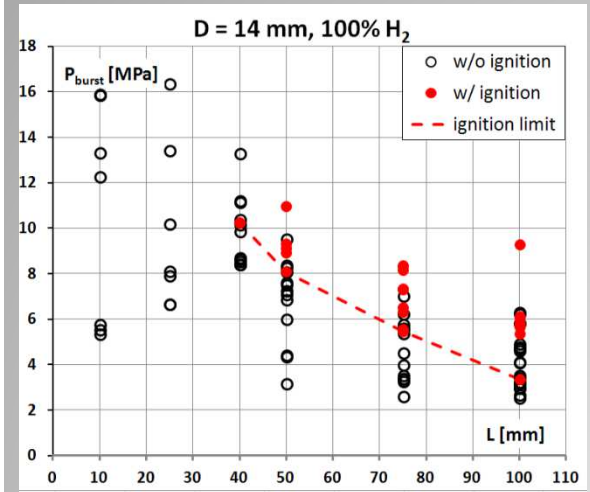
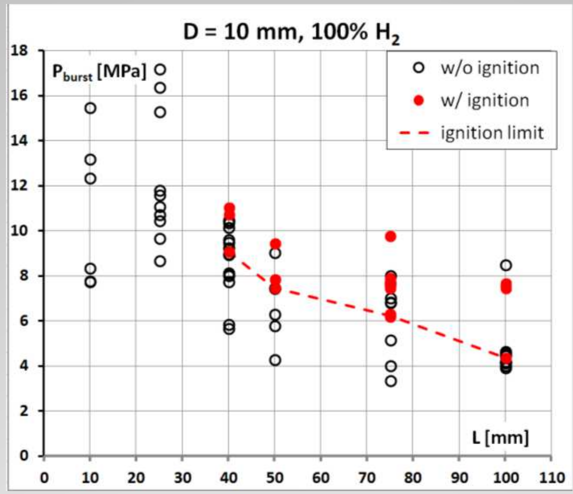
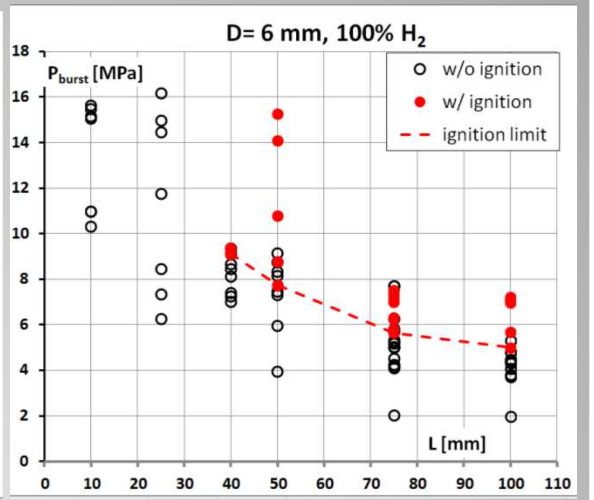
4. Results cont.

100% H_2







- Self-ignition has „stochastic" feature
- For a specific geometrical configuration below certain pressure self-ignition is not possible
- Self-ignition probability increases with tube length increase
- Self-ignition limit curve becomes flatter when tube diameter decreases
- No ignition for tubes with L < 40 mm → nonlinear self-ignition limit curve

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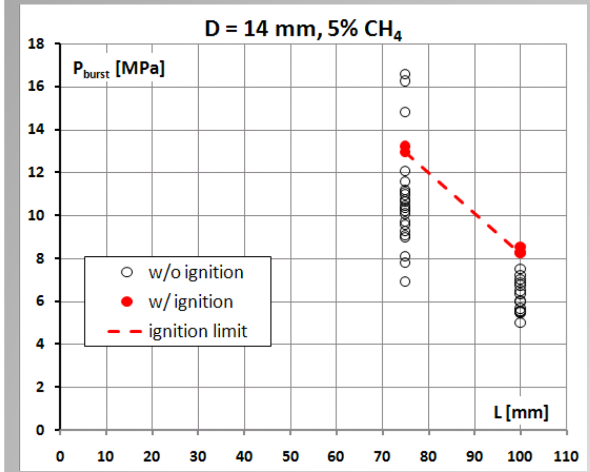
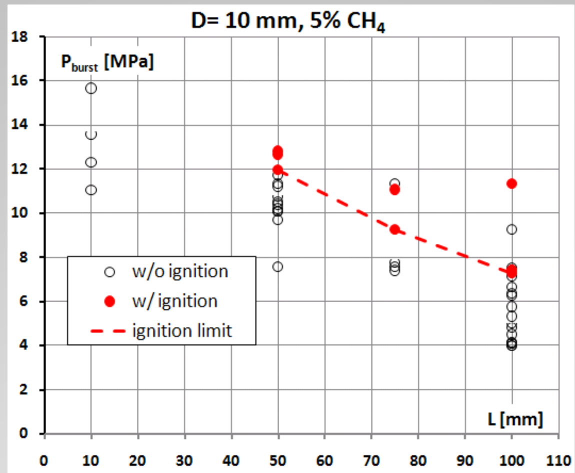
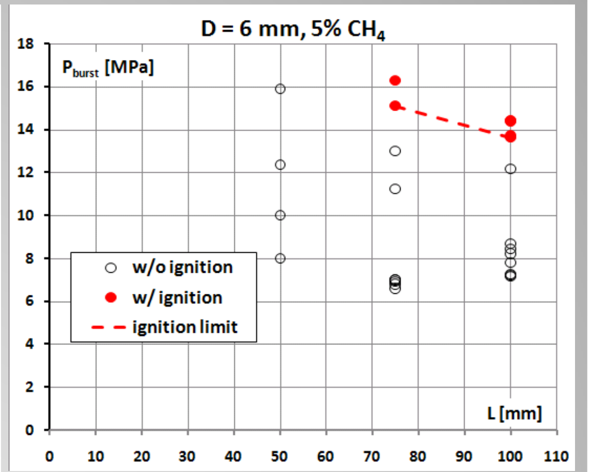
4. Results cont.

5% CH_4







- 5% methane addition increase significantly self-ignition initial pressure

pressure ratios
$P_{burst\ 5\%\ CH4} / P_{burst\ H2}$ →

	d=6 mm	d=10 mm	d=14 mm
L=50 mm	-	1,6	-
L=75 mm	1,94	1,49	2,37
L=100 mm	2,75	1,67	2,47

- For 10% of methane addition and L = 100 mm - no ignition for pressures up to the 16 MPa

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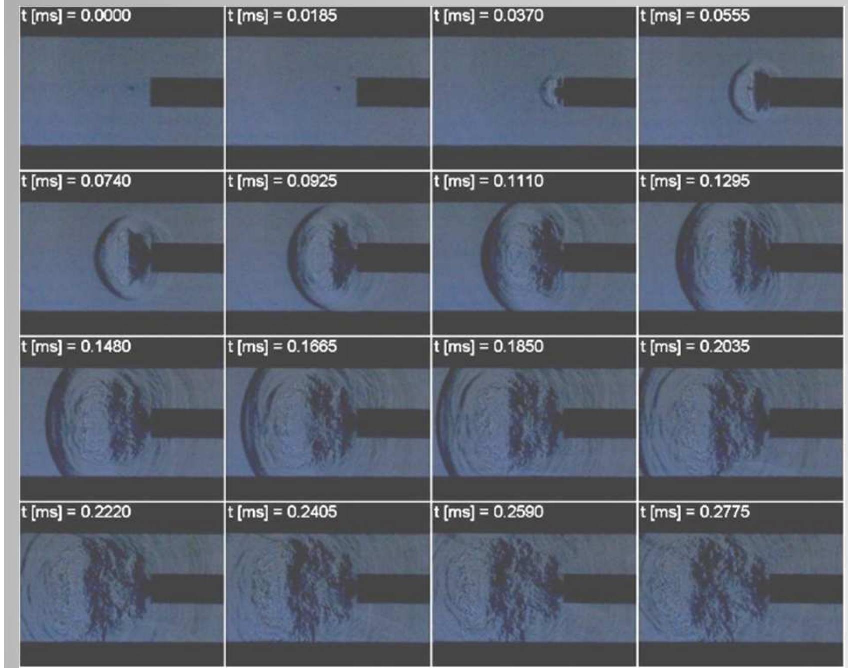
4. Results cont.

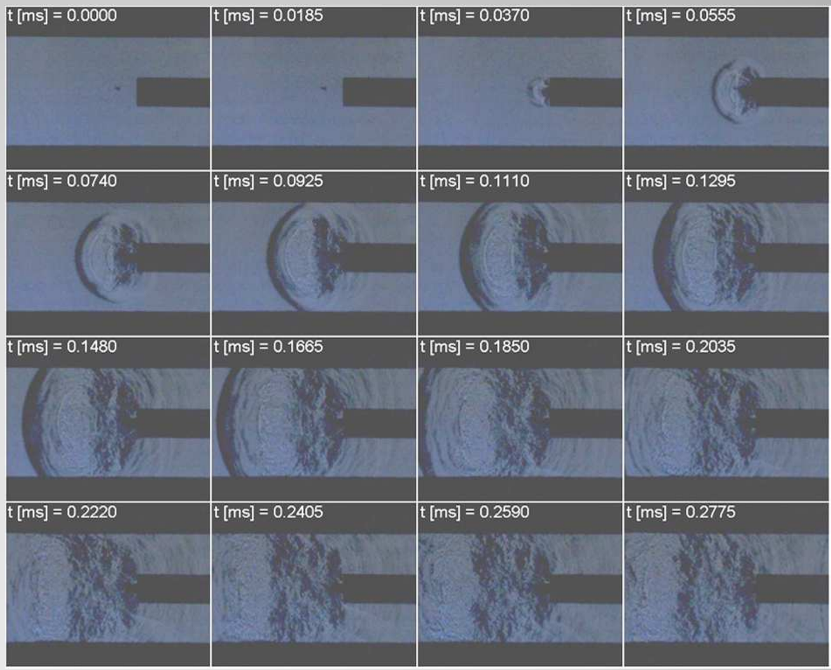
Schlieren images,

camera Photron FASTCAM SA1.1, 54 kfps, shutter 1/297000 s

- Slight differences in images w/ and w/o ignition
- PDs are the main indicators of flame



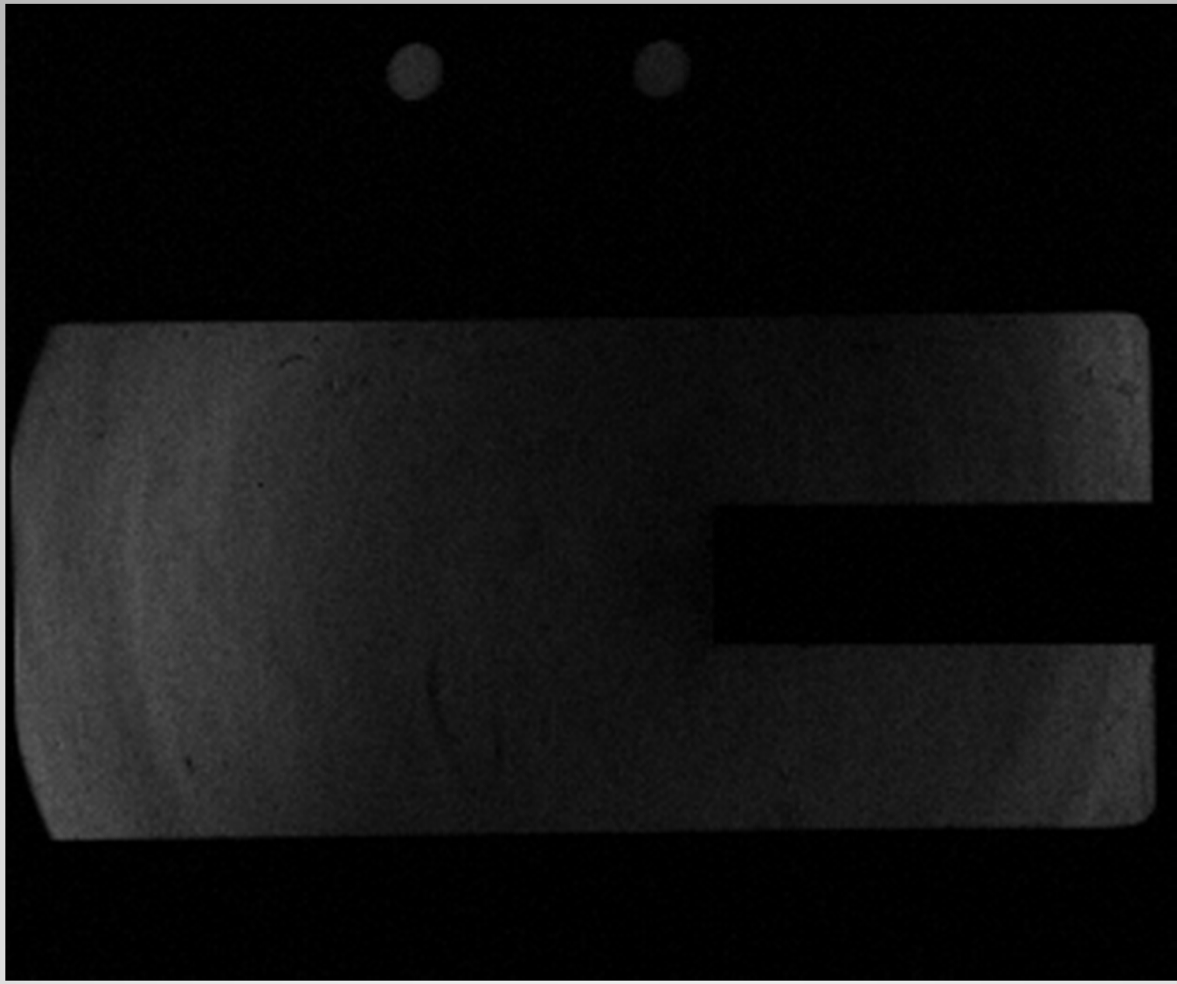
d=6 mm, L=75 mm, P_{burst} = 7,15 MPa
w/ ignition



d=6 mm, L=75 mm, P_{burst} = 4,28 MPa
w/o ignition

4. Results cont.

Schlieren images, 125 kfps, camera Photron FASTCAM IS-1M



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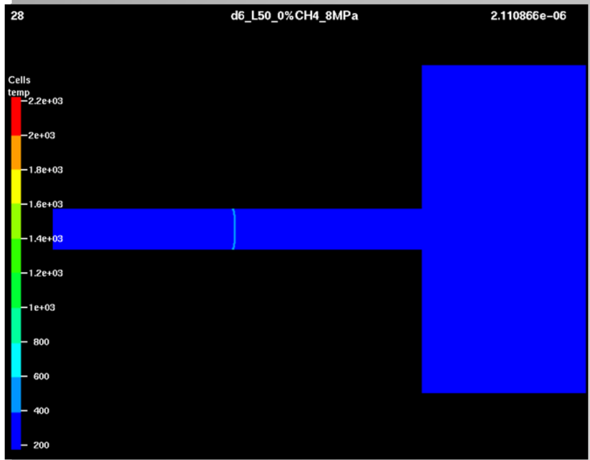
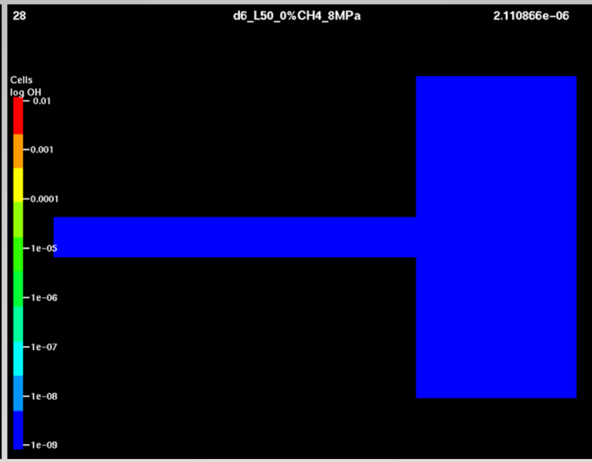
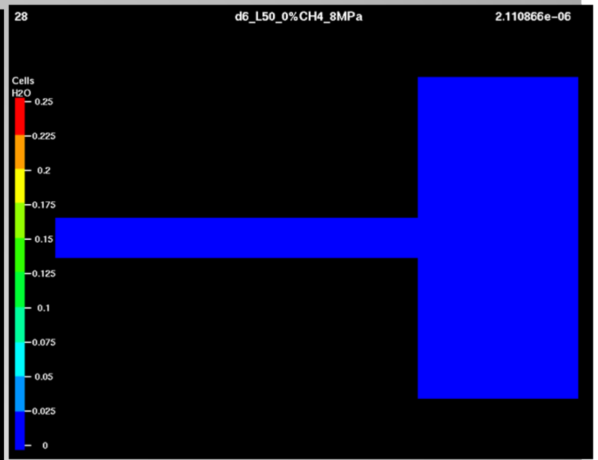
4. Results cont.

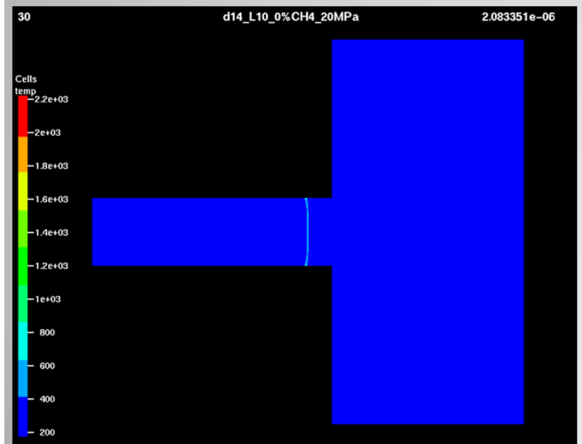
Temperature [K] | OH mass fraction [-] | H_2O mass fraction [-]

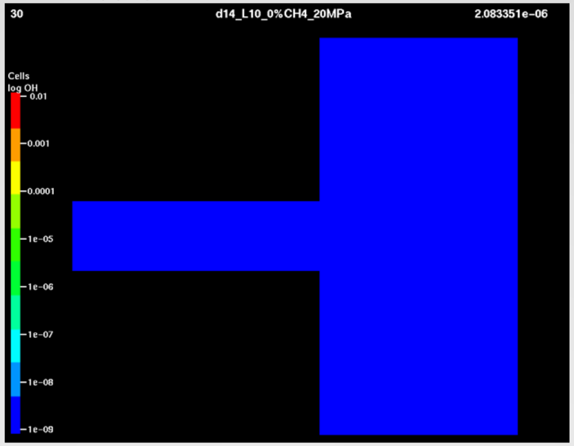


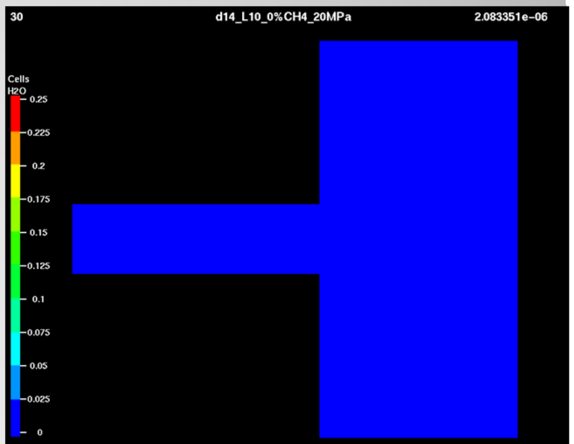




d = 6 mm, L = 50 mm, P_{burst} = 8 MPa, ignition in a tube near wall







d = 14 mm, L = 10 mm, P_{burst} = 20 MPa, „diffusive ignition”

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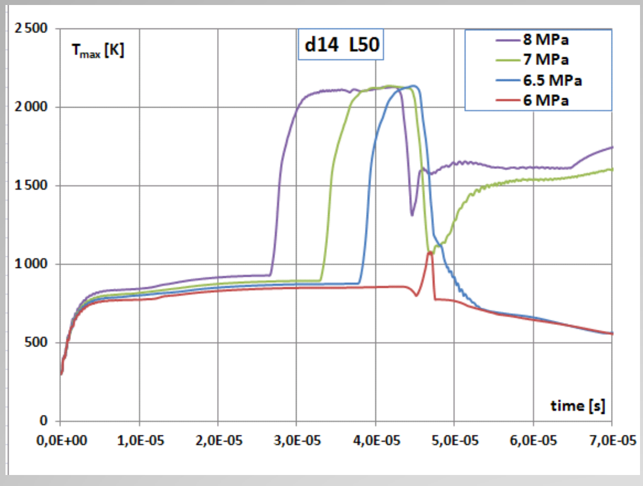
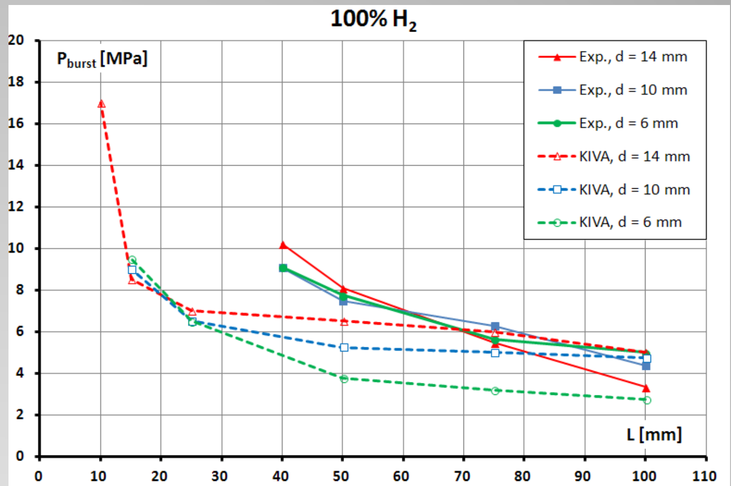
4. Results cont.





Numerical simulations:

- Ignition in longer tubes: near the wall of the channel where complex shock-wall and shock-shock interactions are present increasing the temperature and mixing process,
- ignition in shorter tubes (L = 10 mm) ignition took place in a similar way as described by Wolański&Wójcicki „diffusive ignition" occurred just after leading shock wave, the burst pressure necessary for this kind of ignition is considerably higher than for cases with longer tubes,
- diameter influence on the self-ignition occurence visible for longer tubes (L > 25 mm)

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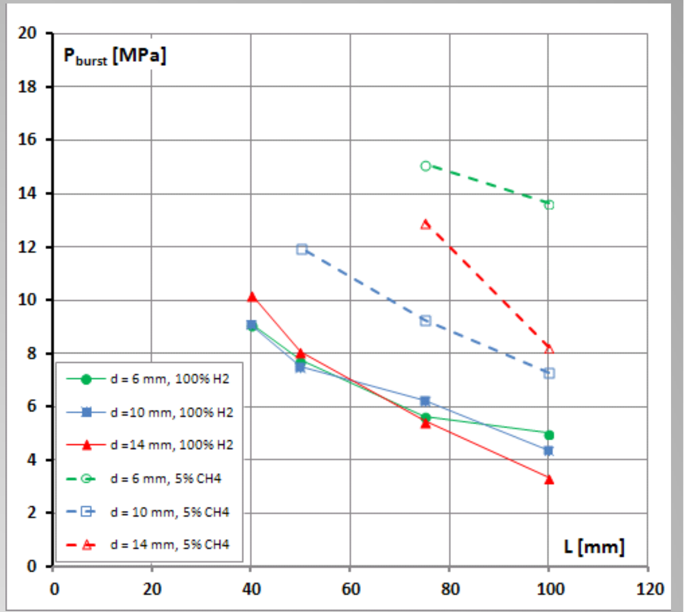
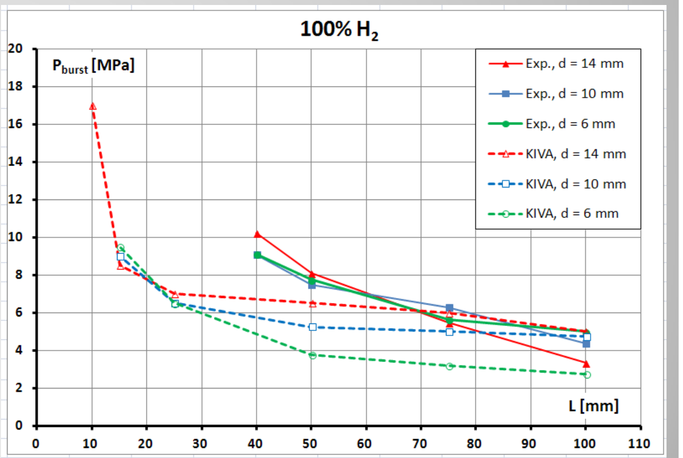
5. Conclusions

- Hydrogen may ignite when released into the air only above certain initial pressure – stochastic feature
- Adding 5% of methane to hydrogen increases significantly (1.5 – 2.7 times) the pressure necessary for self-ignition the mixture
- self-ignition did not occur for 10% of methane addition for pressures up to 15 MPa and L = 100 mm
- non-linear pressure to L dependence
- no ignition occured for tubes with L < 40 mm



Numerical simulations:
- 2 types of ignition:
  - longer tubes (L > 25 mm) – ignition near the wall of the channel
  - shorter tubes (L < 25 mm) -„diffusive ignition"
- Tube length dependence – similar as in experiments
- tube diameter influence visible for longer tubes



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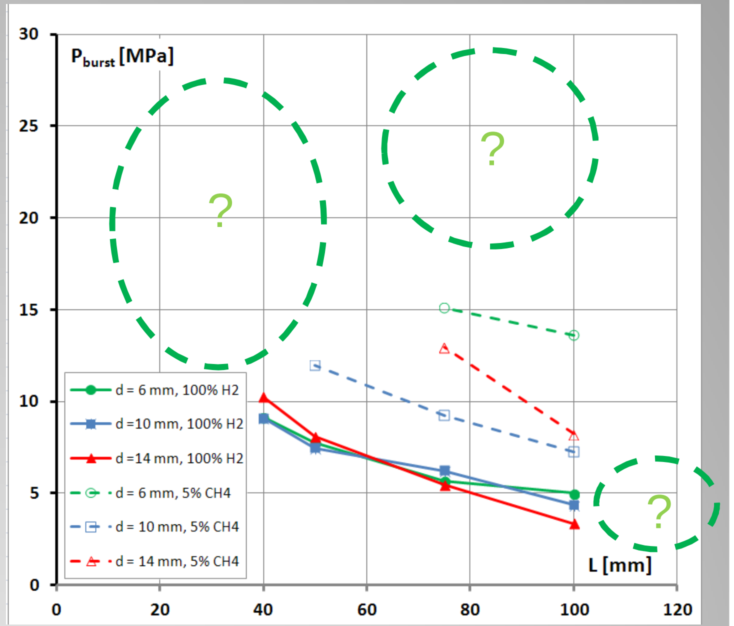
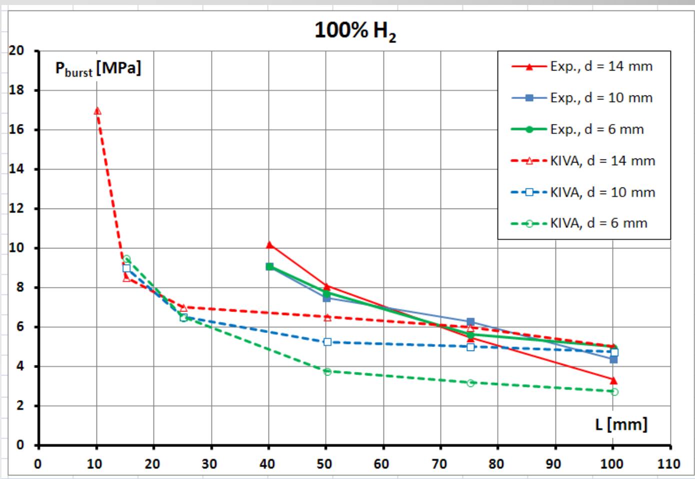
6. Future research

Experimental research:
- more tests for 5% CH_4
- higher pressures (for 10% CH_4)
- tubes with L < 40 mm
- hydrogen-nitrogen mixtures?
- diameter influence for longer tubes more evident?



Numerical research:
- simulations with detailed H_2-CH_4 reaction mechanism
- membrane rupture rate



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7. Acknowledgements

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8. References

P. Wolanski, S. Wojcicki, *Proc. Combust. Inst.* 14 (1972) 1217-1223.

V.V. Golub, D.I. Baklanov, T.V. Bazhenova, M.V. Bragin, S.V. Golovastov, M.F. Ivanov, V.V. Volodin, *J. Loss Prev. Process Ind.* 20 (4-6) (2007) 439-446.

F.L. Dryer, M. Chaos, Z. Zhao, J.N. Stein, J.Y. Alpert, C.J. Homer, *Comb. Sci. Technol.* 179 (4) (2007) 663-694.

T. Mogi, D. Kim, H. Shiina, S. Horiguchi, *J. Loss Prevent. Process. Ind.*, 21(2) (2008) 199-204.

T. V. Bazhenova, V. V. Golub, I. N. Laskin, N. V. Semin: 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.

T. Mogi, Y. Wada, Y. Ogata, A.K. Hayashi, *Int. J. Hydrogen Energy* 34 (2009) 5810-5816

P. Oleszczak, P. Wolanski, *Shock Waves* 20 (2010) 539-550.

W. Rudy, A. Dabkowski, A. Teodorczyk, *Archivum Combustionis* 30 (1-2) (2010) 37-52, available online: http://archcomb.itc.pw.edu.pl

H.J. Lee, Y.R. Kim, S.H. Kim, I.S. Jeung, *Proc. Combust. Inst.* 33 (2011) 2351-2358.

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

Y.R. Kim, H.J. Lee, S. Kim, I.-S. Jeung, *Proc. Combust. Inst.* 34 (2) (2013) 2057-2064

S. Kim, H.J. Lee, J.H. Park, I.-S. Jeung, *Proc. Combust. Inst.* 34 (2) (2013) 2049-2056

J.X. Wen, B.P. Xu, V.H.Y. Tam, *Combust. Flame* 156 (11) (2009) 2173-2189

B.J. Lee, I.-S. Jeung, *Int. J. Hydrogen Energy* 34 (2009) 8763-8769

M.V. Bragin, V.V. Molkov, *Int. J. Hydrogen Energy* 36 (2011) 2589-2596.

E. Yamada, N. Kitabayashi, A. Koichi Hayashi, N. Tsuboi, *Int. J. Hydrogen Energy* 26 (2011) 2560-2566.

A. A.Amsden: *KIVA-3V: A Block-Structured KIVA Program for Engines Vertical or Canted Valves* , Los Alamos National Laboratory report LA-13313-MS, July 1997.

A.A. Konnov: *Detailed reaction mechanism for small hydrocarbons combustion Release 0.5*, 2000.

Alcock, J.L., Shirvill, L.C., and Cracknell, R.F. (2001) Compilation of Existing Safety Data on Hydrogen and Comparative Fuels. Report EIHP 2, Fifth Framework Programme (1998–2002), Shell Global Solutions.

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