

# Temperature effect on explosion parameters of hydrogen-air deflagrations in presence of water vapor

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Results of investigation of hydrogen-air deflagrations phenomenon in closed vessel in various initial temperatures and volume fraction of water vapor are presented in following paper. Tests were performed in apparatus which construction complies with EN 15967 recommendations—20-litre sphere. Studied parameters were explosion pressure ( $P_{ex}$ ) and maximum explosion pressure ( $P_{max}$ ). Defining the influence of the initial conditions (temperature and amount of water vapor) on the maximum pressure of the hydrogen-air deflagration in a constant volume was the main aim. Initial temperatures were equal to 373K, 398K and 413K. Initial pressure was ambient (0.1 MPa). Hydrogen volume fraction differed from 15% to 80%, while humidity volume fraction from 0% to 20%. Ignition source was placed in geometrical center of testing chamber and provided energy between 10-20J from burnout of fuse wire with accordance to abovementioned standard. Common features of all experimentally obtained results were discussed. Maximum explosion pressure ( $P_{max}$ ) decreases with increasing the initial temperature. Furthermore, addition of the water vapor for constant initial temperature decreases value of  $P_{max}$  and shifts the maximum peak to the direction of lean mixtures. Data provided in paper can be useful in assessment of explosion risk of industry installations working with hydrogen-air atmospheres with high water vapor addition.

**Keywords:** explosion pressure, hydrogen, closed vessel, elevated temperature, water vapor, deflagration

## Introduction

Explosion pressure  $P_{ex}$ , besides flammability limits, is a key parameter that features hazardous materials in the field of their combustible nature. According to definition it is the highest pressure that occurs in closed vessel (or any confinement) during the explosion of flammable mixture in air or in air with inert gas. This value is measured for specific volume fraction of fuel [1]. Conducting a series of measurements for different volume fractions of fuel leads to discovering its maximum value, i.e. maximum explosion pressure  $P_{max}$  [1].

There are a lot of well-known examples of industrial processes at which elimination of flammable atmosphere is impossible due to their economical and/or physicochemical determinations. The active methods of explosion protection are the only solution in order to avoid pressure loading. An example is using venting devices with no-reusable elements (e.g. bursting membranes) or with reusable elements (e.g. weight-loaded explosion doors). Successful designing of mentioned systems is impossible without knowledge of a specific explosion parameters which can be confirmed by various norms, standards and guidelines [2, 3].

Studies prove that determination of explosion parameters is dependent on factors that can be classified into two (or three) groups: factors related to the testing procedure and/or apparatus (e.g. volume and shape of testing vessel or amount of energy, type and placement of ignition source), factors related to physicochemical background of determination process (e.g. temperature, pressure, turbulence of flammable atmosphere at the moment of ignition or humidity of oxidizing medium) and (in some cases) human factors [4]. Knowledge

of the factors that influence  $P_{ex}$  can be crucial for designing proper explosion protection systems.

Hydrogen is considered to be fuel of the future [5]. There are still some unresolved issues concerning usage of hydrogen that need to be addressed. Hydrogen can be extracted from natural gas and coal beds, which results in the great carbon dioxide emissions into the atmosphere. Another method of producing discussed element is electrolysis (still not efficient). In this process chemical bond between constituting atoms of water compound is being broke due to direct electrical current [5]. Hydrogen can also be obtained in atomic power plants, due to zirconium-steam reaction at high temperatures. Necessity of investigation of how hydrogen-air explosion parameters are dependent on humidity, initial temperature and fuel volume fraction therefore seems justified.

First sources of information about explosive nature of hydrogen were presented by numerous authors [6-8] in the past. Recently a lot of scientific effort was put into understanding the flammability characteristics of hydrogen. Some explosion parameters were determined for hybrid mixtures of hydrogen with addition of different flammable dusts, e.g. graphite [9], cork [10] or active carbon [11]. Hydrogen-air mixture combustion parameters, like flame speed, flame duration, peak pressure, maximum explosion pressure and deflagration indexes, were studied by Li et al. [12], also at the presence of methane. Great contribution into understanding  $H_2$  destructive potential has Van den Schoor [14-16], whose studies were focused on methods for determination of flammability limits in methane/hydrogen/air mixtures [13], upper explosion limit of lower alkanes and alkenes in air at elevated pressures and temperatures [14], influence of the ignition source location on the determination of the explosion pressure at elevated ini-

tial pressures [15] and upper flammability limit of methane/hydrogen/air mixtures at elevated pressures and temperatures [16]. Holborn et al. [17] investigated possibility of modeling the effect of water fog on the upper flammability limit of hydrogen-oxygen-nitrogen mixtures. Ren and Zhang [18] evaluated influence of hydrogen distribution in presence of air on measured flammability limits. Rudy et al. [19] studied (both experimentally and numerically) spontaneous ignition of hydrogen and hydrogen-methane jets into air. Liu and Zhang [20] reported significant influence of initial pressure and temperature on flammability limits of hydrogen-air mixtures. Sanchez and Williams [21] prepared state-of-art paper in which they have presented wide and comprehensive overview on hydrogen flammable nature that was recently reported in the literature.

Besides hydrogen, Razus et al. [22-25], di Benedetto et al. [26-29], Li et al. [30, 31], Flasińska et al. [32], Grabarczyk et al. [33] and Gieras et al. [34-36] independently determined various explosion parameters of several hydrocarbons in closed vessels of different geometry, both in shape and volume. Earlier, theoretical work was made by Fan with Crowl [37] and by Bradley with Mitcheson [38]. Their achievements were developed further by Giurcan et al. [39] who attempted to predict flammability limits of fuel-air and fuel-air-inert mixtures from explosion parameters determined in closed vessels.

This paper presents research results defining explosion pressure  $P_{ex}$  for hydrogen-air mixtures with and without presence of water vapor (humidity) at various ratios. Initial temperatures were equal to 373K, 398K and 413K. Initial pressure was ambient. Hydrogen volume fraction differ from 15% to 80%, while humidity volume fraction from 0% to 20%.

## Experimental designs

Measurements were performed in 20-liter spherical explosion vessel that complies with EN 15967 recommendations [1]. Picture and scheme of testing apparatus is shown in Figure 1. Ignition source was supplied by overvoltage of fuse wire type Kanthal D with 0.2 mm diameter. This implementation allows receiving energy between 10 and 20J, which do not influence obtained results [1]. Performing tests at elevated initial temperature was possible due to thermal insulation and temperature stabilization modules.

Apparatus has also safety module, mixture preparation system module and data acquisition system. First consist a bursting membrane with activation pressure equal to 20 bars. Second is a set of following devices: pressure transducer, signal amplifier and computer that records phenomenon with adequate sampling frequency. Obtained signals were smoothed according to algorithm based on concept of moving average. Last module works on basis of partial pressure method.

Procedure started with clearing (creating absolute pressure) testing vessel and gradually adding further constituents. Application of constituents was as follows: water (with automatic pipette), hydrogen and synthetic air (less than 5 ppm of water). Water priority was justified by need of its full evaporation. After each step it was necessary to note pressure inside the vessel as (according to Dalton's Law and ideal gas model) volume fraction of single constituent is equal to its par-

tial pressure divided by total pressure in vessel. It was crucial to consider residual amount of air in vessel while calculating volume fraction of each component. Pressure gauge installed in apparatus allowed to measure underpressure with accuracy up to 1 mbar. Volume of channels that provides constituents is assumed to be negligible in comparison with whole vessel.

Experiments were performed mostly for fuel-rich atmospheres in which addition of water in vapor state (humidity) decreases amount of oxidizer in a whole mixture. For composition with hydrogen volume fraction lower than its stoichiometric value, the amount of oxygen is great enough for reaction to be nearly perfect, while difference between heat capacities of oxygen-nitrogen mixture and water in vapor state is not significant enough to influence the  $P_{ex}$  values.

Upper flammability limits of hydrogen are (depending on the source) equal to 75% for ambient conditions (298 K and 0.1 MPa) and measured according to T method of EN 1839 standard [40]. Before performing measurements, additional pre-experimental calculations were made, which covered influence of temperature on upper flammability limit of hydrogen. According to Liu and Zhang [20] for ambient pressure (0.1 MPa) UFLs of H<sub>2</sub> in 294K, 313K, 333K, 348K and 363K are equal (respectively) 76.5%, 82.5%, 84.5%, 85.5% and 86.5% with possible error of  $\pm 0.5\%$ . Above values were extrapolated with Curve Fitting Toolbox [41] in Matlab 2014b [42] for 378K, 398K and 423K giving following values of UFLs: 87.1%, 89.0% and 91.7%. Those values were assumed to be indicative boarder volume fractions of hydrogen for following measurements.

Amounts of water applied inside chamber were measured with automatic pipette and were equal

0.5 ml, 1 ml and 2 ml. Tests for dry mixtures were also performed (0 ml of water). Hydrogen volume fractions in experiments varied from 15% to 80% (or less if addition of water decreases upper flammability limits faster than temperature increase it) with 5% step. Synthetic air with less than 5 ppm of water was applied into the vessel as the last constituent until total pressure of mixture was equal to the atmospheric one. Oxygen to nitrogen ratio was equal 0.2939 per its provider (The Linde Group—Poland [43]).

## Data evaluation

Experimental results were analyzed with Matlab [42]. With use of Curve Fitting Toolbox [41] tendencies of  $P_{ex}$  versus hydrogen volume fractions were computed for every case (Figure 2-4).  $P_{ex}$  were described as follows:

$$P_{ex} = f(X, v_{H_2}, T_0) \quad (1)$$

where:

- X is volume fraction of water in its vapor state,
- $v_{H_2}$  is volume fraction of hydrogen and  $T_0$  is initial temperature of vessel. Maximum values of  $P_{ex}$  are the maximum pressures of the constant volume explosions ( $P_{max}$ ) (see Table 1).

Amounts of water applied inside chamber were measured with automatic pipette and were equal 0.5 ml, 1 ml and 2 ml. Same amount of water in its liquid state ( $v_{H_2O}^{(l)}$ ) occupy different volume at different temperature. Table 1 contains information

Table 1. Maximum explosion pressure ( $P_{max}$ ) and volume fraction of water in vapor state (X) for measurement in each temperature ( $T_0$ )

$T_0$ [K]	378K		398K		423K		
	$v_{H_2O}^{(l)}$ [ml]	$P_{max}$ [bar]	X [% vol.]	$P_{max}$ [bar]	X [% vol.]	$P_{max}$ [bar]	X [% vol.]
0		4.88	0%	4.57	0%	4.31	0%
0.5		4.78	4.18%	4.48	4.48%	4.13	4.77%
1		4.66	8.35%	4.31	8.95%	3.98	9.54%
2		4.35	16.71%	3.87	17.90%	3.46	19.08%

about volume fraction of water in its vapor state (X) obtained from applied  $v_{H_2O}^{(l)}$  into the testing vessel. Values of parameter X were obtained from tables containing physicochemical properties of water and steam [44] and confirmed experimentally with measurements of partial pressures of humidity (injected water).

For each composition of hydrogen-water-air mixture at every temperature measurements in number of five were performed. Standard deviations ( $\sigma$ ) were calculated for each case. Maximum standard deviations ( $\sigma_{max}$ ) and mean standard deviations ( $\sigma_{mean}$ ) were calculated for each temperature and were equal to 0.22 bar and 0.06 bar for 373K, 0.18 bar and 0.04 bar for 398K, 0.17 bar and 0.09 bar for 423K respectively. Markers on plots represents experimental values, while lines stands for computed tendencies of  $P_{ex}$ .

## Results and discussion

Figure 2-4 presents obtained results for 373K, 398K, 423K respectively. It can be seen that maximum pressure ( $P_{max}$ ) decreases with increasing the initial temperature, which is in agreement with literature [31, 39]. It is an expected outcome, because density decreases with increase of temperature. Consequently, in the same volume can be stored less matter (e.g. fuel), so less heat from combustion can be released and lower pressure arise will be observed. What is more, addition of the water vapor for constant initial temperature decreases value of  $P_{max}$  and slightly shifts the maximum peak to the direction of lean mixtures. Secondly,  $P_{max}$  values were measured for hydrogen volume fractions close to stoichiometric values (~30%), but from side of fuel rich mixtures. Fuel surplus is essential for effective extraction of energy from combustion reaction, because it ensures that every oxygen molecule will be paired with fuel molecule. Additionally,  $P_{ex}$  tendencies for different temperatures seems to converge towards LFL. Authors interpret this effect on ground of negligible change of LFL value between 373K and 423K. It was also noticed that explosion pressures assume lower values for fuel rich mixtures than for fuel lean mixtures, but range of flammable fuel volume fractions on their side (fuel rich mixtures) is wider. Additionally, lack symmetry between fuel rich and fuel lean mixtures was observed. All  $P_{ex}$  tendencies for various amount of water vapor as well as for various initial temperatures are arranged into layers in (almost) whole range of hydrogen volume fractions. For such cases the influence of temperature on UFL is too gentle to affect the  $P_{ex}$  characteristics as a function of hydrogen volume fraction, therefore no trend inversion can be observed.

The difference between boundary temperatures (373K and 423K) are equal 0.57 bar, 0.65 bar, 0.68 bar and 0.89 bar for  $v_{H_2O}^{(l)}$  equal 0 (dry composition), 0.5 ml, 1 ml and 2 ml

respectively. It is noticeable that differences increase with increase of amount of water additive ( $v_{H_2O}^{(l)}=0$ ). Authors assume that initial temperature have an impact on physicochemical properties (like heat capacity or density) of both water and steam.

## Conclusion

This paper presents the results of experimental research performed in a 20-litre closed spherical explosion vessel where combustion of hydrogen-air mixture was investigated. The results show that initial temperature influence maximum pressure of explosion. Addition of water vapor to the hydrogen-air mixture resulted in decreasing value of the  $P_{max}$ . Following measurements may be needed for promoting a database of explosion parameters of mixtures based on  $H_2$  in a wide range of initial conditions. Presented results can also be useful in evaluating explosion risk of and/or in selection of explosion protection systems for hydrogen-based flammable atmospheres. Results obtained by described means possess high level of safety for industrial use.

## Acknowledgements

This work was financed by Faculty of Power and Aeronautical Engineering of Warsaw University of Technology from its Dean's subsidy for years 2014 and 2015. Authors also would like to express their highest gratitude for Mr. Andrzej Kołaczowski from ANKO-lab Company [45] for his contribution as a designer and supplier of testing apparatus and as a consultant in designing presented experiments. Fellow researchers from Scientific and Research Center for Fire Protection in Jozefów also deserves thankfulness [46].

## Nomenclature and abbreviations

- $p_0$  – initial pressure
- $P_{ex}$  – explosion pressure
- $P_{max}$  – maximum explosion pressure
- $T_0$  – initial temperature
- $v_{H_2}$  – volume fraction of hydrogen
- $v_{H_2O}^{(l)}$  – amount of injected water into apparatus in its liquid state
- X – volume fraction of water constituent in its vapor state

Greek letters:

- $\sigma_{max}$  – maximum deviation for measurement in single temperature
- $\sigma_{mean}$  – mean deviation for measurement in single temperature

## Figures

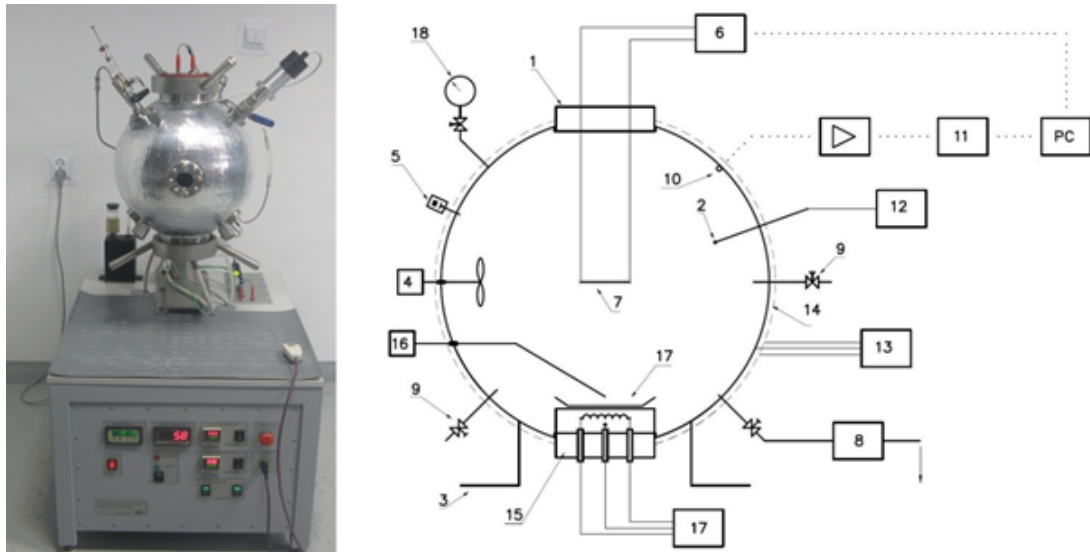


Figure 1 [45]. Apparatus picture and scheme with description: 1) upper head of reactor carrying ignition source module; 2) thermocouple for measuring temperature inside the reactor; 3) supports on which chamber is founded; 4) homogenizing system for preparation of tested mixtures; 5) safety module; 6) trigger for initiating data acquisition system coupled with ignition system; 7) ignition source; 8) vacuum pump with filter and shut-off single-sided valve; 9) 1/2" sockets for connecting additional components or modules if needed; 10) dynamic pressure sensor with signal transducer and analog amplifier; 11) data acquisition system that links PC with dynamic pressure sensor; 12) device for regulation of measured value (with display screen) of temperature inside reactor; 13) system for temperature stabilization of testing reactor (immersion heaters, thermocouples and digital controller); 14) thermal insulation of reactor (outer casing that separates surrounding and testing vessel); 15) lower head with evaporation module; 16) system for feeding  $H_2O$  component in its liquid state into evaporation module; 17) evaporation module (heated tray with temperature stabilization system); 18) pressure gauge for measuring static pressure inside reactor during measurement preparation procedure (disconnected from apparatus during actual measurement)

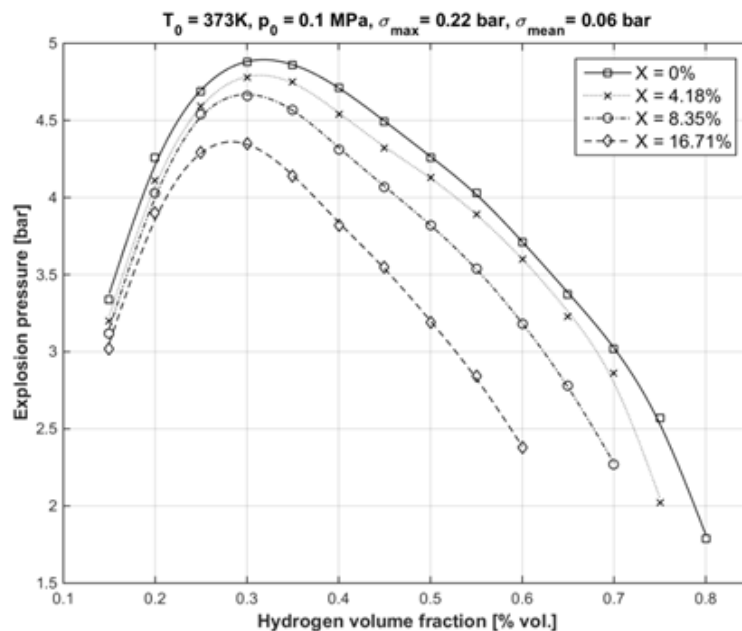


Figure 2. Explosion pressure tendencies for initial temperature equal 373K for different volume fractions of fuel (hydrogen) and volume fractions of water in its vapor state ( $X$ )

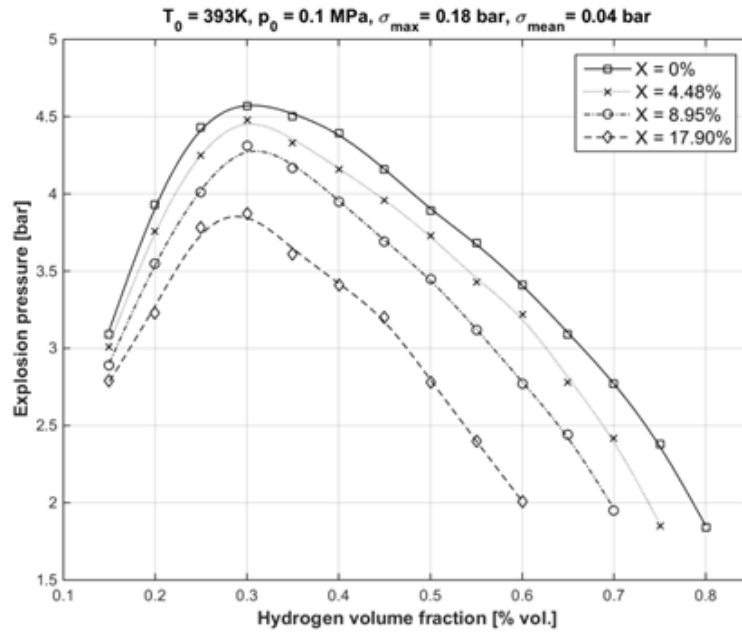


Figure 3. Explosion pressure tendencies for initial temperature equal 398K for different

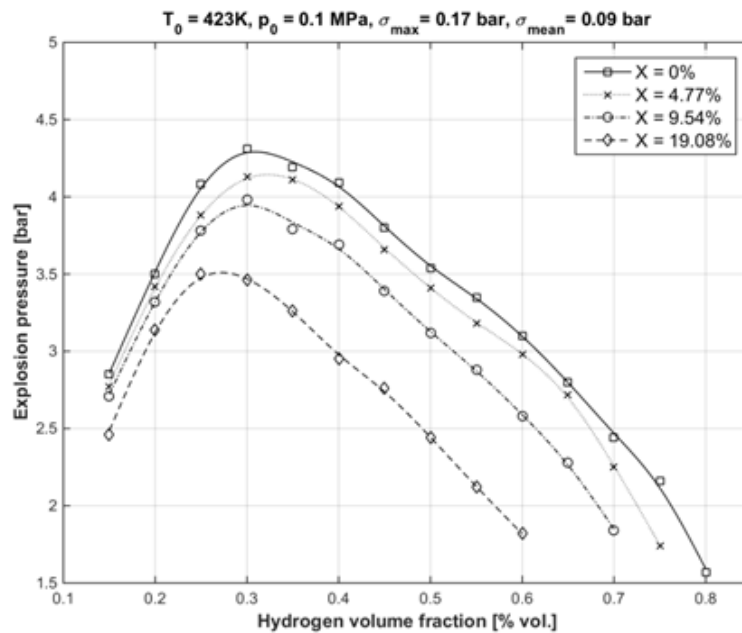


Figure 4. Explosion pressure tendencies for initial temperature equal 423K for different volume fractions of fuel (hydrogen) and volume fractions of water in its vapor state (X)

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