The Lower Flammability Limit of Hydrogen

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Abstract

Distribution of pure hydrogen through the existing natural gas grid is one of the options to feed industry and provide a heating source for the domestic environment. Alliander demonstrates this with her project in Lochem, where 12 houses are being heated with 100% hydrogen[1]. However, hydrogen is considered to be a highly flammable and explosive gas. In most literature no distinction is made between the flammability limits and the explosion limits, leading to the common understanding that hydrogen/air mixtures of 4% and upwards are explosive. Furthermore, the necessary ignition energy is commonly perceived as extremely low.

In 1952, The US Bureau of Mines published the article 'Limits of flammability of gases and vapors'[2]. Their research shows that hydrogen in a mixture between 4% and 6% only ignites molecules that are directly above the burning molecule. Between 6 % and 9 % the neighboring molecules will also be ignited, and only above 9 % hydrogen in air the flame will spread in all directions. Unfortunately, this knowledge did not become common, and much literature states that hydrogen can explode at 4% in air.

Kiwa Technology realized a test setup to visualize the research explained above. This resulted in high-speed videos, made from ignition of both hydrogen and natural gas at differing concentrations. These videos show that at low concentrations, the probability of ignition of hydrogen is lower compared with natural gas. It was discovered that in practice in this test set up, it was not even possible to ignite hydrogen at concentrations up till 6%. This can be of particular relevance for the safety of service engineers and firefighters. Currently, explosion limits (LEL/UEL) of flammable gases, such as hydrogen, are determined by the detachment and upward movement of the flame, or the formation of a halo in a test-tube[3]. This can also be observed at the lower percentages as described above. There is a huge difference in risk appreciation whether a mixture will only burn (upwards, sideways or downwards) or produce the amount of combustion gases that can lead to an explosion with damage.

[1] https://www.alliander.com/en/news/a-global-first-alliander-heats-homes-using-hydrogen-through-the-existing-gas-grid/
[2] https://digital.library.unt.edu/ark:/67531/metadc12662/m2/1/high_res_d/Bulletin0503.pdf
[3] https://www.nen.nl/en/nen-en-1839-2017-en-229549

Alliander and Kiwa Technology urge the international research and standardization community to conduct research to diversify in differing practical implications of flammable and explosive situations. By doing so a firm base can be created to reflect the practical outcome in standardization. Also, the possibility should be investigated to come with more fitting LEL/UEL definitions than currently given in EN 1839:2017, where flammability and explosion are considered to be the same. This may accelerate a future where hydrogen is part of the common, public energy system.

Introduction

Distribution of pure hydrogen through the existing natural grid is one of the options to feed industry and provide a heating source for the domestic environment. Alliander, being one of the leading regional gas grid operators in the Netherlands, demonstrates this with a trial in Lochem, where 12 houses are being heated with pure hydrogen, whilst using the current gas grid. In preparation of this trial, Alliander conducted various research into the physics of hydrogen and associated risks and the potential need for mitigating measures. Hydrogen is considered to be a highly flammable and explosive gas. Most literature (including relevant norms) does not distinguish between flammability and explosion limits of these gasses, leading to the common understanding that hydrogen/air mixtures are explosive from concentrations as low as 4% hydrogen in air. Furthermore, the ignition energy is commonly perceived as extremely low. We see this reflected for instance in -EN 1839:2017. This implies a higher hydrogen risk profile, compared to natural gas. Alliander also perceived hydrogen to have a high safety risk profile then natural gas. However, this was challenged during preparation of the Lochem trial.

In 1952, The US Bureau of Mines published a 170-page article in Bulletin 503, on the 'Limits of flammability of gases and vapors. This research shows that a hydrogen air mixture between 4% and 6% only ignites molecules that are directly above the burning molecule. Between 6% and 9% the neighboring molecules will also be ignited; only with concentrations above 9% hydrogen in air will the flame front will spread in all directions. At percentages below 9%, the upward convection is stronger compared to the flame speed. This knowledge did not become common, and much literature wrongly states that hydrogen can explode at 4% hydrogen in air.

To understand the results of the Bureau of Mines, Kiwa Technology, a Dutch technological research consultancy firm, designed a test set-up, in order to visualize the ignition and flame front of hydrogen at low concentrations with varying ignition sources. This was done to share the resulting high-speed videos with field engineers, firefighters the Dutch safety regulator gas and relevant permitting bodies.

Theoretical background

This part provides the theoretical background on ignition energy and flammability of hydrogen in comparison to natural gas. The Minimum Ignition Energy (MIE) of hydrogen is 0,0017 mJ at the stoichiometric mixture. Most ignition sources can reach this and therefore ignite hydrogen. However, the ignition energy is a function of the gas/air ratio with much higher ignition values at low hydrogen in air concentrations. The ignition energy for a hydrogen mixture between 0-10% is comparable to methane (figure 1).

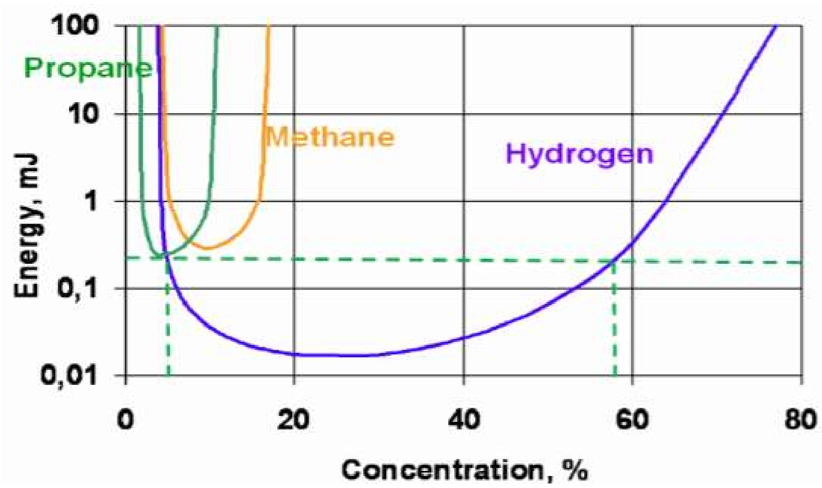

Figure 1: The ignition energy is depending on the amount of gas in air.

The MIE of hydrogen is reached at the stochiometric mixture of 29,5%, whereas the MIE of natural gas is reached at a stoichiometric mixture of 9,5% gas in air. This is not the only difference: with natural gas the LFL(5.1%) and LEL(5.3%)[4] almost have the same value, and these are often interchanged. For hydrogen these values are further apart. The theoretical LFL is 4,1 % and the LEL is not an exact value and depends on multiple variables. It is somewhere between 15% and 20% hydrogen in air also depending on the definition of an explosion. Most literature states that hydrogen has an LFL/LEL for hydrogen of 4.1%. This low concentration and needed ignition energy, would imply that hydrogen has a higher risk profile, compared to natural gas. However, the article of the bureau states of mines on ‘Limits of flammability of gases and vapors’ shares a more nuanced view on flammability limits of hydrogen.

Limits of flammability of hydrogen-air mixtures saturated with water vapor

	Limits, percent Lower	Limits, percent Higher
Upward propagation	4. 1	74
Horizontal propagation	6. 0	
Downward propagation	9. 0	74

Table 1: flame propagation of hydrogen. Source: Bureau of Mines, Bulletin 503

[4] There are some variations because natural gas is a mixture of methane with higher hydrocarbons and differs depending on the extraction location.

As can be seen in table 1 it is possible to ignite hydrogen at 4.1% but the flame only propagates in an upward direction. The main driver for this is, that at this percentage the upward convection is stronger than the flame speed. In addition, the flame temperature of the burning molecules is around 350 °C, which is insufficient to ignite neighboring molecules in a horizontal direction. The temperature needed is 585 °C. This knowledge did not become common, and many literatures wrongly states that hydrogen can explode at 4% hydrogen in air.

Test set up

Kiwa Technology designed a test setup to visually show lower flammability limits and the process of combustion (including impact on cotton) by exposing differing hydrogen-air mixtures to varying ignition sources. The test box used, measured 23,5 cm x 23,5 cm x 42,5 cm (width x depth x height). The top of the test box was equipped with a simple pressure release system, to prevent unwanted build-up of pressure and mitigate the potential risk of an explosion. The setup was fitted with ignition sources that are common in the build environment (e.g. within a residential home):

- Common sparkplug like used in boilers.
- Typical doorbell ringer.
- Light switch connected to a bulb (75w)
- A burning cigarette.

A tube was mounted (airtight), through the side of the textbox to allow for the adding of gasses under controlled circumstances. Also, a cotton fiber was mounted hanging in the test box.

Figure 2: Test setup to visualize hydrogen flammability. Source: Kiwa Technology

A wide variety of tests was executed: different hydrogen air mixtures (2% increments) ranging from 4% up to 10% were added to the test box, whilst activating the different ignition sources in turn. For comparative reasons, the same tests were repeated with natural gas air mixtures. The resulting gaseous behavior was filmed with help of a high-speed camera fitted with schlieren optics. This method of filming was necessary to make the ignition-behavior visible to the human eye.

Results

Above (please see paragraph theoretical background), 4,1% hydrogen into air was mentioned as a found LFL. This limit was not confirmed during the practical tests; during the tests it was not possible to ignite the mixture at the LFL of 4,1%. The following behavior was recorded with different mixtures:

- Ignition of a hydrogen mixture was unsuccessful with a 6% hydrogen into air mixture or less, regardless of the source of ignition
- Ignition of an 8% hydrogen air mixture was successful, with the flame front primarily moving upwards with a relative steady pace.
- Ignition of a 10% hydrogen air mixture was successful, with the flame front spreading in all directions in a fast and turbulent way.

Stills from the schlieren-camera recording are shown in figure 3: the movement in the left picture (6% hydrogen in air) is only the rising of the heated mixture, without any flames.

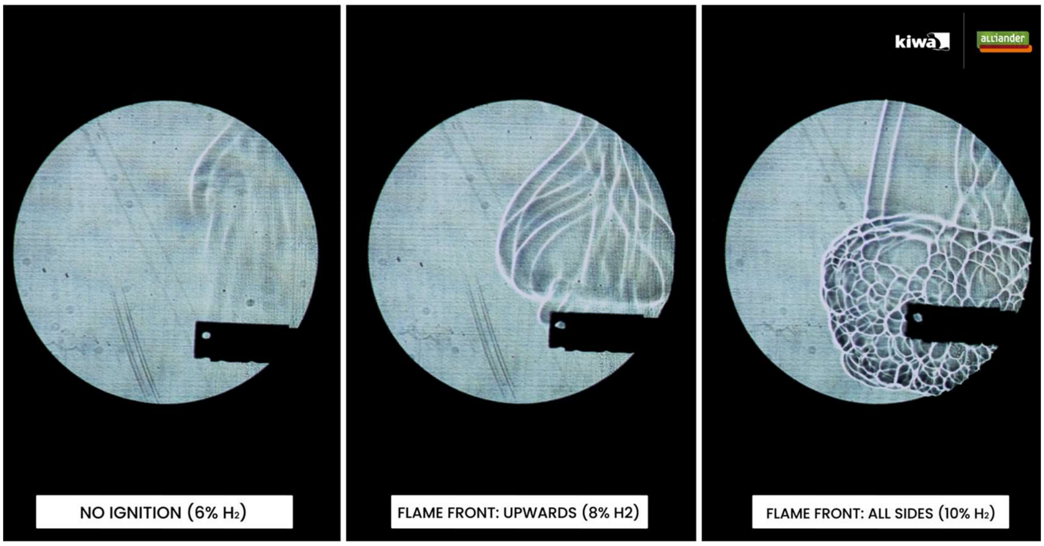

Figure 3: Flame front with different hydrogen-air mixtures

The lower flammability limit and development of flame front was found to be dependent on the concentration of hydrogen into air only, and not dependent on the tested sources of ignition. The burning cigarette, as ignition source, was unable to ignite any hydrogen air mixture: the cigarette did not become hot enough, with a max temperature of 400 °C. This is consistent with the theoretical background, as explained above.

The mounted cotton fiber provides an indication of impact of the flame front on materials in its radius. It is here where the reference tests with methane gas show a profound difference:

- A methane air mixture with a gas concentration of 6% in air ignites; the radiant heat instantly burns the cotton fibers completely. This mixture is $2/3^{rd}$ of the optimal 9.5% stochiometric methane gas in air concentration, leading to $2/3^{rd}$ of the potential maximum radiant heat.
- A 10% hydrogen-air mixture ignites as well, but the flame front radiates insufficient heat to burn the cotton fibers. A 10% hydrogen-air mixture is $1/3^{rd}$ of the optimal stochiometric hydrogen in air concentration, and therefore also $1/3^{rd}$ of the potential maximum radiant heat.

Relevance of the test results

The relevance of difference between the LEL and LFL, as well as the differing flame fronts of hydrogen and natural gas may be profound for a gas grid operator. During the tests a maximum 10% gas in air was chosen as a practical limit to explain the relevance in practice. First the chance of an occurrence has been considered.

- The chance that a mixture will ignite below 10% is for hydrogen lower than for natural gas in the test. In the same test set up it was not possible to ignite hydrogen below 6%, but it was possible to ignite natural gas at 5.25% gas in air.

If the effect of an occurrence is being considered the following can be found, for the mentioned low concentration of hydrogen:

- Although there is ignition at 8% in the test the flame only propagates upwards. This means people and material below the ignition point are not immediately affected.
- Due to the fact that the flame only propagates upwards the flame can be self-extinguishing, so if the leakage – which is by definition at a lower point – keeps supplying hydrogen this hydrogen is not being burned by the upward propagating flame. This to the contrary with natural gas, the flame front propagates in all directions and if the leakage persists the flame will not be self-extinguishing. The natural gas flame will burn back to the point of leakage providing the flame with energy.
- The energy of a hydrogen flame is not able to burn cotton, a natural gas flame does burn cotton. This means for a service engineer, who is being confronted with a hydrogen flame, less danger than with a natural gas flame. This also means that a hydrogen flame is not able to ignite building materials like wood, plastic's etc. Natural gas ignites building materials causing damage.
- Below 10% hydrogen burns, but no explosive mixture was found in practice or theory. Natural gas mixtures can explode and may cause more damage than a hydrogen flame.

As can be seen the chance of ignition as well as the effect of ignition for hydrogen gas in air mixtures up till 10% may be smaller than for natural gas. This means that the risk will at least be the same but probably smaller for hydrogen air mixtures below 10% than with natural gas.

In this paper a practical upper limit of 10% hydrogen or natural gas in air is used for the comparison. Alliander researched her database on natural gas leakages in buildings. It is found that the vast majority of leakages in buildings are caused by leakages of couplings or corrosion leaks. Hydrogen in itself has no effect on the formation of leakages in the same pressure range. So the found data set on natural gas seems valid. The smallest confined space in a house in the Netherlands is the meter cabinet, so this is the area where a concentration built up will happen the quickest. In this meter cabinet the meters for electricity, gas and water are being placed. As another preparation for the Lochem trial, Alliander build, together with DNV Groningen, a meter cabinet to measure the concentration of gas in air for natural gas and hydrogen for typical leaks. A concentration of around 0.2% up till 0.3% gas in air has been found for natural gas as well as hydrogen. Although the amount of leakage of hydrogen in volume will be 1,4 (total laminar) up till 3 times (turbulent) more, the concentration is about the same at the four measurement points. The higher buoyance of hydrogen compensates for the extra volume. The Nuclear Research Group (NRG) also performed computational Fluid Dynamics (CFD) calculations for the meter cabinet as has been shown in figure 4.

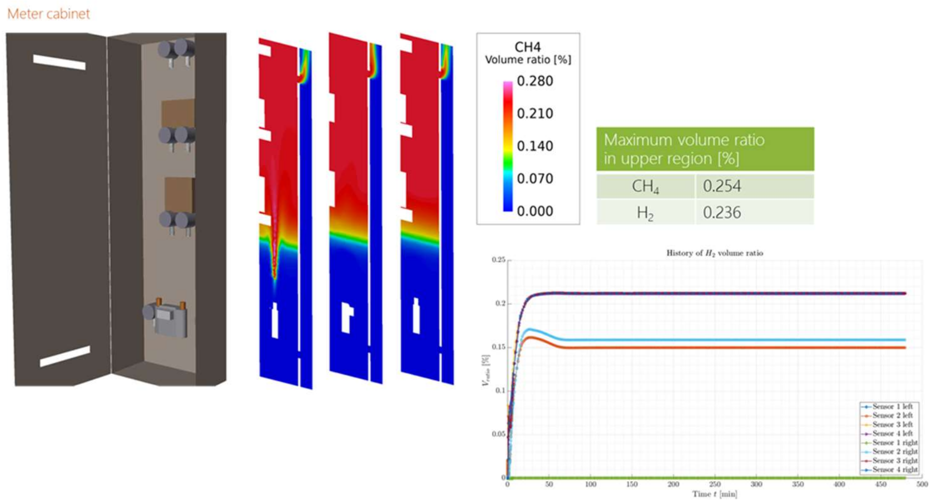

Figure 4 Computational Fluid Dynamics simulations of normal leakages in a meter cabin

As can be seen in figure 4 the same results with the model have been found as in practice. The results of these practical results and the results of the modelling of the meter cabinet seem convincing, that a regular leakage in a building will not lead to high concentrations and remain below the practical upper limit of 10% gas in air. Of course, there is a small chance that a bigger leakage might occur, e.g. when coincidentally a hole is drilled in the in-house piping. To prevent the resulting high out flow, Alliander installed an access flow valve in the saddle between the main and service line connection for each of the houses included in the Lochem trial. The flow allowed is big enough to provide the condensing boiler with enough hydrogen, but a higher rate than the maximum needed for the boiler will lead to a closing of the access flow valve, in turn preventing percentages gas/air mixtures surpassing 10%.

The Minimum Ignition Energy (MIE) of hydrogen is reached at the stochiometric mixture of 29,5%. This is relevant to grid operators, as this is a rather high percentage with a much lower probability of occurrence in practice then the stochiometric mixture of natural gas which is around 9.5% gas in air. Safety measures in the natural gas grid are developed to prevent gas concentrations from rising above 5.1 % natural gas in air. This 5.1 % is chosen as this is in practice both the lower flammability limit and the lower explosion limit of natural gas. Following the same line of thought, the lower flammability limit of hydrogen could be the governing concentration for safe working guidelines.

Call to research and standardization

In preparation of the Lochem trial, Alliander rediscovered the bureau of mines Bulletin 503 paper and validated their conclusions with a practical test together with Kiwa Technology. The resulting video is regularly used by Alliander to share this knowledge with for example permitting bodies. It is found to be a good way of communicating the findings of the Bureau of mines have been made visible and are being placed in a practical perspective.

It was found that literature and standards seldom distinguish between the lower flammability limit and the lower explosion limit of hydrogen air mixtures. Whereas in this paper it is argued that in the practical safety consequences there is a huge difference between these two. Consequently, it is urged to the scientific and standardization community to do further research on this difference and reflect these insights in standards such as EN-1839 or ISO-TR-15916.