

HYDROGEN STORAGE OPTIONS

OVERVIEW AND COMPARISON OF EXISTING TECHNOLOGIES



BSC THESIS REPORT
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Title

Hydrogen Storage Options - Overview and Comparison of Existing Technologies

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ABSTRACT

The current climate crisis and its effects on life on Earth demands societies to move towards more environmentally friendly behavior, involving the substantial reduction of greenhouse gas emissions (GHGs). For the energy sector, such a shift would encompass leaving the fossil fuel era behind and replacing it with renewable energy sources. A promising option in that regard is the use of hydrogen as an energy carrier, which is free of GHGs when the right production process is used (Green Hydrogen). Hydrogen can be implemented in nearly every sector and for a great variety of applications. However, there are still some barriers to its implementation. Especially hydrogen storage seems to be a relatively immature, but rather pressing issue. Therefore, hydrogen storage technology options are investigated, and an overview is being created in which 6 categories are used: Free H₂ Gas (FHG), Free H₂ Liquid (FHL), Free H₂ Solid (FHS), Compound H₂ Gas (CHG), Compound H₂ Liquid (CHL), and Compound H₂ Solid (CHS). After this categorization, the options are compared based on 4 comparison criteria: technological specifications, technology efficiency & readiness, safety, and financial projections. Combining these criteria leads to a top 3 of best options for hydrogen storage. Besides, a top 3 of best options is being created based on the targets for hydrogen storage set by the US Department of Energy (US DOE). Both assessments lead to the conclusion that ammonia, methanol, liquid & compressed hydrogen storage are the most promising technologies currently available.

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LIST OF ABBREVIATIONS

<u>GHGs</u>	Greenhouse gases
<u>RQ</u>	Research question
<u>FHG</u>	Free Hydrogen gas
<u>FHL</u>	Free Hydrogen liquid
<u>FHS</u>	Free Hydrogen solid (<i>adsorption</i>)
<u>CHG</u>	Compound Hydrogen gas
<u>CHL</u>	Compound Hydrogen liquid
<u>CHS</u>	Compound Hydrogen solid (<i>absorption</i>)
<u>MOFs</u>	Metal Organic Frameworks
<u>US DOE</u>	United States Department of Energy
<u>CC</u>	Comparison Criteria
<u>I</u>	Targets (<i>set by US DOE</i>)

INTRODUCTION

The current climate crisis and its (negative) effects on life on Earth demands societies to move towards more environmentally friendly behavior, involving the substantial reduction of greenhouse gas emissions (GHGs). For the energy sector, such a shift would encompass leaving the fossil fuel era behind and replacing it by renewable energy sources, a process that has been referred to as 'decarbonization' or 'greenization' (Bhuiyan & Ibragimova, 2022) & (Acar & Dincer, 2019).

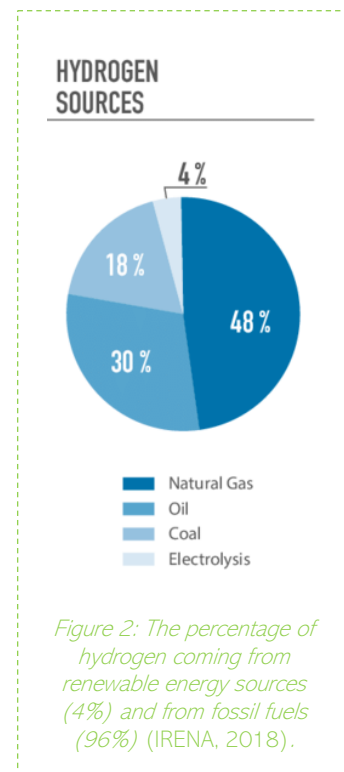
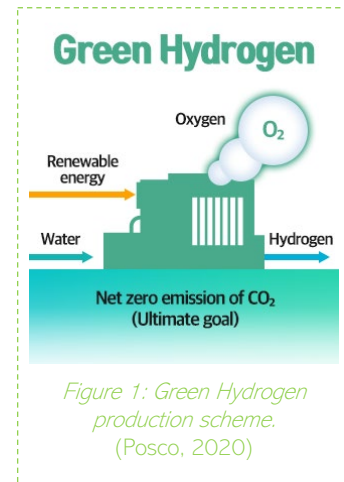
In that regard, hydrogen as an energy carrier could be an interesting alternative (IRENA, 2018). Specifically Green Hydrogen (figure 1), which is produced through the electrolysis of water, fueled by renewable energy sources such as solar and wind power, and does not emit CO₂ emissions (Dincer, 2012). Compared to the fossil fuel-based industries that dominate the world, hydrogen could be a promising alternative for countries on their way to reach the sustainability goals set by the Paris Agreement in 2015: limiting global warming (mainly related to GHGs) to a maximum of 2°C (United Nations Climate Change, n.d.). Hydrogen is envisaged to provide 18% of the global energy demand by 2050 (IRENA, 2018).

Hydrogen is considered most effective as decarbonization agent or as grid balancing service, levelling out the variation in energy consumption and production systems (Oliveira et al., 2021). It could likely live up to the 15% seasonal storage demand needed to fully depend on green (renewable) energy rather than fossil fuels (Oliveira et al., 2021). It could play an important role in the future of renewable energy generation, working towards a so-called Hydrogen Economy in which hydrogen is the primary energy carrier (Abe et al., 2019).

However promising this all sounds, in reality it is not that simple. The total share of hydrogen from renewable energy sources is still only 4% of the total hydrogen production in the world (figure 2), which means that the largest part is still reliant on fossil fuels and does not meet net-zero emission targets (IRENA, 2018). Besides, there seems to be too little capacity to produce the amount of renewable electricity needed to shift to solely Green Hydrogen (Bhuiyan & Ibragimova, 2022). But that's not all, more processes involving hydrogen face implications (Dincer, 2012).

The electrolysis of water for Green Hydrogen production, for example, requires pure water as input to reach optimal efficiency (Urquhart, 2021). That asks for additional desalinization and purification, two energy intensive and costly processes (Urquhart, 2021) & (Beswick et al., 2021). The large energy consumption - partly for electrolysis and partly for other processes - makes that the efficiency of this system does not compete with other types of energy production (Bhuiyan & Ibragimova, 2022). In addition, unforeseen negative impacts on the environment, such as the byproduct 'brine' (a highly concentrated salt with harmful chemicals), may arise (Bhuiyan & Ibragimova, 2022) & (Khan et al., 2021).

Other problems arise with the storage of hydrogen - disregarding its production method - both related to the technology as well as the infrastructure (Bhuiyan & Ibragimova, 2022). Although this aspect is still relatively immature compared to hydrogen production, it might be the most pressing one, especially



considering hydrogen has the possibility to function as a grid balancing service (Longoria et al., 2021) & (Oliveira et al., 2021). Proposed hydrogen storage technology options are not yet ready to be implemented for various reasons, amongst which hydrogen's explosive nature. (Bhuiyan & Ibragimova, 2022) & (Abe et al., 2019). Besides, a hydrogen economy would require a new hydrogen infrastructure, but creating this will bring about costs and possibly other unforeseen consequences (Bhuiyan & Ibragimova, 2022).

Hydrogen has the potential to be applied as an energy carrier in almost all sectors: transportation, commercial, industrial, residential, and portable (Satyapal, 2017). On top of that, it can function in a lot of applications, ranging from vehicles to portable power and from specialty vehicles (i.e. forklifts) to storage and backup. Currently, hydrogen is particularly applied in heavy industries, amongst which refineries (37%), and space applications (1%) (Dias et al., 2020) & (IRENA, 2018). It thus seems to play a vital role in the transition towards a zero-carbon energy sector, or maybe even society. However, only if all barriers are overcome, especially hydrogen storage issues.

Objective

The first part of this thesis aims to create an overview of the storage possibilities for both compound and free hydrogen, categorized into the phases of storage (gas / liquid / solid). The second part aims to compare the hydrogen storage options, focusing on predefined comparison criteria. These are:

- **Technological specifications**, related to the storage conditions;
- **Technological efficiency & readiness**, containing storage efficiency, scale of application and hydrogen & energy content;
- **Safety**: explosiveness and toxicity of compounds;
- **Financial projections**: a cost estimation for hydrogen storage options.

Lastly, the data retrieved during the comparison is combined to form a top 3 of most promising hydrogen storage options, also measured to the goals set by the US Department of Energy (DOE). An excel file with all relevant parameters is provided as an appendix.

Research questions

MAIN RESEARCH QUESTION.

What are currently the most promising and realistic options for hydrogen storage, functioning as a grid balancing service?

RQ1.

What are the hydrogen storage technology options for free and compound hydrogen in gas, liquid, and solid phase?

RQ2.

How do hydrogen storage technology options compare with respect to the predefined comparison criteria: technological specifications, efficiency & readiness, safety, and financial projections?

METHODOLOGY

The goal of this thesis is to create an overview of the already existing technologies regarding the storage of hydrogen, followed by a thorough comparison based on largely quantitative, and partly qualitative properties. The main part of this thesis involves a literature review, with the purpose of gathering information and data to create overview tables for the comparison. This requires thoroughly going through technical articles on specific topics related to hydrogen storage, preferably published in or after 2017, due to the relatively fast developing knowledge in this field. Values retrieved from such articles are not always in the right units and can ask for recalculation, a very basic mathematical application where numbers are converted from MJ to kWh ($1\text{ kWh} = 3,6\text{ MJ}$), € to \$ ($\text{€}1,- = \$0,98$), MPa to bar ($\text{MPa} \cdot 10 = \text{bar}$), K to °C ($1\text{ K} = -272,15^\circ\text{C}$) or \$/kg to \$/ton ($\$1/\text{kg} = \$1000/\text{ton}$). After this step, parameters within the same criteria are compared, providing a reflection of the table in that section.

Finally, the sections are combined in an excel file - without specific calculation steps - to get the 'bigger picture'. A top 3 of best practices in this field will be formed solely based on the parameters provided in this thesis (and the appended excel file), and measured with the targets set by the US DOE. It is therefore not an advice, but rather a general assessment.

To get to the top 3 based on the comparison criteria from [Hydrogen storage technologies | comparison](#), all values in the appended excel file (in the second sheet "Parameters with conditions") are color coded (green cells are desirable, red cells are not) and the number of green cells is counted. This list is then again given a color code and the highest numbers are considered the best options. Penalty points are manually assigned to hydrogen carriers scoring high while some of their characteristics is not desirable, which lowers their value. This results in the top 3 as given in column AC in the excel file.

To get to the top 3 based on the US DOE targets, the table given in [Hydrogen storage technologies | targets](#) is added to the appended excel file in the first sheet "Comparison parameters". In the categories linked to the targets, a color coding is applied (values are colored red when they fit the target). Then, for each option the number of red values is manually counted and added to column W. This results in the top 3 as given in column AA in the excel file.

Combining the comparison parameters in an excel file makes future modelling possible. This thesis thus aims to function as a basis for future research involving modelling of hydrogen storage technologies based on the parameters provided in the excel sheet in the appendix.

THE CHARACTERISTICS OF HYDROGEN

Hydrogen storage functions as a grid balancing service, levelling out the variation in energy consumption and production systems (Møller et al., 2017). However, storage options seem to be limited and restricted by some of hydrogen's characteristics.

Even though hydrogen is the most abundant element in the universe, pure hydrogen gas accounts for only a small fraction of Earth's atmosphere (Encyclopedia Britannica, n.d.). Interactions with other particles can cause this light element to easily escape into space (National Center for Biotechnology Information, 2022). Nevertheless, hydrogen is observed quite often in other forms, as part of a chemical compound, such as water (H₂O) and organic matter (carbohydrates) (Encyclopedia Britannica, n.d.). Hydrogen is the first element in the periodic table, with atomic number Z=1 (Møller et al., 2017).

At normal temperature, the gas is not very reactive without a catalyst - such as air - to bring about an explosion, whereas at high temperatures it is very reactive (Lenntech, n.d.). This leads to difficulties for its storage and implementation, especially considering it is known as the most flammable substance. Through adsorption, hydrogen can turn steel brittle, risky for infrastructural purposes. Too much hydrogen gas in an area can cause oxygen deficiency, resulting in unconsciousness or death amongst people and animals. Regardless of its phase, hydrogen has a gravimetric energy density of 33,3 kWh/kg, which means that its potential energy storage capacity is about 3 times greater compared to fossil fuels (Preuster et al., 2017).

Hydrogen's phases

Hydrogen can be present in gas, liquid, and solid phase. Basic properties are listed in [table 1](#).

Hydrogen gas dominates at ambient conditions (20°C and 1 bar) but can also be compressed for storage purposes. Hydrogen is a colourless, odourless gas, chemically referred to as H₂ (Lenntech, n.d.). It has a density of 0,0899 g/L at room temperature, which is rather low: approximately 14,4 times smaller compared to the density of air (Lenntech, n.d.).

Liquid hydrogen comes with a higher hydrogen and energy density than that of hydrogen gas (Preuster et al., 2017). This is more interesting for storage purposes, but it should also be noted that it comes with a high pressure and low temperature requirement. Hydrogen's boiling point is at 13,8K or -252,8°C, which means that below this temperature - under cryogenic circumstances - the gas will change into a liquid (National Center for Biotechnology Information, 2022).

Solid-state hydrogen can either be found as solid molecular hydrogen or, when put under a huge amount of pressure as observed in Jupiter's core (more than 2Mbar), this type can turn into solid metallic hydrogen (National Center for Biotechnology Information, 2022). Further cooling and/or compression will turn liquid hydrogen into its solid-state (Gregoryanz et al., 2020). However, when referring to "solid-state hydrogen storage", hydrogen is usually still in gaseous or liquid form but forms new solid-state compounds through chemical processes such as adsorption or absorption (Hassan et al., 2021).

Table 1: Basic properties of hydrogen in solid, liquid, and gaseous state (Lenntech, n.d.).

	Hydrogen solid	Hydrogen liquid	Hydrogen gas
Temperature range	Lower than -259,2°C melting point	Between -259,2°C and -252,8°C	Higher than -252,8°C boiling point
Density	⊖ not relevant	70,85 g/L ²⁶	0,0899 g/L at 20°C
Volumetric energy density	⊖ not relevant	2,36 kWh/L ²⁶	1,32 kWh/L ¹³ at 700 bar
Gravimetric energy density	33,3 kWh/kg ²⁶		

HYDROGEN STORAGE TECHNOLOGIES | OVERVIEW

To make a proper assessment of the feasibility of currently proposed hydrogen storage technologies, it is of importance that first an overview of these technologies is being created. Especially considering the enormous number of possibilities. Therefore, this chapter focuses on the following research question:

RQ1. What are the hydrogen storage technology options for free and compound hydrogen in gas, liquid, and solid phase?

There are a lot of possibilities or ideas to store hydrogen nowadays, which makes a correct categorization system a requirement (Andersson & Grönkvist, 2019). For that reason, the options discussed in this paper are categorized by the phase (gas / liquid/ solid) of the storage method, both for free hydrogen and for hydrogen compounds. The result is presented in the schematic overview on the next page (figure 3). However, it should be noted that categorization exists in many forms and not one system is ultimately correct. Additionally, it is considered beyond the scope of this research project to investigate all possible options and therefore only an overview of the most discussed options in literature is given.

The classification system used in this thesis is based on two groups:

- Free hydrogen refers to hydrogen in its pure form, not bound to any other element or chemical. Storage occurs in vessels, pipelines, cavities or in adsorbents, without chemical interaction.
- Compound hydrogen refers to hydrogen bound to another element through chemical processes, forming a new compound.

Within these categories, hydrogen can be present as a solid, liquid or gas, creating a total of 6 categories: Free Hydrogen Gas (FHG), Free Hydrogen Liquid (FHL), Free Hydrogen Solid (FHS) on the one side and Compound Hydrogen Gas (CHG), Compound Hydrogen Liquid (CHL), and Compound Hydrogen Solid (CHS) on the other.

Hydrogen storage sometimes makes use of adsorption processes, where compounds attach to the surface of the adsorbent, divided into physisorption and chemisorption (Helmenstine, 2019) & (Mohan et al., 2019). Physisorption (physical adsorption) processes are based on weak Van der Waals bonds between hydrogen and another compound, whereas chemisorption (chemical adsorption) processes make use of stronger chemical bonds (Agboola & Benson, 2021). These processes play a role in Free Hydrogen storage in Solids (FHS) (Hassan et al., 2021).

For solid-state compound hydrogen storage (CHS) this is similar, making use of absorption processes, where compounds are taken up by the volume of the absorbent (Helmenstine, 2019) & (Hassan et al., 2021).

Chemical hydrides, used for chemical hydrogen storage, are - besides from being compounds - not necessarily linked to a certain category (or in this case: storage phase), but include all compounds obtained by chemical reactions (Hydrogen Tools, n.d.). Chemical hydrides use lighter elements compared to metal hydrides, which means that hydrogen is released more easily generally (Hassan et al., 2021). Examples mentioned in this thesis are ammonia (NH₃) and LOHCs. Therefore, in the schematic overview on the next page, this category is linked to both the liquid and gas phase of compound hydrogen storage.

Schematic overview of available hydrogen storage technologies

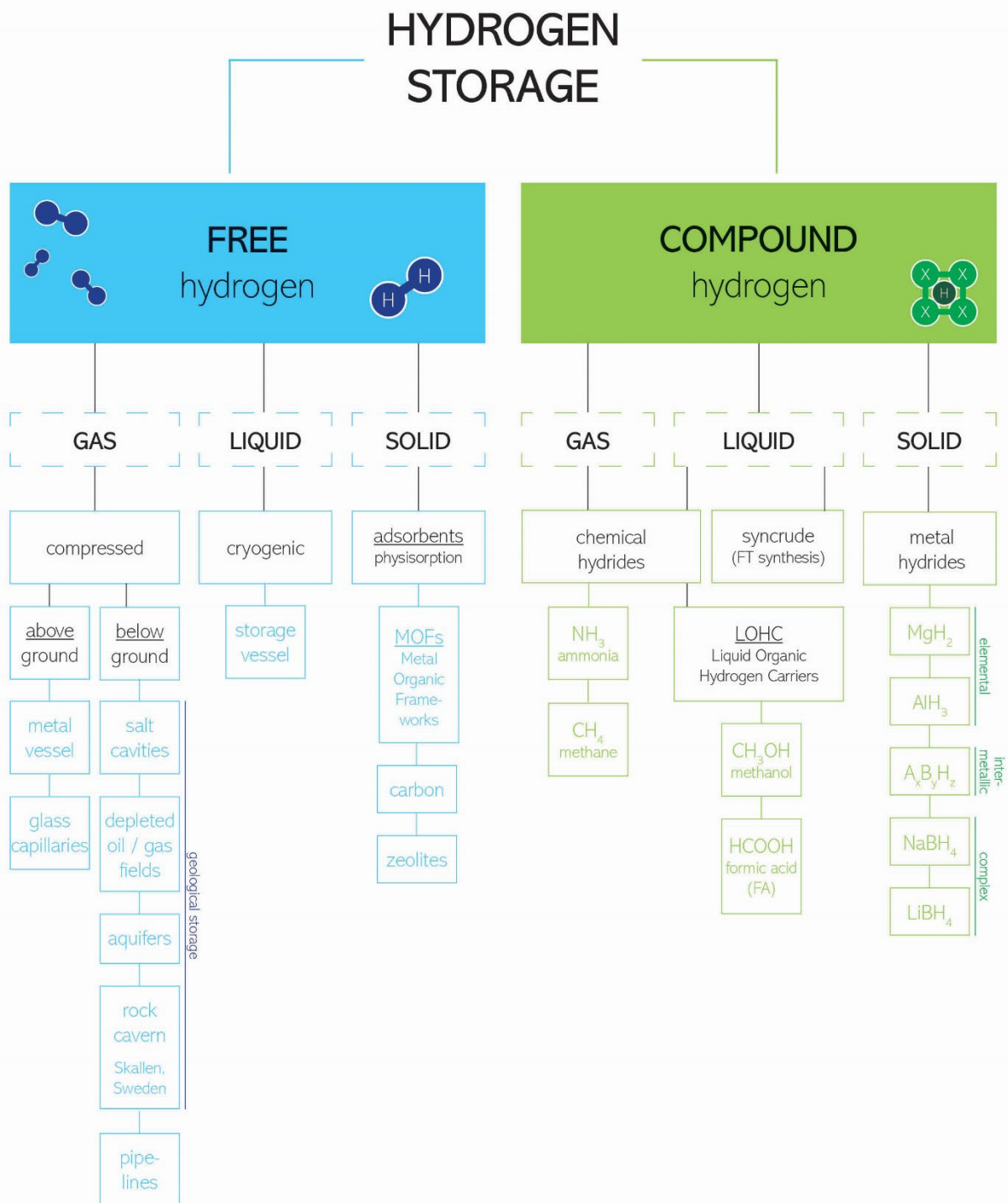


Figure 3: Overview of the main (most discussed) hydrogen storage possibilities, categorized into free and compound hydrogen and sorted by hydrogen phase.

1. Free Hydrogen Storage: gas

Hydrogen storage in its gaseous state requires compression due to its extremely low density of only 0.089 kg/m³ under normal conditions (Hassan et al., 2021) & (Elberry et al., 2021). Its density is thus determined but also limited by the achievable storage pressures, which in their turn depend on the limitations and safety considerations of the operational system (Andersson & Grönkvist, 2019). Currently, storage systems at relatively large scale do not exceed 100-200 bar of pressure. At this pressure, hydrogen's density reaches a value of 7.8 kg/m³. Although compression requires a lot of energy, it is not comparable to the 64% higher energy requirement for liquid storage applications (Elberry et al., 2021). Small scale onboard storage, as used in fuel cell vehicles (FCVs), can reach pressures of 700-1000 bar (Andersson & Grönkvist, 2019) & (Hassan et al., 2021). However, this brings about high energy requirements and operation costs.

Compressed hydrogen gas storage can be divided into 2 categories:

Above ground storage occurs mainly in metallic vessels. It is usually less preferred compared to below ground storage since it brings about much higher costs (Andersson & Grönkvist, 2019). This option is relatively immature for hydrogen storage purposes but has been applied largely for fossil fuel gas storage. However, it seems as though currently used vessels are also compatible for hydrogen. There are different types of metallic vessels, amongst which pipe storage, but there seem to be a few technical obstacles. Another option is the use of glass structures in the form of capillary arrays, at present only implemented at microscale (Preuster et al., 2017). This system could store hydrogen under high pressures up to 1000 bar and possibly reach energy storage capacity targets. However, this technology is relatively immature and needs further investigation before implementation at a larger scale becomes a possibility.

Below ground storage makes use of certain geological characteristics (Andersson & Grönkvist, 2019). The most viable option seems to be compressed hydrogen gas storage in salt cavities as it has low construction costs, low risks and is easy to use (fast withdrawal and injection rates). The only problem when it comes to salt cavities is that these do not exist at every desirable location on Earth since it is dependent on geological conditions. Other options that might be more available around the globe - but are still limited - are depleted oil or gas fields and aquifers.

Another promising option for hydrogen gas storage is pipeline storage (Andersson & Grönkvist, 2019). This has already been used quite some time for the storage and transport of fossil fuels but has proven to be an interesting option for hydrogen as well.

Underground vessels are usually not implemented as they have little advantage over other storage systems (Andersson & Grönkvist, 2019). Nevertheless, there are experiments using a lined rock cavern as underground storage vessel in Skallen, Sweden, which appears to be a good option.

2. Free Hydrogen Storage: liquid

Liquid hydrogen has attracted more attention due to its high hydrogen content, and relatively low risks and costs (Zheng et al., 2021). To liquefy hydrogen, the gas needs to be cooled to approximately $T=20\text{K}$ at normal pressures (1 bar) or higher (Yanxing et al., 2019). The process of cooling down a gas makes that liquid hydrogen is also referred to as cryogenic hydrogen, which relates to the low temperatures at which liquid hydrogen is handled (Encyclopedia Britannica, n.d.). Both terms are used interchangeably.

This liquefaction process can increase the density of hydrogen to 70kg/m^3 without having to apply external pressures (Andersson & Grönkvist, 2019). Around 30-45% of energy in the form of hydrogen is used for this process alone, a drawback of the technology (Qyyum et al., 2021) & (Wijayanta et al., 2019). It also brings about massive installation costs and availability issues (Andersson & Grönkvist, 2019). On top of that, a relatively big part of the hydrogen and energy input is lost through evaporation, also known as boil-off, possibly risking the safety of its application (Preuster et al., 2017). One of the reasons for that is heat transfer, which forms the main threat to the implementation of hydrogen as a liquid. Thus, this requires more research to improve (Wijayanta et al., 2019).

An advantage of liquid hydrogen storage is its high level of purity, not requiring any further steps after hydrogen release (Wijayanta et al., 2019). In addition, the auto-ignition temperature is relatively high ($535\text{ }^{\circ}\text{C}$), reducing spontaneous explosion risks. A drawback, on the other hand, is that the infrastructure should be made suitable for liquid hydrogen.

Liquid hydrogen can be stored in specially designed cryogenic storage vessels, both for terrestrial and marine applications (Preuster et al., 2017) & (Wijayanta et al., 2019). These should be suitable for the operation at extremely low temperatures and be able to withstand pressures (Air Products and Chemicals, 2014). Due to the hazards, the location of liquid hydrogen storage tanks is strictly regulated. Storage usually involves a tank with pressure relief valves, vaporizer, and controls. Purging is required before hydrogen is added to the system or before the system is opened, to prevent explosion hazard.

3. Free Hydrogen Storage: solid-state

Solid-state storage of free hydrogen is based on an entirely different process compared to gas or liquid hydrogen storage. For it is not the hydrogen which is present in solid form, rather in liquid or gas state, but the adsorbent. Through physisorption, liquid or gaseous hydrogen attaches to the surface of adsorbents, which can also be perceived as shifting from a gas or liquid to a solid surface (Hassan et al., 2021). Since this process makes use of weak Van der Waals bonds between hydrogen and the adsorbent, external conditions such as a lowering of the temperature and an increased pressure need to be applied to achieve more energy dense storage (Andersson & Grönkvist, 2019). Besides, this exothermic reaction needs appropriate heat management to reach higher adsorption rates. On top of that, hydrogen storage capacity is also limited by the need for additives to make the process more effective. Lastly, it should be kept in mind that extra space is needed for the adsorbent during storage in a vessel for example, which can also limit the applied pressure.

Frequently discussed adsorbents suitable for storing hydrogen in literature are Metal Organic Frameworks (MOFs) and carbon structures (Andersson & Grönkvist, 2019) & (Hassan et al., 2021). Probably since these are also the most successful options regarding scale and hydrogen storage density (Andersson & Grönkvist, 2019). Almost all other adsorbent-based applications are still only available on laboratory scale. Still, this type of hydrogen storage is investigated quite a lot due to its promising properties, described by Hassan et al. (2021) as: “fast kinetics, good reversibility as well as high hydrogen storage capacity”. But again, low density poses as a threat for this technique. Thus, more research is needed to make adsorbents a viable hydrogen storage option for the future.

Metal Organic Frameworks (MOFs) exist in various forms, showing a crystalline structure (Shet et al., 2021). Due to its high porosity and surface area, it is a promising adsorbent for hydrogen. Inert materials could help improve its hydrogen content as well as cryogenic conditions, but more research is needed on suitable agents to optimize the process (Shet et al., 2021) & (Hassan et al., 2021). MOF-5 has shown to be the most mature among all MOFs, with optimal results at cryogenic temperatures and normal pressure (Hassan et al., 2021). It is positively affected by polarization and decreased pore sizes.

Carbon structures, specifically porous carbon, can occur in multiple forms and their capacity to store hydrogen mainly depends on the surface area and the presence and volume of (micro)pores (Mohan et al., 2019) & (Broom et al., 2016). Activated carbon is the most well-known example of a carbon structure (Hassan et al., 2021). This adsorbent works best under cryogenic temperatures and high pressures. The same counts for carbon nanostructures, such as carbon nanotubes from graphite, but thermal treatments and metal doping have been proposed as improvements and more research should be done for higher storage capacities (Mohan et al., 2019).

Zeolites are another example of a promising hydrogen storage adsorbents, the first inorganic compound proposed, which faces about the same issues and provides the same advantages as MOFs and carbon structures (Hassan et al., 2021) & (Halder & Ghoshal, 2022).

Even though hydrogen can be stored using adsorbents, these complexes in their turn need to be stored in tanks, like the Adsorptive Hydrogen Storage Tank as discussed by Broom et al. (2016) and Mohan et al. (2019). They conclude that more research is required to design the ideal storage tank as there are too many problems with the currently proposed systems, of which low gravimetric and volumetric capacities are the main. On top of that, this hydrogen storage technique (using adsorbents) is amongst one of the least mature options proposed yet (Halder & Ghoshal, 2022).

4. Compound Hydrogen Storage: gas

Other than physical storage of free hydrogen in liquid and gaseous form or solid-state adsorption, there is the option to store hydrogen as a compound. In that case, hydrogen is bound in another chemical, which can again be in gaseous, liquid, or solid form. Gaseous compound hydrogen refers to the gaseous state the compound is in after bonding with hydrogen. The two most well-known examples are ammonia and methane, with chemical formula NH_3 and CH_4 respectively.

Methane (CH_4) is the “carbon-based carrier with the highest storage capacity” for hydrogen (Preuster et al., 2017). It is mainly produced from hydrogen and CO_2 through the Sabatier reaction, a strongly exothermic chemical reaction that requires heat removal. This is also a drawback of this option as pure CO_2 is required as input, which can have scale and cost implications. The same counts for the other way around: it is hard and costly to recover hydrogen from methane in a significantly pure form. The steam-methane reforming (SMR) process produces hydrogen, albeit not free of CO_2 (Chen et al., 2020). A newly proposed option that is more environmentally friendly is the Catalytic Decomposition of Methane (CDM), but this is still not commercially available. The disadvantages of recovering hydrogen from methane make methane more interesting as another energy carrier rather than a hydrogen storage system (Preuster et al., 2017). It is for that reason that methane is barely discussed in scientific articles covering hydrogen storage options, but only in the hydrogen production category.

Ammonia (NH_3) is a valuable option due to its relatively high hydrogen storage density and flexible application in mobile and stationary storage systems and utilization (Aziz et al., 2020). It is usually produced following the Haber-Bosch process, requiring high temperature and pressure and the help of a catalyst for optimal results. Ammonia as energy source can either be used directly or indirectly. In the first case, ammonia is used in a fuel cell or combustion engine, sometimes mixed with other fuels, which can then be converted into electric energy free of CO_2 (Aziz et al., 2020) & (Hassan et al., 2021). The second option is to convert ammonia to hydrogen, which is then used as energy source. This can, however, not be done using only one step and requires additional purification steps (Preuster et al., 2017). Ammonia has the advantage of not producing any CO_2 during its use, making CO_2 -free energy (Aziz et al., 2020). On the other hand, ammonia can produce NO_x during combustion, another greenhouse gas (GHG). Plus, ammonia is characterised as a toxic gas, which can lead to safety and health issues for people and the environment (Preuster et al., 2017). Appropriate management is therefore needed to make ammonia a truly viable option. Liquifying ammonia can lead to higher volumetric energy densities, beneficial for transportation for example through pipelines or for storage purposes (Aziz et al., 2020). The best option so far for stationary storage is a light and low-cost tank under atmospheric pressure and relatively low temperature ($-33,4^\circ\text{C}$). Existing propane infrastructure is also appropriate for ammonia storage.

5. Compound Hydrogen Storage: liquid

Hydrogen can also be stored in compounds that are liquid at ambient temperatures, forming so-called chemical hydrides (Hassan et al., 2021). Although it should be noted that this term is not specific to liquid phase applications.

Liquid Organic Hydrogen Carriers (LOHC) are liquid compounds that can store hydrogen reversibly due to a covalent bond between the two (Hassan et al., 2021). They exist in many forms, for example as cycloalkanes, ammonia-borane based systems, nitrogen-containing aromatics, toluene, and heat transfer fuels to name a few (Hassan et al., 2021) & (Preuster et al., 2017). LOHCs have a great capacity to store hydrogen, they release only little heat during production, are safe and abundant, and can easily be used. Besides, high-quality pure hydrogen can be obtained, and the current fuel infrastructure is compatible (Hassan et al., 2021) & (Preuster et al., 2017). On the other hand, high costs are generated and high temperatures, high pressures and the addition of catalysts required (Hassan et al., 2021). Linked to LOHCs are formic acid and methanol (Hassan et al., 2021).

Formic acid (HCOOH) appears to be a hydrogen carrier with limited functions, probably only applicable in small-scale off-grid energy purposes (Preuster et al., 2017). This is partly due to the corrosive nature of formic acid - making costly infrastructure adjustments a prerequisite - and partly due to the process of its creation. CO₂ hydrogenation, also used to produce methanol, is not very favourable and efficiencies are low (Preuster et al., 2017) & (Onishi et al., 2018). Theoretically, however, formic acid as hydrogen carrier appears to be an interesting option because of its relatively high density, resulting in a considerable volumetric hydrogen storage capacity (Preuster et al., 2017). Hydrogen can be released from formic acid through a catalytic process as described by Onishi et al. (2018) and Preuster et al. (2017). A mixture of high pressure H₂ and CO₂ gases is formed and separating them only requires lowering its temperature (Onishi et al., 2018). To conclude, formic acid formation is not that simple, but its use as hydrogen carrier could be beneficial.

Methanol (CH₃OH) is formed by hydrogenation of CO₂ using catalysts (Preuster et al., 2017). This reaction has promising reaction rates, nevertheless it is limited by (thermodynamic) process parameters, making a full conversion unrealistic. Methanol can serve as a fuel directly, in a combustion engine or more efficiently in a Direct Methanol Fuel Cell (DMFC), or as a hydrogen carrier. In case of the latter, hydrogen needs to be released from the methanol in a relatively uncomplicated manner. This hydrogen carrier provides that advantage through the process of methanol steam reforming or dehydrogenation (producing CO as by-product) (Preuster et al., 2017) & (Kumar et al., 2021). Another advantage is the relative low price of methanol.

Syncrude is a mixture of long chain hydrocarbons, showing similarities to crude oil (Preuster et al., 2017). Production occurs through Fischer-Tropsch (FT) synthesis, which uses CO₂ and hydrogen as input. It is expected that Syncrude will become more like existing fuels in the future, thus be able to use the infrastructure already in place, a huge advantage for its implementation. However, the FT process could use some optimization as it now requires high temperatures (200°C +) and moderate pressures (10-40 bar).

6. Compound Hydrogen Storage: solid

Hydrogen can react with other chemicals to form a solid via the process of absorption (Hassan et al., 2021). In literature, this is also referred to as chemical hydrogen storage, although this term is not specific to solid-state applications (Preuster et al., 2017). Metal hydrides are the main category and can be divided into 3 groups: elemental, intermetallic and complex hydrides (Andersson & Grönkvist, 2019).

Though solid-state storage of hydrogen through adsorption and absorption (in hydrides) might look similar at first glance, there are very important differences. The bonds with hydrogen in absorption are much stronger than the weak Van der Waals bonds in adsorption (Andersson & Grönkvist, 2019). Besides, hydrogen does not remain in its pure form - as is the case in adsorption processes, thus classified as Free Hydrogen Storage - but rather fits into the new structure and forms a completely new compound. An advantage of absorption is its possibility to store hydrogen reversibly, using desorption practices (Hassan et al., 2021).

Metal hydrides provide a safe manner of hydrogen storage in that relatively low temperatures are required for the operation of the process and hydrogen is released through an endothermic reaction (Hassan et al., 2021). Especially lightweight elements are advantageous as weight is a limiting parameter, decreasing hydrogen storage densities. Recently, magnesium compounds have gotten the most attention due to their many advantages. However, the optimal combination of elements is still researched (Preuster et al., 2017). On top of that, hydrogen storage in the form of metal hydrides is hindered by "slow kinetics, low reversibility and high dehydrogenation temperatures" (Hassan et al., 2021). Optimization occurs through surface modification - hydrogen can best attach to nanoparticles with a large surface area - and additives (Preuster et al., 2017). However, working with nanoparticles requires the use of inert materials to prevent degeneration through exposure to air or moisture, which should also be incorporated into the design of an appropriate storage tank. It now looks like metal hydrides can only provide a solution for powering small electronics and remain on a small- and very-small-scale. Large-scale options are hindered by the considerable drawbacks.

Within metal hydrides, 3 groups can be distinguished:

Elemental hydrides are bonds between hydrogen and 1 other metal, a binary compound (Andersson & Grönkvist, 2019). Examples are magnesium hydride (MgH_2) and aluminium hydride (AlH_3), two of the most promising compounds in this category.

Intermetallic hydrides can be seen as optimized elemental hydrides since an extra element (largely rare earth and transition metals) is added, giving it the chemical formula $\text{A}_x\text{B}_y\text{H}_z$ (Andersson & Grönkvist, 2019) & (Hassan et al., 2021). The combination of a weakly and a strongly bonded element make that hydrogen storage capacities improve. However, intermetallic hydrides are still not viable to use for large-scale storage or mobile applications due to low storage densities and high material costs. On the other hand, sectors could apply this technique in cases where its disadvantages – mainly weight – turn out to be useful, such as in forklifts.

Complex hydrides are generally formed through a reaction of two hydrides, forming a covalent bond (Preuster et al., 2017). The product contains boron, nitrogen or aluminium combined with alkali metals such as Li or Na. Examples are: LiBH_4 , NaAlH_4 and LiNH_2 . The highest storage densities are reached when boron functions as the central atom. An advantage of this hydrogen carrier is the relative mild conditions under which hydrogen can be released. Be that as it may, the other way around is a lot harder to achieve and requires significant costs and effort.

HYDROGEN STORAGE TECHNOLOGIES | COMPARISON

An overview of the hydrogen storage technology options is provided in the previous chapter. This now asks for a comparison, resulting in a general idea of best practices in this field. The comparison is based on multiple predefined criteria. This section seeks to answer the following research question:

RQ2. How do hydrogen storage technology options compare with respect to the predefined comparison criteria: technological specifications, efficiency & readiness, safety, and financial projections?

The great variety in proposed technologies creates difficulties for a complete and concise comparison. Still, some properties can function as a base for comparison, resulting in the predefined comparison criteria.

Comparison criteria in this chapter are:

- Technological specifications of experimental setups define how realistic the option is to apply regarding technological adjustments related to temperature, pressure, and additive or catalyst requirement. A distinction is made between hydrogen storage parameters ([table 2](#)) and hydrogen uptake and release (hydrogenation and dehydrogenation respectively) parameters ([table 3](#)).
- Technological efficiency & readiness determine the viability of the investigated hydrogen storage options. Efficiencies need to be able to compete with other sources of energy and thus drastically increase ([Bhuiyan & Ibragimova, 2022](#)). In this chapter, round-trip and conversion efficiencies are differentiated ([table 5](#)). Other efficiencies, related to fuel cells for example, are considered beyond the scope of this research. Readiness is related to the scale of application (large to very small or not realistic), hydrogen content (in wt%) ([table 4](#)), volumetric and gravimetric density ([table 5](#)).
- Safety should be considered, as hydrogen is an inflammable gas and leakage can lead to an explosion, potentially endangering people, animals, and the environment ([Bhuiyan & Ibragimova, 2022](#)). For the purpose of this research, a distinction is made between a compound's toxicity and flammability risks ([table 6](#)), the sole focus of this part. For visualization purposes, the schematic overview of the previous chapter is adjusted to integrate the risks ([figure 5](#)).
- Financial projections largely determine if a technology will be implemented. A future Hydrogen Economy asks amongst others for lowered costs ([Khatriwada et al., 2022](#)). [Table 7](#) lists the estimated costs for storing hydrogen using the different techniques. Costs related to other parts of the process are considered beyond the scope of the research.

1. Technological specifications

A good starting point for assessing the future of investigated hydrogen storage options is provided by the gathered information in the tables below ([table 2 & 3](#)). First, the hydrogen storage parameters – pressure, temperature, and additive requirement – are listed to understand the current state of the hydrogen storage technologies and thus the need to develop more ([table 2](#)). In some cases, usually for solid-state or compound-based storage, it is also relevant to get insight into the processes of hydrogenation and dehydrogenation required before and after storage respectively. For that reason, the same parameters – pressure, temperature, and additive requirement – for hydrogen uptake and release are listed in a separate table ([table 3](#)) to get an overview of the implications of certain processes and the underlying requirements of hydrogen storage.

Hydrogen storage parameters

Hydrogen storage as compound and free hydrogen requires a certain pressure and temperature, which are listed in [table 2](#) for all investigated options. The most optimal situation is one with a pressure close to **1 bar** (atmospheric pressure), a temperature close to **room temperature** (around 20°C) and **no additive or catalyst** requirement, because that requires the least technological adjustments and is thus easiest. However, in practice this is almost never the case, as can be seen in [table 2](#) where all parameters that do meet these desired values are given a light blue colour.

It should be noted that all values are under constant change and circumstances might improve with future research and the addition of catalysts and other substances. Besides, the temperatures and pressures presented in [table 2](#) are considered the optimal situation for hydrogen storage currently, but there might be other options that result in lower weight percentage (wt%) for example (Hassan et al., 2021).

Table 2: Operational parameters - pressure, temperature, and additives requirement - needed for hydrogen storage technology options. If there is not enough information available, a star (★) is added.

Cat.	Carrier	Required Pressure	Required Temperature	Catalysts & Additives
FHG	Compressed hydrogen gas	350-700 bar ²²	normal ²²	✗
	Glass structures	over 1000 bar ²⁶	-40 - 60°C ²⁷	✗
FHL	Liquid hydrogen	1 bar ³⁰	-253°C ^{20,4}	✗
FHS	MOFs (MOF-5)	1 bar ¹³ for adsorption	cryogenic ¹³	✓ ¹³ metal ions: Li ⁺ Cu ⁺² Mg ⁺²
	Carbon structures	high ¹³	cryogenic ¹³	✓ ¹³
	- Activated carbon	80 bar ¹⁹	25°C ¹⁹	
	- Graphite	100 bar ¹⁹	25°C ¹⁹	
	- Carbon nanofibre (CNF)	120 bar ¹⁹	27°C ¹⁹	
	Zeolites	★ 10 – 100 bar ² general for sorbents	room T ^{13,23} ~ -196°C ² general for sorbents	✓ ¹³ Ni ²⁺ Co ²⁺
	GENERAL ADSORPTION	40 bar ⁴	-176°C ⁴	depending on sorbent type
CHG	Ammonia (liquid)	normal ^{5,8}	-33,4°C ^{5,8}	✓ ¹³
		9,9 bar ⁵	25°C ⁵	
		8 bar ⁸	ambient ⁸	
	Methane	⊖	⊖	⊖
CHL	Syncrude	Similar to existing fuels ²⁶	Similar to existing fuels ²⁶	✓ ²⁶ Co-based
	Methanol (LOHC)	50 bar ⁴	250°C ⁵	✓ ²⁶
		1 bar ^{5,8}	25°C ^{5,8}	
	Formic Acid (LOHC)	105 bar ⁴	100-180°C ⁴	✓ ²⁶
CHS	Metal hydrides (general)	moderate ¹³ 10-40 bar ¹¹	low ¹³ 20°C ¹¹	✓ ²⁶
	- Elemental	★ ⁴ AlH ₃	<70°C ⁴ AlH ₃	✓ ⁴
		30 bar ⁴ MgH ₂	300°C ⁴ MgH ₂	
	- Intermetallic	50 bar ⁴	<80°C ⁴	★
	- Complex	100 bar ⁶ Values based on Fuel Cell Coupled Solid State Hydrogen Storage Tank	200°C ⁶	✓ ⁴

Abbreviations

FHG: Free H₂ gas

FHL: Free H₂ liquid

FHS: Free H₂ solid (*adsorption*)

CHG: Compound H₂ gas

CHL: Compound H₂ liquid

CHS: Compound H₂ solid (*absorption*)

Symbols

✓ = preferable

✕ = not needed

⊛ = unknown, no info available

⊙ = not applicable

Reflection of storage parameters

Table 2 shows the wide range of pressures and temperatures required for hydrogen storage for the investigated options in this thesis. It can be concluded from the light blue marked cells that not one option meets the optimal conditions mentioned before (1 bar, room temperature and no additives/catalysts). However, there are some options that meet 2 out of 3 requirements: compressed hydrogen gas (normal temperature, no additives) and methanol (normal pressure and temperature). That doesn't mean that these are also the best options though. This is generally dependent on factors like the applied technique.

Remarkable is the case of methane as hydrogen carrier. Its gaseous state makes it relatively hard to be stored and transported. Options to change this are liquefaction by converting methane to its liquid alcohol (methanol), or storing it, just like gaseous hydrogen, in MOFs for example (Department of Energy, Office of Science, 2015). These extra steps sound a bit cumbersome. Besides, articles on methane as a hydrogen carrier are very scarce, which implies that it is not a realistic option to use methane to store hydrogen. Chen et al. (2020) mention methane as a source of hydrogen, using it for hydrogen production rather than storage. They also imply that hydrogen storage is only interesting in methane if it is first converted to another compound. Therefore, in table 2 a ⊙ sign is added for methane.

Hydrogen uptake and release parameters

Hydrogen storage is not always the same as hydrogen uptake and release, especially in the case of compound hydrogen storage and free hydrogen storage in solid-state. In that case, it is useful to consider the pressure and temperature requirements for hydrogenation and dehydrogenation processes. Table 3 gives an overview of these parameters. It should be noted that the options listed are under constant change. Besides, catalysts can improve the circumstances listed and are most of the time not accounted for in the values for temperature and pressure. Optimal conditions are highlighted in blue.

Table 3: Operational parameters - pressure, temperature, and additives requirement - needed for hydrogen uptake and release.

Cat.	Carrier	H ₂ uptake (hydrogenation)		H ₂ release (dehydrogenation)		Add.
		Pressure	Temperature	Pressure	Temperature	
FHG	Compressed hydrogen	⊖	⊖	⊖	⊖	⊖
	Glass structures	⊖	⊖	⊖	⊖	⊖
FHL	Liquid hydrogen	⊖	⊖	evaporation ¹⁸		⊖
FHS	MOFs	1 bar ¹³	cryogenic ¹³	★	★	★
	Carbon structures	★	★	★		✓ ²⁸
	- Activated carbon	3 MPa ²⁸ 30 bar	77K ²⁸ ~-196°C			
	- Graphite	11,35 MPa ²⁸ 113,5 bar	room T ²⁸ ~20°C			
	- Carbon nanofibre (CNF)	10,10 MPa ²⁸ 101 bar	room T ²⁸ ~20°C			
	Zeolites	900 bar ¹³ ~100 bar ²³ Co ²⁺ /Ni ²⁺ zeolites	350°C ¹³	1,5 bar ²³ Co ²⁺ /Ni ²⁺ zeolites	★	✓ ¹³
CHG	Ammonia	100-300 bar ²⁶	400-500°C ⁵	250 bar ²	400-700°C ^{5,2}	✓ ^{26,5} (iron) ³¹
		Haber-Bosch reaction (unsustainable) ³¹		endothermic		
	Methane	<100 bar ¹⁰	250-400°C ²⁶	★	★	✓ ²⁶
		Sabatier reaction / methanation		hard & costly: generally not considered ²⁶		
CHL	Syncrude	10-40 bar ²⁶	200-250°C ²⁶ (LTFT)	★	★	★
		Fischer-Tropsch reaction		---		
	Methanol (LOHC)	~40-60 bar ³ + homogeneous catalysts	~140°C ³ + homogeneous catalysts	25-50 bar ³¹	200-300°C ³¹ lower with cat.	✓ ⁵
		exothermic, limited by thermodynamic constraints		endothermic, methanol steam reforming ²⁶		
	Formic Acid (LOHC)	120 bar ¹⁴	120°C ¹⁴	dependent on CO ₂ recovery	<100°C ⁴ > 50°C ¹⁸	✓ ¹⁴ Ru- based
		Also: electrochemical		---		
CHS	Metal hydrides	dependent on type / particle size ²⁶		1.5 bar ⁶ complex hydr.	≥300°C ^{13,4} for MgH ₂ 100°C ⁴ for AlH ₃ <180°C ⁶ complex hydr.	✓ ²⁶ + surf. modif. /doping
				endothermic		

Symbols

✓ = preferable

✕ = not needed

⊕ = unknown, no info available

⊖ = not applicable

Reflection of (de-)hydrogenation parameters

The values in [table 3](#) show great variety and only a few cells display the optimal conditions (highlighted in blue). However, regarding hydrogen storage options, the parameters shown in [table 2](#) are far more important. Comparing parameters in [table 3](#) and the different processes they are based on is also not very realistic, but still there are a few (general) conclusions to be drawn from this. For example, additives are almost always required to optimize processes and it is expected that the same counts for the options with a ⊕.

Hydrogen storage in the form of compressed hydrogen gas or liquid hydrogen can be perceived as the most beneficial in the sense that no hydrogenation or dehydrogenation processes are required. However, these options do require specific circumstances for storage that might be less the case for other options.

Remarkable is the large number of empty spaces in [table 3](#). These can mostly be attributed to categories comprising of a group of hydrogen carriers rather than solely one, as is (in part) the case for MOFs, carbon structures, zeolites, and metal hydrides. A reason for the lack of data here could be that these techniques are relatively new and research on these topics is still rather immature. Furthermore, as stated before, these options exist of a group of carriers, which makes assigning a value even more complicated.

Methane is a different case. The few papers about methane-based hydrogen carriers usually conclude that a hard and costly process is needed to obtain hydrogen from methane, making it a generally not considered hydrogen carrier. That is possibly also the reason why the dehydrogenation cells for methane have a ⊕.

Syncrude is also a case on its own, because in literature it is almost only perceived as a final product rather than a hydrogen carrier. Therefore, dehydrogenation processes are almost never discussed. An assumption is that syncrude will be used directly in the same way fossil fuels are implemented nowadays and thus does not need such a process. Besides, syncrude is not very often related to hydrogen apart from the Fischer-Tropsch reaction for its formation.

2. Technology efficiency & readiness

Implementing technologies requires assessing their readiness level and ease of integration in the current society. A technology is ready if it is in the Deployment Phase (TRL 9) (figure 4), according to the system invented by NASA (SNN, n.d.) & (Tzinis, 2021). For it to be there, a technology needs to be technically and commercially ready, the only step left is to market it. It is therefore hard to make a good comparison based on data for technologies that are still developing. On the other hand, it is very useful to understand what is possible in a certain situation and what can be improved on.

This technology readiness assessment is focussed on gathering information related to the scale of application and the hydrogen content of the different carriers in the first part (table 4). The second part focusses on volumetric and gravimetric densities, and round-trip and conversion efficiencies (table 5).

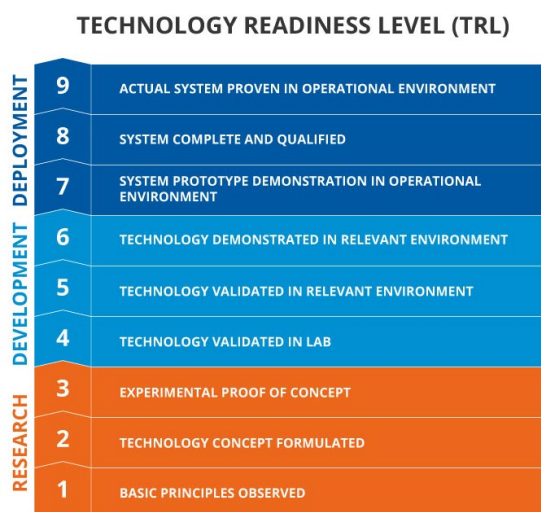


Figure 4: Technology Readiness Levels (TRL) explained. A technology is ready if it reaches Deployment Phase 1 (TWI, n.d.).

Hydrogen technology readiness parameters

These first technology readiness parameters relate to scale and capacity of the storage system. In this case, scale of application refers to the scale at which a hydrogen storage option is currently employed, or the scale it is expected to function on in the future. An explanation is given in blue. Hydrogen content shows what part of the storage system is hydrogen in wt%, with an optimal - though unrealistic - value of 100 wt%. A summary is given in table 4 below, red cells are lowest values, green cells are highest values.

Table 4: Technology readiness parameters: the scale of application, hydrogen storage capacity and total hydrogen content.

Cat.	Carrier	Scale of application	Hydrogen content (wt%)
FHG	Compressed hydrogen gas	① ²⁶	max. 4,2 wt% ¹³ 13 wt% ¹⁹
	Glass structures	④ ²⁶ microscale	10 – 14 wt% ²⁰
FHL	Liquid hydrogen	② - ① ²⁰	Size dependent
FHS	MOFs	③ ²⁰ currently at laboratory scale	4,5 wt% ¹³ max. at cryogenic T
	Carbon structures	③ ²⁰ currently at laboratory scale	8 wt% ²⁰ dependent on type:
	- Activated carbon		5,5 wt% ¹⁹
	- Graphite		4,48 wt% ¹⁹
	- Carbon nanofibre (CNF)		6,5 wt% ¹⁹
	Zeolites	③ ²⁰ currently at laboratory scale	9,2 wt% ²⁰ max 2,9 wt% ²³ without additives
CHG	Ammonia	① ^{5,26} promising for transport	17,8 %wt ¹³
	Methane	⑤ ²⁶ not suitable for hydrogen storage	25 %wt ²⁶
CHL	Syncrude	① ²⁶	★

		in today's storage and transport infrastructure	
	Methanol (LOHC)	③ ²⁶ commercial use in DMFCs	12,5 %wt ²⁶
	Formic Acid (LOHC)	③ ²⁶ small, off-grid appl.	4.4 %wt ²⁰
	GENERAL LOHC	① - ③ ²⁶ large- and small scale application of LOHCs	
CHS	Metal hydrides	④ - ③ ²⁶ application scale	max. 12.6 %wt ²⁰
	- Complex (LiBH ₄)		18,5 %wt ¹⁵
	- MgH ₂		7,66 wt% ¹⁵

Explanation symbols (table 4):

- ① = Large scale (energy grids / existing infrastructure) ^a
- ② = Medium scale
- ③ = Small scale (lab?)
- ④ = Very small scale (microscale?)
- ⑤ = Not realistic

Reflection of readiness parameters

From the scales of application mentioned in the table above (table 4), it can be concluded that methane is not suitable for hydrogen storage purposes, scholars usually define it as not realistic. Formic acid will probably also not be applied on a large scale and is expected to only be available at small-scale, off-grid applications. More or less the same counts for metal hydrides, which are expected to be most promising for small consumer electronics (Preuster et al., 2017). All other options could have a future in hydrogen storage. Especially compressed and liquid hydrogen, ammonia and syncrude for they are expected to be (or already are) available at a large scale. Techniques still in development towards a possible larger scale application are sorption-based options in the category FHS, which are drastically in need of improvement before being commercialized (Moradi & Groth, 2019).

On the contrary, hydrogen content - also referred to as gravimetric hydrogen storage capacity - in weight percentages is readily available in literature. Only in the case of syncrude, a complex mixture of hydrocarbons, and liquid hydrogen, reason, there is no relevant data available. All other values are compared and categorized into highest (blue cells) and lowest (green cells) values in table 4. Most promising in this regard appears to be the option to store hydrogen as a gaseous compound (CHG), like ammonia and methane. Complex metal hydride hydrogen storage also scores relatively high concerning its hydrogen content (wt%). Lowest values are for formic acid, carbon structures and MOFs. For the latter two options, this is probably because research on this topic is relatively immature.

Note that capacity (storage in tons per day) is not part of the technology readiness parameters, because this is generally not a limiting factor. At least, not in mature technologies.

Hydrogen carrier densities & efficiencies

The second part of this technology readiness parameter comparison comprises of volumetric and gravimetric densities, and round-trip and conversion efficiencies.

Volumetric energy density is the amount of energy stored per volume, with unit Wh/L, or in the case of hydrogen kWh/L, or MJ/L (Wiki Battery, 2022). Gravimetric energy density is the amount of energy stored per weight, with unit Wh/kg or in the case of hydrogen kWh/L, or in MJ/kg. Recalculation of all values from MJ/L or MJ/kg to kWh/L or kWh/kg respectively, requires values to be divided by 3,6 (1 kWh = 3,6 MJ). Those new values are given in green in [table 5](#). Remarks are in blue. Red cells are lowest values, green cells are highest values.

Round-trip efficiency refers to the “percentage of electricity put into storage that is later retrieved” (Mey, 2021). In the case of hydrogen storage this more or less comes down to the storage efficiency, a parameter more often mentioned in literature. Conversion efficiency refers to the process efficiencies of hydrogenation and dehydrogenation reactions.

Table 5: Technology readiness parameters: volumetric and gravimetric density, and round-trip and conversion efficiency.

Cat.	Carrier	Volumetric Density	Gravimetric Density	Efficiency	
				Round-trip	Conversion
FHG	Compressed hydrogen gas	1,32 kWh/L ¹³ at 700 bar	33,3 kWh/kg ²⁶	97,1% ⁹ pipe storage 98% ⁹ salt caverns 94,9% ⁹ Vessel	72,4% ⁸ from water electrolysis
	Glass structures	★	★	★	★
FHL	Liquid hydrogen	2,36 kWh/L ²⁶	33,3 kWh/kg ²⁶	30-40% ²⁶ thermodyn. 53% ⁸ storage efficiency	44,7% ⁸ liquefaction
FHS	MOFs (MOF-5)	0,021 kg H ₂ / L system ¹³ 31.1 g H ₂ / L	0,038 kg H ₂ / kg system ¹³	★	★
	Carbon structures	★	★	★	★
	- Activated carbon - Graphite - Carbon nanofibers (CNF)	★	★	★	★
	Zeolites	★	★	★	★
CHG	Ammonia (liquid)	12.7 MJ/L ⁵ 3,53 kWh/L	18.6 MJ/kg ⁵ 5,17 kWh/kg	99.3% ⁸ storage efficiency	25% ⁵ conversion rate 300 bar 76% ⁸ Haber-Bosch 69% ² NH ₃ to H ₂
	Methane	0.0378 MJ/L ¹² ambient pressure 0,0105 kWh/L	55,6 MJ/kg ¹² ambient pressure 15,4 kWh/kg	95% ⁸ storage (gas, 60bar)	87.9% ⁸ methanation
CHL	Syncrude	10000 kWh/m ³ ¹⁶ value for crude oil 10 kWh/L	41,9 MJ/kg ²⁹ value for crude oil 3,53 kWh/kg	★	46-67% ²⁵ power-to-fuel

	Methanol (LOHC)	3,3 kWh/L ²⁶ 15,8 MJ/L ⁸	31,1 MJ/kg ²⁹ 8,64 kWh/kg	95% ⁸	83.5% ⁸
	Formic Acid (LOHC)	1,79 kWh/L ²⁶	⚡	⚡	48% ¹⁴ + Ru-catalyst
	GENERAL LOHC			~71% ¹ Storage efficiency	
CHS	Metal hydrides (general)		50 kWh/100kg ¹¹ 0,50 kWh/kg based on GKN Powder Metallurgy's Hy2Green storage tanks	~78% ¹ Storage efficiency	⚡
	(examples)	0,71 kWh/L ¹³ for complex hyd. 5.55 kg/100L ¹⁵ for MgH ₂ 7.42 kg/100L ¹⁵ for AlH ₃ 5.72 kg/100L ¹⁵ for NaBH ₄ 6.11 kg/100L ¹⁵ for NaBH ₄			

Reflection of density & efficiency parameters

Table 5 shows a variety of volumetric and gravimetric densities, sometimes multiple values per storage option. Highest and lowest values are highlighted in green and red respectively. Comparing the (recalculated) green values results in the following conclusions:

- Pure hydrogen in liquid or gaseous form has the highest gravimetric density (33,3 kWh/kg), followed by about half of that value for methane (15,4 kWh/kg). Other options (as far as available) appear to be far behind.
- The highest volumetric density can be appointed to syncrude (10 kWh/L), although this number is based on the assumption that syncrude is comparable to crude oil and brings a lot of uncertainty. Second best is ammonia (3,53 kWh/L), closely followed by methanol (3,3 kWh/L). The differences in this category are smaller compared to those of gravimetric density.
- Literature sometimes provides volumetric and gravimetric densities in different units (kg/100L) than used in table 5, which makes recalculation and appropriate comparisons difficult. Besides, some articles use wt% as gravimetric density, a unit originally belonging to gravimetric capacity (Broom et al., 2019).
- Storage efficiencies are highest for gaseous hydrogen, ammonia, methane, and methanol (95-98%, 99,3%, 95% and 95% respectively). Exceptionally low storage efficiencies - and large uncertainty - are found for liquid hydrogen (30-50%).
- Conversion efficiencies vary greatly amongst all options, with the highest percentages for methane (87,9%) and methanol (83,5%), and the lowest for liquefaction (44,7%).
- The highlighted cells in table 5 show that **methanol** has the most beneficial properties regarding volumetric density and both round-trip and conversion efficiencies. This option scores highest for 3 out of 4 properties (most cells in blue) and does not contribute to the lowest values (no green cells).
- A lot of values are still missing in this table (indicated with a ⚡), which indicates that some hydrogen storage options are relatively immature, and more research is needed in the future to find the most beneficial situation regarding efficiencies and densities.

3. Safety

Safety is evaluated in this case by the explosive or toxic nature of the compounds and their environmental or health hazards. A schematic overview is provided in figure 4 below, a table with the data and a reflection follow table 4.

Schematic overview of hazards associated with hydrogen carriers:

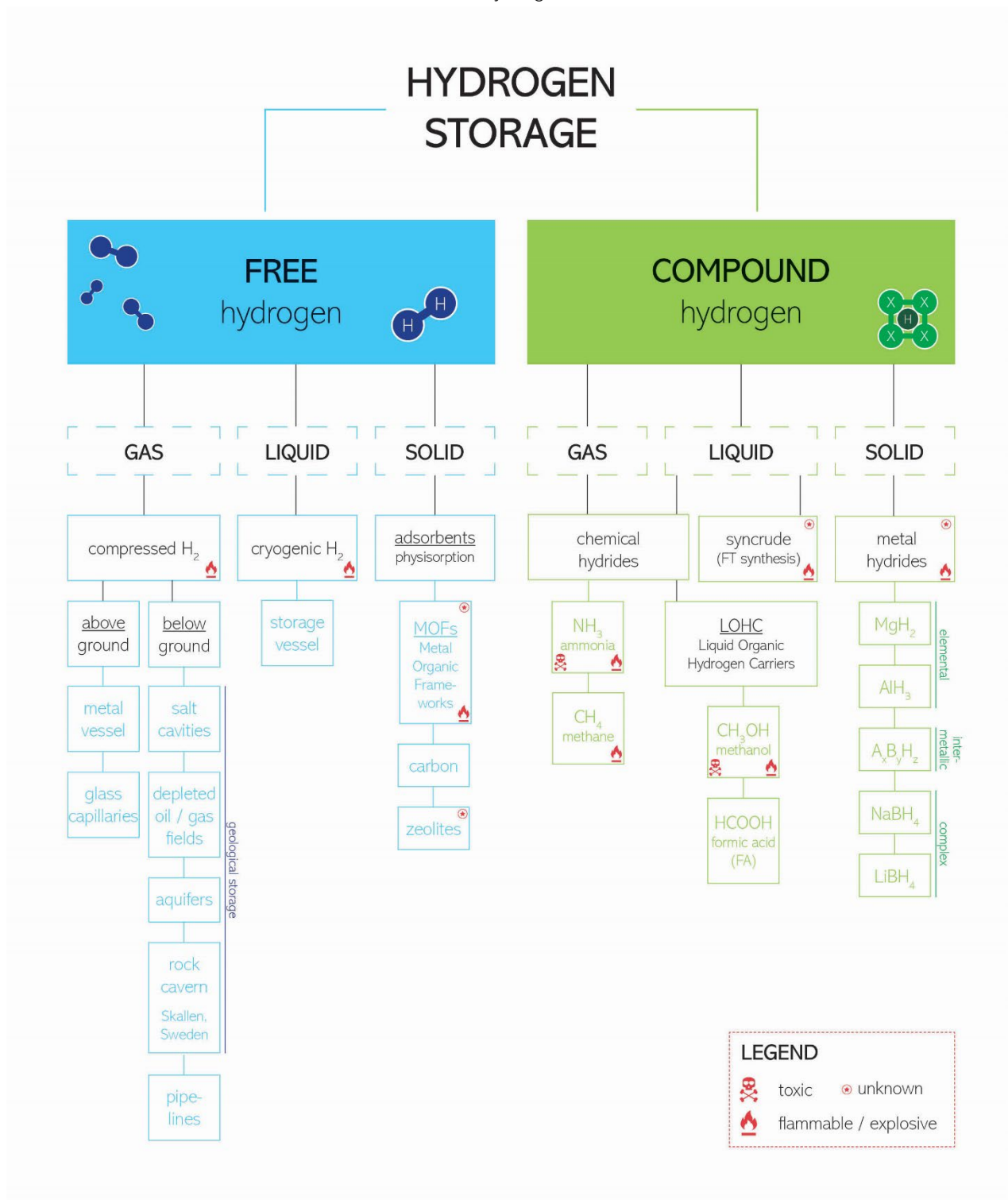


Figure 5: Overview of possible hydrogen carriers and their hazards. Regarding safety, only toxicity and flammability are reported. If unknown, a little star is added.

Table 6: Hazard classification of investigated hydrogen carriers. A check (✓) is used if the hazard fits the compound and a cross (✗) is used for the opposite. If too little information was available, a star is added.

Cat.	Carrier	Flammable / explosive	Toxic	Remarks
FHG	Compressed hydrogen gas	✓✓	✗	Asphyxiant
FHL	Liquid hydrogen	✓✓	✗	Frostbite
FHS	MOFs	✱	✓	Many forms
	Carbon structures	✗	✗	Weakly expl. Health hazard
	Zeolites	✱	✗ / ✱	Little info
CHG	Ammonia	✓	✓✓	Corrosive, env. hazards
	Methane	✓✓	✗	
	Syncrude	✓ / ✱	✗ / ✱	Based on crude oil Health + env. hazard
CHL	Methanol (LOHC)	✓✓	✓	Health hazard
	Formic Acid (LOHC)	✗	✗	Corrosive
CHS	Metal hydrides	✓✓	✱	Taken as a group

Symbols

✓ = this is a characteristic of this compound (more = more extremely)

✗ = this is not a characteristic of this compound

✱ = unknown, no info available

Reflection of safety assessment

From table 6 it can be concluded that flammability is especially an issue for compressed hydrogen gas, liquid hydrogen, methane, methanol, and metal hydrides. Followed by a lower level of explosiveness for ammonia and possibly syncrude. Toxicity is mainly a problem for ammonia, but also - albeit less extreme - for MOFs and methanol. The most promising options in this regard thus seem to be the use of carbon structures or formic acid for hydrogen storage.

Extra information on safety for each hydrogen carrier is provided below:

Hydrogen is classified as an “Extremely flammable gas” (National Center for Biotechnology Information, 2022). It reacts violently with certain chemicals or when heated. It is not regarded as toxic, but rather as an asphyxiant, causing a reduction or replacement of oxygen, possibly risking health due to lack of oxygen (Chemscape Safety Technologies., n.d.). If hydrogen becomes liquid, additional hazards are mentioned, mainly related to the extremely low temperature, such as: “contact with liquid hydrogen will cause frostbite or severe burns of the skin” (National Center for Biotechnology Information, 2022) & (Air Products and Chemicals, 2014). Liquid hydrogen vapor is also highly flammable.

MOFs are a bit harder to classify since there are so many different structures. The synthesis of MOFs is associated with fire and explosion hazards as well as with toxicity (Escobar-Hernandez et al., 2021). However, this is not always related to the compound itself but also to the added chemicals (such as solvents) for example. Regarding the toxicity of MOFs themselves, it can again be stated that this is dependent on various conditions (Niklas, 2019). They as well as their toxicity can be tuned according to their application. For MOF-5, the most promising MOF in hydrogen storage, its toxicity is considered high. Flammability is not mentioned.

Carbon structures are generally nontoxic when referred to activated carbon, but they can cause irritation (General Carbon Corporation, 2018) & (Silberman et al., 2022). When powdered, it is weakly explosive under the right circumstances, but this is not considered a hazard. Carbon nanostructures made of graphite are classified as irritants and are suspected to cause cancer (National Center for Biotechnology Information, 2022). No toxicity or flammability claims are made.

Zeolites, a variety of aluminosilicates, are still under investigation regarding their safety and not a lot of information is available yet. However, it has been found that micron-sized zeolites are considered non-toxic (Lehman & Larsen, 2014).

Ammonia is classified as a “Flammable gas” that can cause “severe skin burns and eye damage” (corrosive) (National Center for Biotechnology Information, 2022). It is both toxic to humans, when inhaled, and even more extremely for aquatic life (environmental hazard).

Methane is classified as an “Extremely flammable gas” (National Center for Biotechnology Information, 2022). As it is a compressed gas usually, it can explode if heated. No specific toxicity hazards are mentioned.

Syncrude is hard to classify as it comprises of a mixture of long chain hydrocarbons, with similarities to crude oil (Preuster et al., 2017). No information on hazards assigned to syncrude is available. Crude oil, however, is classified as a “flammable liquid and vapor” with both health and environmental hazards (Whiting, 2013). Although, its complexity make classification difficult still, and it is not exactly the same as syncrude.

Methanol is classified as a “Highly Flammable liquid and vapor” (National Center for Biotechnology Information, 2022). It is toxic if swallowed, inhaled or if in contact with skin. Another health hazard is that it can cause damage to organs.

Formic acid is classified as a corrosive compound both for metals and tissue, causing “severe skin burns and eye damage” (National Center for Biotechnology Information, 2022). No explosiveness or toxicity is reported.

Metal hydrides are again complex to properly classify as there are many different forms in which they occur. Magnesium hydride (MgH_2), for example, is extremely reactive with water and air (Rivard et al., 2019). Generally speaking, metal hydrides are considered “highly flammable”, corrosive, reacting violently, with health hazards (CAMEO Chemicals, n.d.). There is no specific information regarding their toxicity.

4. Financial projections

Hydrogen storage technologies only really have a future in society if the costs connected to the operation of the technology itself, and related investment costs are as low as possible. Therefore, an estimated calculation of costs per technology option are provided in this chapter. These mainly relate to levelized costs of hydrogen storage (LCHS) solely. Costs related to the maintenance of storage systems, the transport of hydrogen or the purchase of materials are considered beyond the scope of this research project. Production costs are incorporated, though very limited, as it is considered an important aspect but also not the scope of this thesis. The goal is to find out what it would cost to store 1 ton of hydrogen, a realistic future objective. Table 7 gives an overview of the calculations of the costs for hydrogen storage in different scenarios. Recalculating all values from \$/kg to \$/ton results in a value a factor 1000 greater and from €/kg to \$/ton results in a value 980 greater (1 euro = 0,98 dollar). These recalculated values are shown in green below the retrieved values in table 7. In some cases, more values are listed. This is because different sources mention different prices, but it is unclear which is more accurate.

Table 7: Financial projections of hydrogen storage options.

Cat.	Carrier	Costs	Remarks
FHG	Compressed hydrogen gas	~\$1.10/kg of hydrogen ²¹ ~\$2.40/kg of hydrogen ²¹ ~\$1.54/kg ²¹ ~\$0.27/kg ²¹ ~\$1.81/kg €6.6/kg ¹⁷ €7.9/kg H ₂ ²⁶ €0.06/kg H ₂ ²⁶ \$1810/ton	High-pressure tubes (500 bar) Central production + pipelines Compression Storage Compression + storage Production costs (wind energy) Construction cost Operation cost
	- salt caverns	\$1.61/kg ²⁶ \$1610/ton	Levelized costs
	- above ground	~\$0.14/kg of H ₂ ¹ \$140/ton ~\$0.33/kg of H ₂ ¹ \$330/ton	LCHS LCHS
	Glass structures	★	
FHL	Liquid hydrogen	€1.72/(kgLH ₂) ²⁶ \$1686/ton 1.905M\$/tonne ² \$1905/ton	Liquefaction costs Liquefaction + storage tank
FHS	MOFs (MOF-5)	\$490 / kg H ₂ ¹³ \$490,000/ton	
	Carbon structures	★	
	- Activated carbon		
	- Graphite	★	
CH G	- CNF		
	Zeolites	★	
CH G	Ammonia	~ \$3.51/kg of H ₂ ¹ \$3510/ton	LCHS Air Separation Unit ⁸
	Methane	★	
CHL	Syncrude	1.85 €/l _{DE} ²⁵ 1.554 €/kg \$1523/ton	1 DE = 35.9MJ ²⁵ 1 DE = 0.84kg ⁷
	Methanol (LOHC)	~\$2.25/kg of H ₂ ¹ \$2250/ton	LCHS

	Formic Acid (LOHC)	23.16 \$/kgH ₂ ¹⁴ \$23,160/ton	Levelized costs (ecFA + LCO ₂), described as Scenario 3 in article by Kim et al. (2022), containing production and transportation
	GENERAL LOHC (dibenzyl toluene)	~\$1.20/kg of H ₂ ¹ \$1200/ton	LCHS
CHS	Metal hydrides	~\$0.70/kg of H ₂ ¹ \$700/ton	LCHS

Hydrogen production costs come down to ~\$3.2 to ~\$7.7 per kg of H₂ (Abdin et al., 2022). Considering 1 ton of H₂ is equal to 1000kg, total production costs are **between \$3200 and \$7700**. This should be added to the storage costs to get a more complete picture of the situation.

On top of the costs listed in table 7, electrolyser costs, used in the production of Green Hydrogen, should also be accounted for. There are different electrolyzers that can be used, and their costs vary: "Alkaline between 800 and 1,500 €/kW, PEM between 1,400 and 2,100 €/kW and SOEC above 2,000 €/kW" (Dias et al., 2020).

Storage of ammonia can be reached by an Air Separation Unit, with a total estimated cost of 340 €/kW (Dias et al., 2020). It can produce 3 tons of 99.9% pure ammonia per day, enough to fulfill the requirements in this chapter. However, this process and its costs are relatively sensitive to changes in the size of production.

Reflection of calculated costs

Appropriate comparison of costs is rather difficult based solely on the values listed in the table above (table 7) due to the uncertainties of what they encompass exactly. Generally, though, costs appear to be between 1000 and 2000 \$ per ton of hydrogen that is stored. Highest and lowest values are highlighted in red and green respectively. Outliers are the cases of formic acid (\$23,160/ton) and MOFs (\$490,000/ton), which are significantly higher, and salt caverns on the other end, which could potentially be the least costly (\$140/ton). But again, in this last situation a lot of uncertainty arises.

A reason for the exceptionally high costs could be the inclusion of safety costs and other types of costs not accounted for in the calculation of storage costs for other compounds. The low costs can be attributed to the use of an already existing storage system (salt caverns) that only need little adjustment (Andersson & Grönkvist, 2019).

Parameters combined

A relevant conclusion can only be drawn if the parameters categorized into technological specifications, technology efficiency & readiness, safety, and financial projections, are combined and measured against the targets set by the US DOE. To do this more easily, an excel file containing all gathered parameters is provided as appendix to this thesis.

HYDROGEN STORAGE TECHNOLOGIES | TARGETS

A comparison of parameters relating to certain aspect of hydrogen storage technologies is most interesting when measured against targets. While most literature discusses the targets regarding green hydrogen production, it is the US Department of Energy (DOE) that has set and communicated targets in the field of hydrogen storage. The main goal is stated by Bakuru et al. (2019): "*The US Department of Energy (DOE) targets for on-board hydrogen storage systems by 2020 include gravimetric and volumetric hydrogen densities of 5.5 wt % and 40 g L⁻¹ under conditions of -40 °C to 60 °C and <100 bar.*"

Other relevant parameters to use for the comparison are described by Schneemann et al. (2018), together with the above stated and filtered on their relevance to this study, [table 8](#) is obtained.

Table 8: Target values set by the US Department of Energy (DOE) for hydrogen storage, filtered by their relevance for this study.

Parameter	Target value
Gravimetric capacity	2,5 kWh/kg
	6,5 wt%
Volumetric capacity	1,7 kWh/L
	0,05 kg H ₂ /L
Storage costs	\$266/kg H ₂ (= \$266000/ton)
Temperature	-40 °C to 60 °C
Pressure	<100 bar

CONCLUSION

This thesis addressed the currently available hydrogen storage technology options. These options were categorized based on the phase of storage (gas / liquid / solid) and the type of bonds (free / compound H_2), following the research question (RQ1): *"What are the hydrogen storage technology options for free and compound hydrogen in gas, liquid, and solid phase?"*. As a second step, the options were compared based on a wide variety of characteristics, following the research question (RQ2): *"How do hydrogen storage technology options compare with respect to the predefined comparison criteria: technological specifications, efficiency & readiness, safety, and financial projections?"*. The main aim with this literature-based study is to find an answer to the main research question: *"What are currently the most promising and realistic options for hydrogen storage, functioning as a grid balancing service?"*. To answer this question, a table incorporating all parameters gathered throughout this thesis is composed and added as an [appendix](#). Based on this, a top 3 of best options can be created.

Defining a top 3 based on comparison criteria

To get to the top 3 of best options for hydrogen storage technologies, the comparison criteria are combined, and the following conclusions can be drawn:

- **Storage** parameters comprise of required pressure, temperature and additives. In this category, methanol and compressed hydrogen score the best, and methane is considered not an option.
- **Hydrogenation and dehydrogenation** parameters refer to the temperature, pressure and additives required for the processes of hydrogen uptake and release. In this regard, compressed and liquid hydrogen appear to be most beneficial as these processes are not required in those cases. But that does not necessarily mean that those options are the best in this category. Another comparison is hard to make as the processes and thus circumstances vary greatly. Again, methane is considered not an option. As is dehydrogenation of syncrude. Lack of data makes it hard to compare MOFs, carbon structures, zeolites and metal hydrides.
- **Technology readiness** parameters involve scale of application and hydrogen content (in wt%). It can be concluded that methane is again not suitable, even though it has a relatively high hydrogen content. Formic acid, and metal hydrides will only be available on a small scale and do not have high hydrogen contents generally. Values are also low for carbon structures and MOFs. Scoring high on both aspects is ammonia.
- **Carrier densities and efficiencies** are most beneficial in the case of methanol (with highest score for 3 out of 4 and no lowest score), closely followed by compressed hydrogen gas and ammonia (with 2 out of 4 and no lowest score). Exceptionally low values are found for the efficiencies of liquid hydrogen, even though its gravimetric density is the highest. The lowest densities are for metal hydrides and methane.
- **Safety** parameters include toxicity and explosiveness of the compounds. In this category, carbon structures and formic acid perform well, with no toxic or flammable classification. On the other side of the spectrum, there is methanol and ammonia, which appear to fit in both classifications, with one even classified as "highly". Research is limited for MOFs, zeolites and syncrude.
- **Financial projections** are best for compressed hydrogen gas storage in salt caverns and least beneficial for formic acid and MOFs.

An overview of this is given in the [appended excel file](#). Overall, the best performance can be attributed to compressed hydrogen gas, followed by ammonia and liquid hydrogen. The third best is methanol. Least promising are methane, metal hydrides, formic acid, and MOFs. This results in the following top 3:

Top 3 best options for hydrogen storage

1. Compressed H_2 gas
2. Ammonia & Liquid H_2
3. Methanol

Based on CC

Defining a top 3 based on targets

In the appended excel file, a column is added to count all cells with red text in it, containing values that are in line with the targets set by the US DOE (as described in the chapter [Hydrogen storage technologies | targets](#)). In total, 7 parameters are compared, of which only 6 are inherently different categories - there are two values for volumetric capacity that are considered one category in this thesis. That means that the highest score is achieved when 6 out of 7 targets are reached.

Targets are reached in 6 out of 7 cases for ammonia, which makes this compound the most promising option for hydrogen storage. It therefore receives first place in the ranking. This place is shared with methanol, which also scored for 6 out of 7 target parameters. The second best, based on the US DOE targets, is hydrogen storage in metal hydrides, with a score of 5 out of 7. A shared third place is given to liquid hydrogen and compressed hydrogen gas, with 4 out of 7. This results in the following top 3:

Top 3 best options for hydrogen storage

1. Ammonia & Methanol
2. Metal hydrides (*in general*)
3. Liquid H₂ & compressed H₂ gas

Based on T

It can be concluded that the top 3 based on the comparison criteria in this thesis and the top 3 based on the targets set by the US DOE differ slightly. This is probably because the comparison criteria also take the safety aspect into account, while the targets do not. Also, the targets are not set for all criteria discussed in this report. A very important thing to take away - setting aside the fact that not every option is investigated enough and lacks data on some criteria - is that in both cases **ammonia, methanol, compressed hydrogen gas, and liquid hydrogen make it to the top 3, these are thus the most promising options**. However, methanol and ammonia are currently classified as both toxic and explosive, which lowers their value (this is included in the top 3 based on the comparison criteria). The same, regarding explosiveness counts for free hydrogen in gas and liquid form. Therefore, it is suggested that more research on this matter follows.

DISCUSSION

The findings in this thesis have provided insight into the possible hydrogen storage technologies and their pros and cons based on the parameters provided through a literature review. Though, it should be noted that research always comes with limitations, as is also the case in this thesis. In this chapter, those limitations are discussed and a recommendation for future research is provided.

Limitations

Due to a lack of data - or an abundance of different contradictory data - in the tables provided in this thesis, a 100% accurate comparison is out of range. Besides, due to the short amount of time available for this research project, it is possible that some hydrogen storage options are, by accident, left out of the overview and comparison. However, close attention is paid to the accuracy of the schematic overview and the data it is based on, categories are not consciously left out. Only in some cases, the abundance of examples within a category (like the different metal hydrides) made it almost obligatory to make a founded choice which to mention. This is again based on the data availability on that matter.

It should be kept in mind that this report is part of a 7/8-week bachelor thesis program and written by a bachelor student in Environmental Sciences. Only basic physics and chemistry were part of the curriculum making it harder to properly judge certain physical or chemical properties - stated in very technical research papers used in this thesis - on their applicability. Therefore, one should not blindly take over the results stated in this thesis, but rather go back to the source of the information.

Recommendations

It is highly recommended that research in this field of research continues, resulting in a higher-level comparison. It is therefore required that research on Metal Organic Frameworks (MOFs), metal hydrides, carbon structures and zeolites advances, and possible new options are introduced. Besides, comparison criteria could be expanded to contain all important aspects of hydrogen storage. Suggested parameters are investment costs for the hydrogen system, type of storage tank needed (and what kind of materials these require), storage capacity (or space demand) of such tanks, human handling (how easy is it to use this type of storage?), ecology (what happens when this type of hydrogen storage is not used anymore?), sustainability (energy requirement for example). But there are without a question much more relevant parameters to investigate. On top of that, more detail could be given to the hydrogen storage parameters and what these mean to the values in the tables, including how the options that do not reach targets could improve or whether they are considered not realistic.

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