



Review article

Metal hydrides for hydrogen storage – Identification and evaluation of stationary and transportation applications

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ABSTRACT

Hydrogen is becoming increasingly important to achieve the valid defossilization goals. However, due to its physical properties, especially the storage of hydrogen is challenging. One option in this regard are metal hydrides, which are able to store hydrogen in chemically material-bound form. Against this background, the goal of this paper is an analysis of possible technical application areas of such metal hydrides – both regarding transport and stationary application. These various options are assessed for metal hydrides as well as selected competing hydrogen storage options. The investigation shows that metal hydrides with a temperature range below 100 °C (e.g., TiFe) are of interest particularly for transportation applications; possible areas of application include rail and marine transportation, as well as selected non-road vehicles. For stationary applications, metal hydrides can be used on low and high temperature levels. Here, metal hydrides with operating temperatures below 100 °C are particularly useful for selected small-scale applications (e.g., home storage systems). For applications with medium storage capacities (100 kWh to 100 MWh), metal hydrides with higher temperature levels are also conceivable (e.g., NaAlH₄). For even higher storage demands metal hydrides are less promising.

1. Introduction

A continued and intensified defossilization of energy systems is crucial to achieve climate neutrality in the near future. During this development, hydrogen generated from renewable sources of energy will most likely become an important secondary energy carrier (e.g., [1]). In this context, the question of how hydrogen for a specific application case can be stored as energy-efficiently, safely and cost-effectively as possible needs to be answered.

Currently, various hydrogen storage options are in different phases of technological development; this is true for the “classical” pressurized storage, for cryogenic liquid hydrogen storage and for a storage in derivatives such as ammonia or liquid organic hydrogen carriers (LOHC) as well as in metal hydrides. The most common types of hydrogen storage to date, cryogenic and pressurized, require high energy inputs for storage (especially cryogenic) (i.e., high pressure level, low temperature) and especially for cryogenic solutions an ongoing energy demand. Metal hydrides can easily realize long-term hydrogen storage without an ongoing energy demand; they only require thermal energy for storage discharge. Therefore, this could be advantageous for certain specific application cases.

Since the 1960s, research has been conducted in the field of metal hydrides [2]. So far, the main research lines focus on the identification and optimal combination of possible storage materials (e.g., reactive hydride composites) to achieve the highest possible gravimetric energy storage density (e.g., [3]). In addition, there are only few specific examples of applications for metal hydrides as truly promising energy storage devices in literature (e.g., [4]); i.e., there is only limited knowledge available related to the most relevant application areas for metal hydrides within the various energy sectors for demand-oriented energy storage.

Against this background this paper aims to contribute to this topic by

- providing a systematic technical analysis of the conditions, challenges and opportunities for the use of metal hydrides for hydrogen storage in various energy sectors and by
- investigating an efficient integration of metal hydrides into the energy system.

For this purpose, after a detailed consideration of the conditions and necessities in each sector, with a distinction and more detailed insights into transportation and stationary use cases, application examples will

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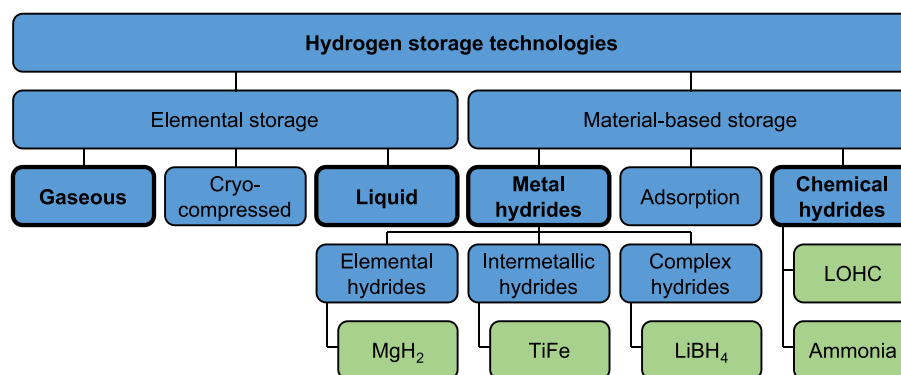


Fig. 1. Overview hydrogen storage technologies, adapted from [6] (MgH_2 Magnesium hydride; TiFe Ferrotitanium; LiBH_4 Lithium borohydride; LOHC Liquid organic hydrogen carrier).

be identified and the respective technical feasibility will be assessed. The base year for this assessment is today (2023) with an outlook on 2040.

2. Basics of hydrogen storage technologies

Hydrogen can be stored in a pure form as well as physically or chemically bounded to and in (gaseous, liquid and solid) materials [5]. Fig. 1 shows these different forms of hydrogen storage together with the classification applied in this work with examples of chemical and metal hydrides. Storage technologies compared in this work are marked in bold.

Below, the theoretical background of the most promising hydrogen storage technologies is discussed including a placement of metal hydrides within other hydrogen storage technologies as well as a brief outline of the core parameters of important forms of hydrogen storage. This is followed by an explanation of different types of metal hydrides currently discussed for potential stationary and transport applications, without providing a full overview of all existing metal hydrides (see e.g., [4]). Explanations are made for the elemental storage forms of liquid and compressed hydrogen as well as for liquid organic hydrogen carriers (LOHC) and ammonia. The latter two are chosen, since LOHC are under discussion for the transport sector in general and ammonia for the maritime sector in particular.

2.1. Metal hydrides

In the case of metal hydrides, hydrogen is chemically bound to metals or metal alloys. Within such a hydrogen absorption process, hydrogen first dissociates when reaching the metal surface and then diffuses into the metal lattice. Here it reacts by releasing heat (exothermic reaction) to form a metal hydride. After the reaction is complete, the hydrogen is bound in the material and is stored under ambient conditions [6].

Pressure and temperature ranges at which the reaction takes place, as well as physical properties of different metal hydrides may differ considerably resulting in a wide range of possibilities for using metal hydrides as a hydrogen storage [6]. According to [6], the general reaction of the process can be described with Eq. (2-1).



ΔH_R : Reaction enthalpy.

To release the hydrogen from the metal by dehydrogenation, heat must be supplied (endothermic reaction) [6].

In addition to the explained thermochemical process for the hydrogenation and dehydrogenation of metal hydrides, which is currently the most studied approach, other methods are being investigated.

One such example is the electrochemical hydrogenation. Here, hydrogen accumulates at the electrodes of a battery through the

decomposition of an alkaline electrolyte. The accumulated atomic hydrogen penetrates the battery electrodes and forms metal hydrides [7]. During experimental investigation of the process in [7], the decomposition of electrodes and the accumulation of hydrogen took place when the battery was charged for several hours and the electrodes were heated up to 800 °C. Storage capacities of up to 20.1 wt% have been achieved, which could make the approach particularly promising for future applications [7,8]. Regarding dehydrogenation, there are also methods beyond the thermochemical process. For example, it is also possible to use a so-called thermal runaway of batteries. Here, dehydrogenation can be carried out under ambient conditions. Depending on the ambient temperature and the hydrogen accumulated at the battery electrodes, the charging current increases sharply after a certain time at a specific applied battery voltage and the electrolyte begins to boil and releases hydrogen. Experiments have shown significantly higher desorption rates than with thermochemical desorption [9].

In this article, however, these alternative processes for hydrogenation and dehydrogenation are not discussed any further, and only the most widespread approach of thermochemical (de)-hydrogenation is considered. In this context, particular emphasis is placed on the usage and supply of heat for (de)hydrogenation when identifying the application cases.

To understand the potentials and limitations of such a metal hydride storage, below a classification is realized by differentiating between elemental, intermetallic and complex metal hydrides [5].

2.1.1. Elemental hydrides

The chemically simplest form of metal hydrides are elemental metal hydrides. Here, hydrogen bonds to only one element (e.g., magnesium, aluminum, titanium).

Magnesium hydride has a theoretical storage capacity of 7.6 wt%. Magnesium is cheap and readily available. However, the chemical bond of magnesium hydride is very strong and about 75 kJ/mol H_2 of energy must be spent for desorption. Moreover, with a desorption temperature of around 300 °C, magnesium hydride is a high-temperature metal hydride, which makes it less suitable for, e.g., transportation applications. However, the charging and discharging behavior can be improved by additives (e.g., titanium compounds), which has been a particular focus of research in the field of magnesium hydrides in recent years. In addition, magnesium hydride shows a high cycle stability. For more detailed descriptions, especially about additives to improve reaction kinetics of magnesium hydride, reference is made to [10–16].

In contrast to magnesium, aluminum is relatively weakly bound to hydrogen, with a desorption energy of around 7 kJ/mol H_2 at a temperature of 100 °C. The theoretical storage capacity of aluminum hydride is very high at around 10.1 wt%. However, since very high pressures are required for charging, the absorption reaction is hardly reversible (a direct reaction of aluminum and gaseous hydrogen is not

Table 1
Parameters of intermetallic hydrides ([20]).

Parameter	AB	AB ₂	AB ₅
Operating temperatures [°C]	0 to 100	−50 to 150	0 to 200
Operating H ₂ pressure [bar]	1 to 30	1 to >1000	0.1 to 500
Storage capacity [wt%]	1.75	1.90	1.50

possible); i.e., regeneration of the aluminum hydride can only occur via electrochemical processes. Analogous to magnesium hydrides, titanium compounds as additives can also improve the performance of aluminum hydrides. For detailed descriptions, reference is made to [17–19].

Titanium, another elemental hydride shows operating temperatures between 650 and 750 °C at low pressures. Hydrogen storage capacity is only around 1 wt% [5,20,21].

2.1.2. Intermetallic hydrides

Intermetallic hydrides show typically lower operating temperatures compared to elemental hydrides. They consist of at least two metals as bonding partners for the hydrogen. The scheme of alloys corresponds to A_xB_yH_z, where an alloy or an element A binds the hydrogen (H_z) strongly and a metal element B binds the hydrogen weakly. In practice, three main ratios of the A_xB_y scheme have emerged: AB, AB₂ and AB₅. Such metal hydrides show extremely different properties, but they all have hydrogen storage capacities generally below 2 wt%. The operating temperatures of such intermetallic hydrides are between −50 and 200 °C (i.e., low-temperature or “room-temperature” metal hydrides and medium-temperature metal hydrides) [20]. Therefore, these hydrides may be well suited to be operated in combination with a low-temperature “waste” heat source. Table 1 shows important parameters.

The alloy lanthanum penta-nickel (LaNi₅) as a typical example shows a maximum theoretical hydrogen storage capacity of 1.5 wt% and can be operated at room temperature with a pressure of 1.8 bar [22]. Due to the high price of lanthanum, alternatives like titanium, zirconium or magnesium, are used. Another alternative is the AB alloy ferrotitanium (TiFe) already used commercially in combination with manganese [23]. This hydride operates below 100 °C and has a hydrogen maximum theoretical storage capacity of around 1.85 wt%. Due to the high price of titanium other alloys and compositions, such as the addition of manganese, are investigated as alternatives [5,20,24].

2.1.3. Complex metal hydrides

Another group of metal hydrides are so-called complex metal hydrides. Here, the hydrogen is bonded with a complex anion in combination with a light metal. Typical anions are aluminum hydrides ([AlH₄][−]), boron hydrides ([BH₄][−]), and amines ([NH₂][−]). Typical light metals are lithium and sodium. Complex metal hydrides have the highest hydrogen storage capacity of all metal hydrides; in the case of metal borohydrides this can be up to 18.5 wt%. However, most complex metal hydrides are high-temperature metal hydrides (partly >300 °C). This is due to the strong bonding; in the case of lithium borohydride (LiBH₄), for example, the desorption energy is 75 kJ/molH₂ with the consequence that a direct application is sometimes impossible in cases, where the high hydrogen storage capacity would be especially advantageous (e.g., transportation applications).

One approach to overcome this disadvantage is to mix other metal hydrides with boron hydrides. The results are reactive hydride composites (RHC) [5]. The combination of LiBH₄ and MgH₂ is one popular example in the literature. MgH₂ acts as a destabilizing additive. By possibly adding an additive such as TiCl₃, the reaction temperature can be lowered from >400 °C at an equilibrium pressure of 1 bar in the presence of LiBH₄ alone to 225 °C by adding the corresponding additives. The reversible hydrogen storage capacity of such an RHC system is 8–10 wt%, while the maximum theoretical capacity of LiBH₄ is 18.5 wt% [25]. Additional additives, in particular other titanium compounds, can further improve the reaction kinetics and reduce the time required for

hydrogen absorption and/or dehydrogenation [26–29].

2.2. Other hydrogen storage options

For the sake of comparison, a short overview of important competing hydrogen storage options follows. This includes the “conventional” storage options of pressurized hydrogen, liquid hydrogen as well as hydrogen storage in liquid organic hydrogen carriers (LOHC) and ammonia.

2.2.1. Gaseous storage

For transportation applications in particular, hydrogen is stored at pressures between 200 and 750 bar at ambient temperatures. Within such a storage device, the volumetric energy density of hydrogen is increased from 10.7 MJ/m³ at standard conditions (1 bar, 25 °C) to up to 4276 MJ/m³ (750 bar, 25 °C). Depending on the weight of these tanks, a hydrogen storage capacity relative to the tank system of 2.5 to 6 wt% can be achieved. Depending on the initial pressure (outlet of the electrolyzer) as well as the implemented compression technology (e.g., reciprocating or ionic compressor) a compression of hydrogen to 750 bar might require around 15% of the hydrogen's lower heating value [30].

2.2.2. Liquid storage

If space is limited, the volumetric energy density of compressed hydrogen might not be sufficient for storing large scale quantities. Therefore, hydrogen in an elemental form can be stored as a liquid. This requires hydrogen to be cooled down to −253 °C resulting in a high energy demand amounting to 20 up to 50% (depending on the respective process) of the lower heating value of hydrogen. In addition, even with a (very) strong insulation, liquid hydrogen exhibits necessarily evaporation losses over time. These so-called boil-off losses can be as high as 1%/d for larger tanks and as high as 3%/d for smaller tanks, causing problems especially in the mid to long-term storage (vehicle standing for several days, seasonal hydrogen storage) [24,30,31]. Additionally, liquid hydrogen is stored typically in spherical tanks characterized by the smallest surface-to-volume ratio and, thus, the lowest possible heat input to minimize boil-off losses. However, for transportation storage applications typically cylindrical tanks must be built being less promising related to the surface-to-volume ratio [30]. The higher volumetric energy content reaches up to 8495 MJ/m³. For such transportation applications, liquid hydrogen tanks achieve hydrogen storage capacities of around 7 wt%, based on the respective system [30].

2.2.3. Liquid organic hydrogen carriers (LOHC)

Liquid organic hydrogen carriers allow hydrogen to be stored in liquid form at ambient conditions. For this purpose, hydrogen is bound to an organic carrier molecule. The hydrogenated liquid compound can be refueled and transported like a conventional liquid hydrocarbon-based fuel. For discharging, hydrogen must be separated from the organic carrier molecule by means of thermal energy and controlled by a catalyst. The hydrogen can afterwards be used, while the carrier molecule has to be stored separately to be recycled and re-loaded with hydrogen. Obstacles of such an approach are the two-tank requirement as well as the energy loss/demand for de-/hydrogenation of the carrier molecule. Process temperatures between 150 and 310 °C can be observed for hydrogenation and dehydrogenation. The hydrogen storage capacity varies between 4.5 and 12 wt% [32].

2.2.4. Ammonia

Hydrogen can also bond at nitrogen to form ammonia (NH₃) to realize a gravimetric energy density of 17.7 wt%. Therefore, existing plants for ammonia production using the Haber-Bosch process can continue to be operated after adaptations. Reaction conditions are at temperatures between 300 and 550 °C and pressures between 200 and 350 bar. Since the process is exothermic, no additional heat is required, but a high-pressure level is necessary. The subsequent extraction of the hydrogen from the ammonia molecule is accompanied by a high

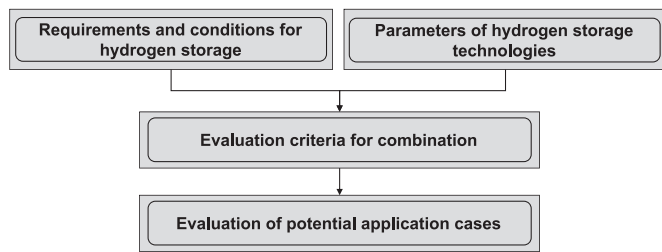


Fig. 2. Approach for identification and evaluation application cases.

(thermal) energy demand. A respective process is thermolysis releasing the hydrogen at temperatures between 400 and 650 °C; subsequently, a further downstream cleaning step is needed to remove unwanted impurities [5,33].

3. Methodical approach

For assessing the technical potential of metal hydrides as an alternative option for hydrogen storage compared to, e.g., physical storage, the methodological approach outlined below is used. This overall procedure is shown in Fig. 2.

- First, representative fields of application in the transportation and stationary sector, where hydrogen storage might possibly become essential in the future, are elucidated focusing on crucial requirements for hydrogen utilization.
- Second, parameters of different representative metal hydrides as well as competing hydrogen storage technologies are quantified to assess the storage performance.
- Third, evaluation criteria are defined for the technical suitability of hydrogen storage technologies based on the identified range of use-case related requirements for hydrogen storage on the one hand, and the identified range of technical parameters/characteristics of the considered hydrogen storage technologies on the other hand
- Fourth, based on the previously defined evaluation scheme, the different combinations of storage technologies and specific application are evaluated.

A detailed description of the different methodical steps is given below.

3.1. Requirements and conditions for hydrogen storage

The basis for assessing the technical suitability of a certain storage technology/option for a specific field of application is the evaluation of the required storage characteristics (e.g., volumetric storage density, charging rates) for each respective field of application. Here, a distinction is made between the requirements and conditions of stationary and transportation storage applications. Further, land-based, maritime and air-based transportation is differentiated. Additionally, the stationary sector is subdivided into the domestic and the industrial subsector.

The requirements and conditions include, e.g., maximum available space for hydrogen storage tank integration (i.e., acceptable tank volumes) or available waste heat temperature levels. The latter is primarily important for the use of metal hydrides due to necessary desorption temperatures. Fig. 3 provides a summary.

3.2. Parameters of hydrogen storage technologies

Assessing technical suitability is a need of an analysis of technical storage parameters. Thus, selected competing hydrogen storage options are evaluated besides metal hydride storage options. Therefore, different parameters, essential for the storage performance, are selected and analyzed for all considered storage possibilities. These parameters include, e.g., gravimetric energy densities, volumetric energy densities or, if applicable, operating temperatures (i.e., desorption or dehydrogenation temperatures for releasing the hydrogen from the respective storage system). Fig. 4 shows the storage technologies and the selected storage characteristics.

3.3. Definition of evaluation criteria for combination

For the assessment of the technical suitability of the considered storage systems for the identified fields of application, an evaluation matrix is defined providing an indication, whether a particular storage characteristic is fully sufficient, sufficient, or insufficient for the particular use case. This evaluation depends in particular on how relevant a requirement is for different field of application; e.g., if only little space is available in a particular use case and, thus, volumetric energy density is very important, a storage with moderate volumetric density might be insufficient, while it could be considered fully sufficient for a use case, where volumetric energy density plays a much less important role. For this categorization, ranges of the selected storage parameters

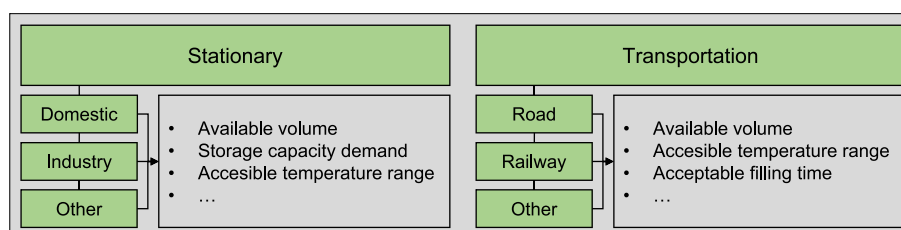


Fig. 3. Methodical step: requirements and conditions for hydrogen storage.

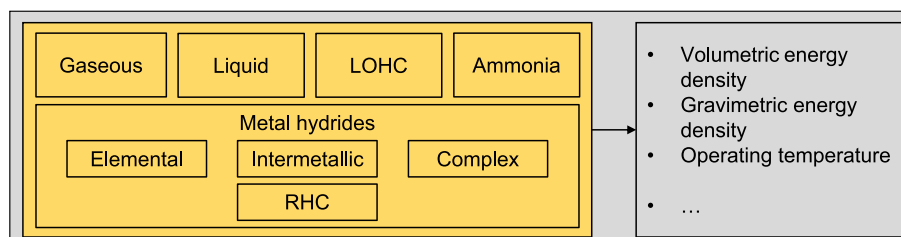


Fig. 4. Methodical step: parameters of hydrogen storage technologies.

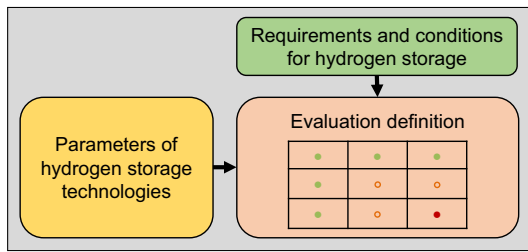


Fig. 5. Methodical step: definition of evaluation criteria for combination.

are defined relative to the best and worst option, so that at least one option can be considered fully sufficient for each use case requirement. The fit between storage parameter and use case requirement or condition is subsequently indicated by a traffic light system. Details are shown in Fig. 5.

3.4. Evaluation of potential application cases

For each of the resulting combinations, consisting of a storage technology and a use case, a qualitative assessment is performed

regarding the technical suitability or adequacy of the particular storage technology parameters for the corresponding field of application. Based on this evaluation, conclusions can be drawn as to which hydrogen storage technology is potentially the most promising for which application area based on the current state of technology. Thus, only a technical perspective is taken, also addressing potentially available storage system sizes. Economic aspects are explicitly left out. This methodical step is shown in Fig. 6.

4. Utilization areas of energy and hydrogen storage and their requirement profiles

To evaluate possible use cases of metal hydrides as energy storage, first the most beneficial use of hydrogen within the overall energy system has to be identified. Several corresponding attempts have been already made (e.g., [34–36]). Typically, hydrogen usage is assessed here within an economic competition with direct electrification; e.g., in the framework of the so-called “Hydrogen Ladder” [35], the use of hydrogen is ranked from economically promising to less promising in a hierarchical way on a qualitative basis.

Here, the focus is shifted to a purely technical assessment because a reliable economic framework conditions of the not yet fully

Evaluation of potential application cases							
Stationary				Transportation			
Storage option / Use case		Stat. 1	Stat. 2	Storage option / Use case		Trans. 1	Trans. 2
Metal hydride 1	Parameter 1	●	●	Metal hydride 1	Parameter 1	●	●
	Parameter 2	●	○		Parameter 2	●	○
	Parameter 3	○	●		Parameter 3	○	●
Metal hydride 2	Parameter 1	●	●	Metal hydride 2	Parameter 1	●	●
	Parameter 2	○	●		Parameter 2	○	●
	Parameter 3	●	●		Parameter 3	●	●
Liquid hydrogen	Parameter 1	●	○	Liquid hydrogen	Parameter 1	●	○
	Parameter 2	●	●		Parameter 2	●	●
	Parameter 3	●	●		Parameter 3	●	●

Fig. 6. Methodical approach: evaluation of potential application cases.

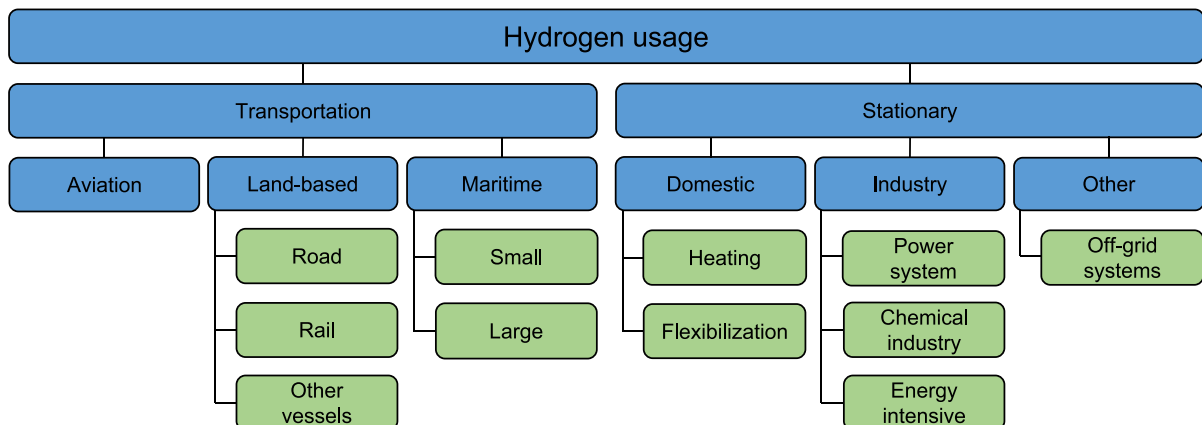


Fig. 7. Potential application cases for hydrogen storage, according to [34–36].

Table 2

Selected technical system targets for onboard hydrogen storage for passenger cars by the United States Department of Energy (DOE) [43].

Storage parameter	2020	2025	Ultimate
Storage capacity system ^a [wt%]	4.5	5.5	6.5
Volumetric density system [kg _{H2} /m ³]	30	40	50
System fill time [min]	3–5	3–5	3–5
Average flow rate [(g/s)/kW]	0.02	0.02	0.02
Minimum/maximum delivery temperature [°C]	–40/85	–40/85	–40/85

^a Based on kg stored hydrogen per kg system.

commercialized and still economically developing technology of metal hydrides is hard to estimate. Regarding possible application cases the division and classification according to Fig. 7 is adopted [34–36]. Because the evolutionary process within the hydrogen production sector is still ongoing, the consideration in which sector hydrogen will play which role within the respective energy system in the years to come is strongly dependent on the assessed time horizon [1]. Additional, when analyzing metal hydrides, possible waste heat potentials together with the respective temperature level should be taken into consideration. These usually play less of a role in the evaluation of “conventional” hydrogen storage options, but are of central importance for the energetically efficient use of metal hydrides requiring heat for the hydrogen release process (desorption).

The scheme in Fig. 7 includes applications where a hydrogen storage tank might be needed for intermediate storage. These different sectors are discussed in detail below.

4.1. Hydrogen storage within the transportation sector

The core aspects for storing hydrogen within the transport sector are volumetric energy density, refueling speed and range (volume, energy content). Weight also plays a significant role in certain transport applications [6]. Thus, the sub-areas listed in Fig. 7 are examined as examples. Acquired data for the various use cases are collected in Tables 3 and 4 and rely on existing examples described below and on a transfer of certain technology parameters of other – e.g., conventional – systems. A special focus is put on the possible waste heat different vehicles can provide, as this is essential for the utilization of metal hydrides.

4.1.1. Land-based vehicles

For the evaluation of road traffic, three different utilization concepts are differentiated: Individual road transport (cars), heavy-duty and bus transport as well as railway traffic and other vehicles, which are not underway on public roads.

Table 3 summarizes and specifies the data for the land-based applications described below; in addition to storage capacity and temperature levels, the hydrogen storage volume and weight as well as refueling times are also shown.

Table 3

Parameters for land-based utilization cases of hydrogen.

Application case	Storage capacity ^a [kWh]	Propulsion technology	Temperature level [°C]	Acceptable filling time	Volume tank system	Weight tank system	Reference
Railway	8000–9000	LT-PEMFC	50–80	Few minutes	Medium relevant	Less relevant	[51,53,54,60]
Road traffic (individual)	200–300	LT-PEMFC	50–80	Few minutes	Very relevant	Very relevant	[41,42]
Road traffic (heavy duty)	1000–2500	LT-PEMFC	50–80	Minutes	Very relevant	Very relevant	[46,51,61,62]
Road traffic (busses)	1100–1500	LT-PEMFC	50–80	Minutes	Very relevant	Very relevant	[49,63]
Non-road vehicles (heavy)	600–700	LT-PEMFC	50–80	From minutes to hours	Very relevant	Medium relevant	[56,58]
Non-road vehicles (heavy goods)	10–500	LT-PEMFC	50–80	From minutes to hours	Very relevant	Medium relevant	[58,64,65]

LT-PEMFC Low temperature polymer electrolyte membrane fuel cell.

^a The storage capacity is based on data in the literature and converted with the lower heating value for hydrogen [66].

4.1.1.1. Individual road transport. Within this transport area, individual transport is most advanced on the pathway towards defossilization. The development in this application area seems currently clearly to be heading towards a complete electrification. This can be explained by increasing capacities and decreasing charging times for batteries; i.e., faster charging time and longer range of hydrogen driven fuel cell vehicles loose increasingly relevance. Ranges of up to 400 km on a full charge and fast charging for a range of 200 km in 15 min are possible [37].

In addition, the total cost of ownership for fuel cell cars is >50% higher than for battery vehicles. This is mainly due to significantly higher CAPEX and OPEX of fuel cell cars [38,39]. Nevertheless, fuel cell vehicles are available for passenger transport; the Toyota Mirai and the Hyundai Nexo are the most prominent examples [40]. The ranges of the designated vehicles are between 650 and 800 km. The waste heat required for the potentially use of a metal hydride storage can be obtained from the polymer electrolyte fuel cell (PEMFC) used typically in such vehicles. Such a PEMFC operates at a temperature level between 50 and 80 °C (Table 3) [41,42].

The United States (US) Department of Energy (DOE) has formulated target values that hydrogen storage systems for passenger car fuel cell vehicles should ideally achieve by certain target years and as final target (ultimate) [43]. Some selected parameters, which are also addressed in the context of this work, are listed in Table 2.

4.1.1.2. Heavy-duty road transport. Although there is an increasing trend towards the development of battery-electric trucks (e.g., [44]), the use of hydrogen in heavy-duty trucks offers still a great potential because still insufficient battery capacities, the long charging time and the resulting short travelling ranges pose a problem. While Diesel and hydrogen vehicles have a refueling time of <20 min, this time can range from 30 min to up to 11 h for battery-electric heavy-duty trucks [45]. Similarly, the range of a battery-electric truck currently amounts to almost 800 km, while that of Diesel trucks is up to 3000 km and that of hydrogen operated heavy-duty trucks can reach up to 1800 km [46]. Thus, the hydrogen-powered heavy-duty truck is a true alternative to the battery-electric vehicle. The most advanced heavy-duty truck in this field at present is the Hyundai Xcient Fuel Cell. Its range is up to 400 km and analogous to individual transport, a PEMFC seems conceivable for heavy-duty transport according to the current state of development and knowledge [46] resulting in a usable temperature level of the waste heat of 50 to 80 °C (Table 3).

4.1.1.3. Bus transport. In contrast to heavy-duty trucks, where the focus is usually on high travelling ranges (especially for long-distance transport), local transport busses usually operate on clearly defined sections at fixed times. The distances to be overcome on a daily basis are <200 km, at least in urban areas, making range less relevant than for cars and long-haul heavy duty trucks [47]. The ranges of battery-electric busses vary from 100 to about 350 km. Without controlled charging, the

Table 4
Parameters for maritime use cases and aviation use of hydrogen.

Application case	Storage capacity ^a [MWh]	Propulsion technology	Temperature level [°C]	Acceptable filling time	Volume tank system	Weight tank system	Reference
Maritime ^b (small vessels)	0.8–18	LT-PEMFC	50–80	From minutes to 1 h	Less relevant	Less relevant	[73,74]
Maritime ^c (large vessels)	10,000–40,000	LT-PEMFC MCFC	50–80 600–700	Up to several hours	Less relevant	Less relevant	[69,75–77]
Aviation	0.03–170	LT-PEMFC SOFC Jet engine	50–80 700–800 500–850	From minutes to 1 h	Very relevant	Very relevant	[71,78–82]

LT-PEMFC Low temperature polymer electrolyte membrane fuel cell; MCFC Molten carbonate fuel cell; SOFC Solid oxide fuel.

^a The storage capacity is based on data in the literature and converted with the lower heating value for hydrogen [66].

^b Small vessels include ferries and small passenger ships.

^c Large vessels include tankers and cargo ships.

charging time is 15 to 270 min [48]. Whereby, the range of fuel cell busses is between 360 and 500 km with a refueling time of occasionally <10 min. As a potential use of metal hydride storage depends on the utilization of heat, for busses operated by a PEMFC thermal “waste” heat is available between 50 and 80 °C [49]. Details are given in Table 3.

4.1.1.4. Railway transport. Railway transport is usually not mentioned as the first area in the transport sector where hydrogen use is discussed due to the much more common operation via electric overhead lines in Europe. However, on average in Europe, only 57% of the rail lines under operation are electrified [50] in particular due to the high cost of laying overhead lines; therefore, especially for rural areas with low frequency services electrification is hardly realizable economically [51]. Battery usage is hard to implement because high power is required, charging takes time, and the range of battery trains is typically too short.

One example for a battery-driven train is the “Flirt Akku” (company “STADLER”) characterized by a travelling range of 220 km [52]. This trains work as a “gap filler” between existing electrified tracks.

Thus, there are no plans known to introduce battery-driven trains on long rural (non-electrified) tracks. Usually, an operation via Diesel-powered trains is realized. But hydrogen operation is particularly suitable for such routes. There are already first routes e.g., in Germany, operated with compressed hydrogen and realizing regional traffic. This involves storing 260 kg of hydrogen, which, at a consumption of 26 kg/100 km, gives a range of around 1000 km [53,54] being most likely a minimum requirement for such applications. Such hydrogen trains also use PEMFC providing waste heat [55] (see Table 3).

4.1.1.5. Off-road vehicles. There are other land-based vehicles that are not (or hardly) present in the public space. Some examples are:

- Construction site vehicles (wheel loader),
- Agricultural machinery (tractor),
- Airport traffic (baggage tug),
- Port traffic (automated guided vehicle (AGV)), and
- Warehouse logistics (forklift).

Despite the different configurations, sizes and usage concepts of such vehicles, they have in common that they either have to handle or transport heavy loads or require counterweights (e.g., forklifts). There are hardly any known hydrogen-powered alternatives to Diesel operation or battery-driven vehicles for the designated off-highway vehicles.

One example is an agricultural tractor from the Fendt company. The vehicle is operated with a PEM fuel cell from a 350 bar pressure tank. It can be operated for 4 to 8 h depending on the load (Table 3) [56]. For other vehicles, mainly battery-electric solutions are currently under discussion. As an example, diesel-powered automated guided vehicles (AGV) at automated container terminals in harbors can be converted to battery-powered vehicles [57,58] (Table 3).

A distinction is made between heavy vehicles (e.g., wheel loaders) and vehicles which transport heavy goods (e.g., forklifts). Analogous to the road-based sector, low ranges and long charging times are currently still barriers to the introduction of battery-electric drives in many of these non-road applications, in particular due to high weights and loads implying high required power [59].

4.1.2. Marine vessels

A wide variety of ship sizes and types exists. For simplicity, only the three ship types ferry, inland cargo ship, and container ship are examined being representative for small and large ships.

The ferry is the only one for which electrification of the propulsion system via batteries is, at present, possible due to its limited range and relatively low load. Battery-electric ferries are already in use or will be

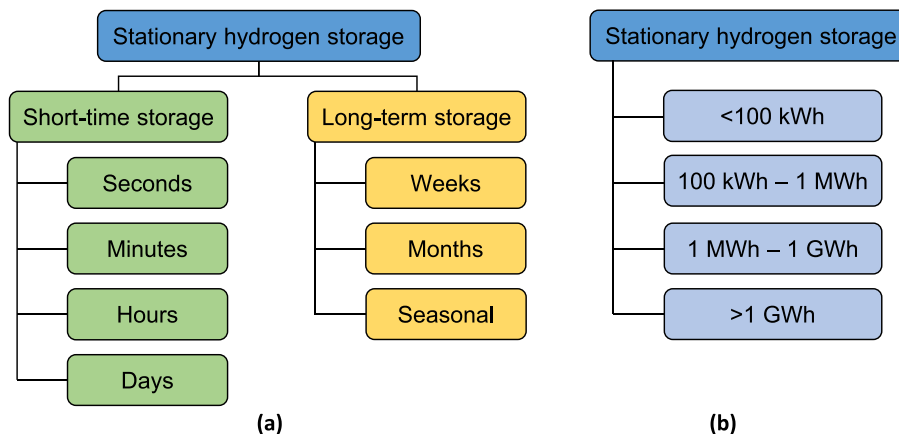


Fig. 8. Classification concepts of stationary energy storage ([83,84]).

Table 5
Parameters for domestic utilization cases of hydrogen.

Application case	Storage capacity [kWh]	Waste heat technology	Temperature level [°C]	Volume tank system	Weight tank system	Reference
Domestic (houses)	200–1500	PEMFC	50–80 120–200	Medium relevant	Medium relevant	[84,87,88,91,92]
Domestic (districts ^a)	200–4000 ^{b,c}	PEMFC	50–80 120–200	Less relevant	Less relevant	[87,88,93–95]

PEMFC Polymer electrolyte membrane fuel cell; PEMEL Proton Exchange Membrane Electrolyzer; AEL Alkaline Electrolyzer; SOEL Solid Oxide Electrolyzer.

^a Includes residential neighborhoods and commercial properties.

^b The storage capacity is based on battery data in the literature and converted with the lower heating value for hydrogen [66] and the higher storage requirements for the use of hydrogen by means of fuel cells compared to battery storage [96].

^c Partially, based on data for battery storage ([97]), conversion losses of PEM electrolyzer and fuel cell taken into account (electrical efficiency) [98,99].

Table 6
Parameters for industry utilization cases of hydrogen.

Application case	Storage capacity	Waste heat technology	Temperature level [°C]	Volume tank system	Weight tank system	Reference
Industry (chemistry)	9–120 MWh ^a	–	Various	Less relevant	Less relevant	[113,114]
Industry (energy-intensive)	3–100 GWh ^a	various	25–1650	Not relevant	Not relevant	[109,111]
Power system (seasonal)	40–4000 GWh	–	–	Not relevant	Not relevant	[103]
Power system (short-time)	3–220 MWh ^{a,b}	PEMEL AEL SOEL	20–100 40–90 650–1000	Onshore: not relevant Offshore: very relevant	Onshore: not relevant Offshore: less relevant	[97,104,119]
Off-grid systems	3 MWh–2 GWh	PEMFC SOFC	50–80 700–800	Various	Less relevant	[115–118,120]

PEMFC Polymer electrolyte membrane fuel cell; PEMEL Proton exchange membrane electrolyzer; AEL Alkaline electrolyzer; SOEL Solid oxide electrolyzer; SOFC Solid oxide fuel.

^a Conversion from Nm³ to kWh according to [121].

^b Based on data for battery storage ([97]), conversion losses of PEM electrolyzer and fuel cell taken into account (electrical efficiency) [98,99].

operated in the future (e.g., in New Zealand, in the Baltic Sea) [67,68].

As an alternative, fuel cell propulsion can be used for ferries. Low-temperature polymer electrolyte fuel cells (LT PEMFC) and molten carbonate fuel cells (MCFC) are particularly suitable technologies, with the majority of research projects being based on LT PEMFC [69]. The resulting temperature levels that could act as a possible heat source for the desorption process of metal hydrides are shown in Table 4.

In the case of inland waterway vessels, the required energy can hardly be provided by batteries. So, for these hydrogen fuel cells can be considered as an alternative to conventional propulsion (Table 4).

4.1.3. Aviation

Air transport is another challenging part of the transport sector in terms of defossilization. In addition to the widely discussed approach of replacing fossil aviation fuels with sustainable aviation fuels (SAF), hydrogen as a fuel offers another option [70]. Hydrogen could be used in fuel cells, with research activities focusing on the application of polymer electrolyte membrane fuel cell (PEMFC) and solid oxide fuel cell (SOFC). Hydrogen can also be used in aircraft turbines commonly used within the commercial aviation industry basically on all distances. But this option is challenging and currently still under development [71,72]. Hydrogen storage in aircrafts need to achieve a high volumetric and gravimetric energy density as well as a high refueling rate in order to limit the time on the ground [71] (Table 4).

4.2. Hydrogen storage within the stationary sector

Different stationary sectors show various storage needs. This is true for storage systems used within the electricity and heating sector, for stationary fuel storage in the transport sector and for raw material storage within the chemical sector [83]. Fig. 8 shows two selected ways to classify these stationary storage options either by time of storage (a [83]) or by storage capacity (b [84]). Especially for the power sector, time-based classification plays an important role in decoupling electricity production based on fluctuating sources of energy from the given consumption of the respective grid [84]. Here, both classifications are taken up (Fig. 8).

These very different stationary energy storage technologies are compared below (Tables 5 and 6). Analogous to the transportation sector, parameters like storage capacity as well as the relevance of weight and volume remain – even if their importance may be less pronounced. The temperature level parameter describes the technology, which, if applicable, uses the stored hydrogen after the withdrawal. Also, other process heat sources are considered (e.g., from the energy-intensive industry). This is again relevant as the efficient desorption of hydrogen from metal hydrides requires the sufficient heat.

4.2.1. Domestic applications

A distinction is made between single private household (houses) and several connected households of a district related to the types of storage requirements for residential latency.

4.2.1.1. Houses. Within the domestic energy system, battery storage systems are currently mainly used to store electricity from photovoltaic (PV) systems to be used during night time [85]. Such storage devices for single-family homes typically have a capacity of 5 to 10 kWh [84]. An alternative concept still subject of research involves a hydrogen system in addition to a PV system. In this case, PV electricity is used for direct electricity supply and for water electrolysis to generate hydrogen being stored within a storage tank. In case of high load or lack of PV power, electricity can be provided by means of a fuel cell using the generated hydrogen from the storage [86].

When storing hydrogen possibly within metal hydrides under ambient conditions, the necessity of at least a partial seasonal storage gains momentum within the public debate. Thus, the storage capacities of existing hydrogen storage providers and additional data are listed in Table 5. Additionally, the conversion technology using the stored hydrogen offers the possibility of heat provision in addition to the possibility of storing electricity (as hydrogen) seasonally [87]. This is of interest both for the use in a heating system and for heating a metal hydride storage tank for hydrogen desorption. Additionally, the issue of rapid loading and unloading is less of an issue compared to transportation use cases. Both low-temperature PEM fuel cells (LT PEMFC)

and high-temperature PEM fuel cells (HT PEMFC) are conceivable for these applications. However, commercial sales are not yet taking place to a large extent. The temperature ranges from 50 to 80 °C (LT PEMFC) and from 120 to 200 °C (HT PEMFC) are possible for a potential desorption process of a metal hydride storage material [87,88].

4.2.1.2. Districts. From an overall energy system perspective, instead of installing energy storage systems for each individual single-family home, neighborhood solutions are an alternative. This increases the degree of self-sufficiency and the share of self-consumption of electricity from photovoltaic systems [89]. A transfer of the concept mentioned above therefore seems conceivable. The main differences to such systems designed for a use in single-family houses are the size of the storage components and the space and volume ratios [84]. Whereas in single-family homes these may be associated with limited space, these parameters play less of a role, especially in newly planned neighborhoods. Also waste heat for regional district heating can be used additionally to provide heat for unloading a metal hydride storage tank [87,90] (Table 5). Most likely, analogous to single family houses, high temperature PEM fuel cells would be included within such district energy systems.

4.2.2. Industrial applications

In industry, there are many areas of application in which energy storage is necessary and will be of increasing importance in the future. In the following, a broad range of applications is tackled without claiming to be exhaustive.

4.2.2.1. Power system. In the case of hydrogen storage requirements for the electricity sector, a distinction could be made between seasonal storage to bridge several months with large-scale storage capacities, and short-term storage with basically short-time storage capacities. For a widely defossilized energy system, the focus is still on the former option, in order to be able to compensate for an insufficient availability of fluctuating wind energy and solar irradiation by seasonally storing surplus electricity [84,100]. Therefore, below between seasonal, large-scale and predominantly centralized as well as short-term, small-scale and probably decentralized hydrogen storage systems is distinguished.

If the share of renewable energies within the electricity system exceeds the demand given within the grid in parallel, hydrogen could be produced by means of electrolysis and stored afterwards. Thus, analogously to storage facilities for natural gas, large-scale hydrogen storage facilities are to be built. Here, especially salt caverns are considered [101,102]. Based on their sizes, the required storage capacities, based on the lowest and highest existing single cavern capacity, are listed in Table 6 assuming that only 70% of the working gas volume of natural gas can be stored in the form of hydrogen [103].

In addition to large-scale seasonal storage, short-time storage (a few hours or days) gains importance due to the increasing expansion of wind farms with the aim of converting parts of the energy into hydrogen to achieve a more even feed-in of wind-borne electricity. Due to the fact that salt caverns are not available everywhere, they are only an option for storing hydrogen provided offshore if the respective hydrogen pipelines are available [86]. If different wind farm projects where electrolyzers will be used in the future are studied [104], electrical outputs between 25 and 1000 MW became obvious. Assuming that a storage capacity of 24 h of hydrogen production of the electrolyzer should be buffered, daily storage capacities between just under 1000 and 55,000 m³ of hydrogen are required (Table 6) [104]. Currently, this hydrogen is stored in pressure tanks operated between 100 and 300 bar [84,105]. For onshore structures, the relevance of volume and weight is of secondary importance due to the typically available space. If, however, storage facilities or electrolyzers are installed offshore, a higher relevance of the volume to be consumed can be assumed due to the limited space available at offshore stations [104].

For other energy storage concepts in combination with wind and photovoltaics, data are given in Table 6. Conversion losses of PEM-based electrolyzers and fuel cells are taken into account, and larger storage capacities are assumed as a result.

4.2.2.2. Energy-intensive industry. The so-called “energy-intensive” industry is often cited as a particular challenge for the defossilization of industry. Assuming energy-intensive industries being defined to spend at least 3% of their production value on energy, this definition includes at least steel production, building materials (especially concrete), glass, non-ferrous metal processing and the chemical industry being considered separately here [106].

The steel industry as one example currently accounts for about 23% of industry-related CO₂-emissions within the EU, mainly emitted during the reduction of iron oxide by using coke [107,108]. Therefore, research is being conducted on direct reduction with hydrogen [109–111]. Concepts are published estimating an additional hydrogen storage requirement [109].

To cover the hydrogen demand emerging from this energy-intensive industries, the need of hydrogen provision by means of pipelines embedded within larger networks is expected. A large-scale hydrogen storage concept is therefore not envisaged for these application cases. For example, a storage capacity of around 100 GWh with a steel production of 1.2 Mt/a is expected. Other sources [109] expect for a steel production of 1 Mt/a hydrogen storage capacity of 90 t. Due to a wide variety of processes within the steel production sector, temperatures between 25 and 1600 °C are available [109]. This would allow the potential use of metal hydrides for a broad variety of different alloys. Further data are listed in Table 6.

4.2.2.3. Chemical industry. Hydrogen is already used in many processes within the chemical industry (e.g., production of ammonia and methanol, refining processes). This hydrogen used today is obtained almost exclusively from fossil fuel resources (natural gas, coal, by-products from refineries); a supply via water electrolysis operated by “green” electricity hardly takes place so far [112].

In order to determine the energy storage resp. hydrogen storage needs for the chemical industry to be defossilized, the current infrastructure must be considered. Up to now, hydrogen, primarily produced from natural gas in Western Europe, is mainly stored and transported via four routes.

- Hydrogen networks are common, especially in areas with a high industrial density and a spatial proximity of suppliers and consumers. For example, these are available in the Ruhr area in Germany able to compensate demand peaks and reduced consumption at individual consumers with a capacity up to 40,000 m³/h [113,114]. Even with increasingly renewable hydrogen supply, it can be expected that these networks will continue to be operated and that no alternative storage technology in the form of large-scale storage facilities will be needed for this specific area of application.
- Other forms of storage currently in use/in development include storage in tankers as liquid hydrogen, in which around 35,000 to 40,000 Nm³ of hydrogen can be stored and transported.
- In addition, hydrogen is usually transported under pressure of 200 bar in permanently mounted pressure pipes with trucks holding around 3000 to 6000 Nm³. As a pilot project, a trailer with a capacity of around 13,000 Nm³ has been announced.
- A small-scale option are individual pressure cylinders [114].

Since the described hydrogen infrastructure and transport options are to be considered independently of subsequent production or conversion processes, no substantial heat source can be assumed for the potential storage and withdrawal of hydrogen. Since hydrogen networks are highly unlikely to be replaced in the future, storage parameters of

Table 7

Parameters of selected metal hydrides.

Metal hydride	Type	Operating Temperature range ^a [°C]	Volumetric density ^{b,c} [kg _{H2} /m ³]	Storage capacity ^b , ^d [wt%]	Charge rate ^e [kg _{H2} /min]	Concept maturity ^f	Reference
LaNi ₅	Intermetallic	20–80	106	1.3	<1.0	Prototypes	[4,6,22,122–125]
TiFe	Intermetallic	30–70	98	1.5	<1.0	Market ready	[6,22,122–124,126,127]
LiBH ₄	Complex	460–1000	91	13.4	<1.0	Lab-Scale	[4,6,22,123–125,128,129]
NaAlH ₄	Complex	100–200	47	3.7	0.5	Lab-Scale	[4,6,22,123–125,128,130]
TiH ₂	Elemental	650–750	39	1.0	<1.0	Lab-Scale	[6,21,124,125,128,131]
MgH ₂	Elemental	250–500	80	5.5	<1.0	Prototypes	[4,6,22,123–125,132]
6Mg(NH ₂) ₂ -9LiH-LiBH ₄	RHC	90–180	74	4.2	<1.0	Lab-Scale	[6,124,133,134]

RHC Reactive hydride composite.

^a Pressures for the named temperature ranges are between 1 and 200 bar [5].^b Reversible volumetric density/storage capacity.^c Missing data for volumetric density is derived from gravimetric density.^d Based on kg stored hydrogen per kg storage material.^e For small-scale, transportation applications.^f Existing concepts are only available in small and medium scale.

the other storage options mentioned are listed in Table 6.

4.2.2.4. Other stationary systems. In addition to the above-mentioned applications for stationary use of hydrogen, there are many more off-grid systems. Typically, hydrogen is used in systems only small in size and are either completely separated from the rest of the power grid (off-grid systems) or have to provide electricity independently for a certain time in the event of a power failure (emergency power). So far, both island grids and such emergency power supplies are often covered by Diesel generators; for example, for an emergency power supply, usual bridging times up to 96 h are requested [115]. For a defossilized energy system, however, the integration of renewable sources of energies, especially electricity from wind and photovoltaics, in combination with battery and hydrogen storage is conceivable [116–118]. Possible applications are usually island grids or, for example, hospitals or data servers being urgently dependent on a stable, continuous power supply. Further data are listed in Table 6 (since a combination of battery and hydrogen storage typically makes sense from a systems point of view, the storage capacity for the entire system is not always shown in Table 5, but only the part of the hydrogen storage).

5. Metal hydrides and other technologies for hydrogen storage

Based, on the explanation and classification of existing metal hydrides given above, two metal hydrides from each of these classes already studied intensively are used as examples for further investigations. The respective characteristics are shown in Table 7 focusing on relevant parameters for possible application cases. Due to the growing interest in this type of metal hydrides in the current research, one reactive hydride composite (RHC) as a subclass of complex metal hydrides is added. For material-based hydrogen storage systems

(metal hydrides and chemical hydrides), material-based parameters and not system data are used in the following. This is due to the fact that only a few material-based hydrogen systems are commercially available to date and there is only little reliable data on individual storage systems.

The following parameters are defined to evaluate the storage performance.

- Operating temperature. This parameter describes the temperature range in which hydrogenation and dehydrogenation take place. Gaseous, compressed hydrogen is stored under ambient temperature and liquid hydrogen at -253 °C; i.e., this parameter is not applicable for such storage options.
- Volumetric energy density. This key figure is defined, depending on the storage technology, as the hydrogen mass stored per volume of the overall tank system (liquid and compressed hydrogen) or per volume of storage material (all other options). The reversible volumetric density is chosen for the storage materials, as this is of relevance for real applications.
- Storage capacity. This parameter is calculated as the hydrogen mass per mass of storage system (liquid and compressed hydrogen) or per mass of storage material (all other options). The reversible storage capacity is chosen for the storage materials, as this is of relevance for real applications.
- Charge rate. This figure described the hydrogen mass which can be stored within 1 min. Charge rates for storage systems from literature are typically only available for small-scale, transportation storage tanks. Thus, the charge rate for small-scale applications is assumed to be transferable to large-scale systems. This is also assumed for competitive hydrogen storage technologies (Table 8).

Table 8

Parameters of selected hydrogen storage options.

Storage technology	Volumetric density [kg _{H2} /m ³]	Storage capacity [wt %]	Charge rate ^a [kg/min]	Operating temperature range [°C]	Existing concepts	References
Liquid	62 ^b	7.2 ^b	1.5–2.0	–253	Established	[6]
Compressed	17–40 ^b	5.5–5.7 ^b	1.5–2.0	Ambient	Established ^f	[6]
LOHC A ^c	56 ^d	6.5 ^d	4.0	300–350	Prototypes ^f	[6,135,136]
LOHC B ^e	55 ^d	5.8 ^d	4.0	180–270	Prototypes ^f	[6,32,137]
Ammonia	121 ^d	17.7 ^d	1.5–2.0	400–650	Established	[5,33,138,139]

^a For small-scale, transportation applications, according to [6].^b Based on the system.^c Dibenzyltoluene, only dehydrogenation process of LOHC for waste heat usage is assumed.^d Based on kg stored hydrogen per kg storage material.^e N-ethylcarbazole, only dehydrogenation process of LOHC for waste heat usage is assumed.^f Existing concepts only available in small and medium scale.

Table 9

Evaluation criteria for evaluating and comparing different hydrogen storage technologies (symbol meaning: green – good, orange – medium, red – poor, “-” – not applicable/no data).

		Parameters use cases			
		Relevance Volume			
		No / less	Medium	High	
Parameters hydrogen storage technologies	Volumetric density [kg H ₂ /m ³]	> 90	●	●	●
		45 – 90	●	○	○
		< 45	●	○	●
			Relevance Weight		
			No / Less	Medium	High
	Storage capacity [wt.%]	> 8.0	●	●	●
		4.0 – 8.0	●	○	○
		< 4.0	●	○	●
			Relevance filling time		
			No / less	Medium	High
	Charge rate [kg/min]	> 4.0	●	●	●
		2.0 – 4.0	●	○	○
		< 2.0	●	○	●
			Operating temperature waste heat upper limit		
	Temperature level	Same value or Deviation < +25%	Deviation < -25%	Deviation > ±25%	
●			○	●	
		Heat usage possible			
		yes	no		
		●	●	●	
		Required storage capacity			
		0 – 100 kWh (small)	100 kWh – 100 MWh (medium)	> 100 MWh (large)	
Concept maturity ^a	Market ready / Established	●	●	● / ○ ^b / ● ^c	
	Prototypes	○	●	●	
	Lab-Scale	●	●	●	

^aBased on 2023; ^b Compressed hydrogen storage system are used today for small and medium scale hydrogen storage; ^c There are no market ready/established concepts in large scale for metal hydrides and LOHC (liquid organic hydrogen carrier).

- Concept maturity. The maturity is defined based on the technology available today (2023) and divided into four stages: Lab-scale, prototype, market ready and established.

For a classification of metal hydride storage, a comparison to other hydrogen storage technologies is performed. These alternative technologies include liquid (cryogenic) hydrogen storage, gaseous high-pressure hydrogen storage as well as hydrogen storage in two different liquid organic hydrogen carriers (LOHC) and ammonia. The first analyzed LOHC (LOHC A) is Dibenzyltoluene. Because of its high dehydrogenation temperature, as an alternative N-ethylcarbazole (LOHC B) is considered. LOHC B shows a lower operating temperature [32]. Detailed data are shown in Table 8.

6. Definition of evaluation criteria

The criteria for the evaluation and comparison of the different technologies are described in Table 9. A traffic light system with three levels is used for this qualitative assessment. Furthermore, the classification into the respective group was made on the basis of the range of

Table 10

Assumptions for concept maturity development until 2040.

		Concept maturity in 2040			
		0–100 kWh (small)	100 kWh–100 MWh (medium)	>100 MWh (large)	
Hydrogen storage technology	Metal hydrides	Lab-Scale ^a	Prototypes	Not used	
		Prototypes/market-ready ^a	Established	Not used	
	LOHC	Ammonia	Prototypes	Prototypes	Not used
		Compressed hydrogen	Established	Established	Established
		Liquid hydrogen	Established	Established	Not used
		Liquid hydrogen	Established	Established	Established

LOHC Liquid organic hydrogen carrier.

^a Concept maturity in 2023.

parameters outlined above; i.e., a “red” rating does not mean that a technology is ruled out for the application in question, but only that it scores worse in comparison to the other options considered here.

6.1. Evaluation of physical criteria for hydrogen storage

The evaluation in Table 9 is defined based on physical properties, relative to the value ranges shown above and the state of concept maturity to date.

6.2. Status and assumptions for the prospects of future hydrogen storage options

Since the further development of the various hydrogen storage technologies is subject to great uncertainty, this paper merely estimates the expected concept maturity. On the basis of the concept maturity, the size classes up to which the storage technologies will be available in 2040 are then indicated (Table 10).

The question to what extent metal hydrides will be suitable for large-scale systems in the future cannot be answered with certainty so far; statements in literature differ clearly (e.g., [20,140]). A relevant parameter of a possible development are the raw material prices for the alloys used in metal hydride storage systems [141]. Based on literature it can be assumed that the use up to a medium-scale range seems likely. To address the difference in the technological maturity of the various metal hydride materials, metal hydrides existing already in form of “prototypes” are assumed to be “market-ready” in 2040. “Lab-scale” metal hydride materials are at least available in form of prototypes in 2040. An estimation for large-scale applications will be omitted due to the given uncertainties (“not used”). LOHCs are at a similar stage of development to metal hydrides, which is why an analogous classification was adopted and large-scale usage in 2040 is excluded (“not used”).

Liquid hydrogen is already used in large-scale storage and can therefore be assumed to be available and “established” in 2040 in all scales [142].

Ammonia has already long been used on a large scale within the chemical industry [138]. Therefore, ammonia is to be regarded as an “established” technology that will most probably be relevant in 2040.

Compressed hydrogen is already widely used in the small- and medium-scale sector and can also be seen as an “established” mature technology. In large-scale applications, according to current knowledge the use of gas networks or salt caverns will be most likely. The term “compressed” in the following and in Table 10 means therefore only the use of pressurized hydrogen in storage tanks and not compressed hydrogen in salt caverns. The use of compressed hydrogen in “conventional” pressure tanks in large-scale systems is unlikely and must be regarded as “not used”.

7. Evaluation results of potential application cases

In the following, combinations of hydrogen storage technologies and applications are evaluated for the transportation and the stationary sector.

Table 11
Rating of different hydrogen storage options for transportation use cases.

Storage option / Application case	Railway	Road	Non-road vehicles (heavy)	Non-road vehicles (heavy goods)	Maritime (small)	Maritime (large)	Aviation
TiFe	Storage capacity available today	●	●	●	●	●	●/●
	Storage capacity available 2040	●	●	●	●	●	●/-
	Temperature level	●	●	●	●	●	●
	Filling time	●	●	-	-	○	●
	Volume	●	●	●	●	●	●
	Weight	●	●	○	○	●	●
LiBH₄	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	○	○	○	●/○	○	○/-
	Temperature level	●	●	●	●	●	○
	Filling time	●	●	-	-	○	●
	Volume	●	●	●	●	●	●
	Weight	●	●	●	●	●	●
NaAlH₄	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	○	○	○	●/○	○	○/-
	Temperature level	●	●	●	●	●	●
	Filling time	●	●	-	-	○	●
	Volume	○	○	○	○	●	○
	Weight	●	●	○	○	●	●
TiH₂	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	○	○	○	●/○	○	○/-
	Temperature level	●	●	●	●	●	○
	Filling time	●	●	-	-	○	●
	Volume	○	●	●	●	●	●
	Weight	●	●	○	○	●	●
MgH₂	Storage capacity available today	●	●	●	○/●	●	●
	Storage capacity available 2040	●	●	●	●	●	●/-
	Temperature level	●	●	●	●	●	●
	Filling time	●	●	-	-	○	●
	Volume	○	○	○	○	●	○
	Weight	●	○	○	○	●	○
6Mg(NH₂)₂·9LiH·LiBH₄	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	○	○	○	●/○	○	○/-
	Temperature level	●	●	●	●	●	●
	Filling time	●	●	-	-	○	●
	Volume	○	○	○	○	●	○
	Weight	●	○	○	○	●	○
Liquid hydrogen	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	●	●	●	●	●	●
	Filling time	○	○	-	-	○	○
	Volume	○	○	○	○	●	○
	Weight	●	○	○	○	●	○
Compressed hydrogen	Storage capacity available today	●	●	●	●	○	●/○
	Storage capacity available 2040	●	●	●	●	●	●/●
	Filling time	○	○	-	-	○	○
	Volume	○	●	●	●	●	●
	Weight	●	○	○	○	●	○
LOHC A	Storage capacity available today	●	●	●	○/●	●	●
	Storage capacity available 2040	●	●	●	●	●	●/-
	Temperature level	●	●	●	●	●	●
	Filling time	●	●	-	-	●	●
	Volume	○	○	○	○	●	○
Weight	●	○	○	○	●	○	
LOHC B	Storage capacity available today	●	●	●	○/●	●	●
	Storage capacity available 2040	●	●	●	●	●	●/-
	Temperature level	●	●	●	●	●	●
	Filling time	●	●	-	-	●	●
	Volume	○	○	○	○	●	○
Weight	●	○	○	○	●	○	
Ammonia	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	●	●	●	●	●	●
	Temperature level	●	●	●	●	●	●
	Filling time	○	○	-	-	○	○
	Volume	●	●	●	●	●	●
Weight	●	●	●	●	●	●	

7.1. Transportation sector

Before evaluating the suitability of the different metal hydrides (and further hydrogen storage technologies) for the defined use cases within the transportation sector, existing examples of the utilization of metal hydrides for energy storage are discussed. Here, only hydrogen storage will be considered and not any other possible application areas of metal hydride storage, such as hydrogen compressors [4].

7.1.1. Existing examples in transportation sector

The most popular example of metal hydride storage within the transportation sector is its use in German and Italian Navy submarines (Class U212A and 214). These are room temperature hydrides with an operating temperature range of 20 to 50 °C. This hydrogen storage is coupled with a fuel cell and a battery [4]. Metal hydrides possible for these applications are TiFe and LaNi₅. The low-temperature level enables waste heat usage of the fuel cell. The low gravimetric energy density is no problem because additionally, a second hydrogen tank with liquid hydrogen is used in these submarines [143].

Another example for the integration of metal hydrides for energy storage is railway transportation. The first experiments with metal hydrides rail-based transport were made in the late 1990s and early 2000s, but only so-called “mini-trains” that were able to carry 4 to 6 passengers were constructed [4]. More extensive use of metal hydrides in train transport has not yet taken place.

Even when the focus in research is less on road transport and more on other non-road vehicles, there are also examples of test vehicles for road traffic. In a pilot study, a small fuel cell vehicle was equipped with a low-temperature metal hydride. In particular, experiments were carried out with the discharge process of the hydrogen, whereby it was found that the energy required to maintain the temperature (40 °C) increases proportionally with increasing discharge rates [144]. Vehicles that operate as factory vehicles in industrial complexes, such as forklifts or baggage tugs at airports, come more into question. One example for a forklift has been developed and operates with a battery and a fuel cell. The hydrogen is stored in a combined pressure and metal hydride storage system. The used metal hydride is a titanium-zirconium alloy belonging to the group of low-temperature intermetallic hydrides [145]. Beyond that, however, there are no successfully developed land-based transportation systems using metal hydrides as hydrogen storage yet.

An example from shipping is a canal boat used for maintenance purposes converted from diesel to fuel cell operation [146]. A PEM fuel cell and a metal hydride storage based on a titanium-manganese alloy were used. The storage was held under a pressure of 20 bar, which results in a combined pressurized-metal hydride storage system. The storage capacity was 4 kg of hydrogen in 30 kg of metal hydride material. The absorption and desorption tests were carried out over two years. The final evaluation showed that operation with high cycle stability with only 5% loss of maximum capacity after 1000 cycles was possible. Charging the storage tank took 2.5 h and the range was between 100 and 700 km, depending on the travel speed [146].

7.1.2. Comparison of application parameters and evaluation of hydrogen storage technologies

The results of the storage technology evaluation for the defined transportation application cases, listed in Tables 3 and 4, and hydrogen storage options, listed in Tables 7 and 8, based on the defined evaluation system are shown in Table 11. Due to the very similar parameters of TiFe and LaNi₅, LaNi₅ is not mapped. TiFe was selected due to the higher market penetration [147]. In addition, the analysis of the data showed that the application cases of individual, truck and bus road traffic were nearly the same, which is why no differentiation was made here.

Table 11 shows that no evaluation could be made for filling time of the assessed non-road vehicles. This parameter varies too much between

different types of vehicles to summarize this in one evaluation point.

Metal hydrides storage systems are not suitable for road transport and aviation, because the weight is too high for all hydrides considered or because the temperature level is in some cases significantly higher than the existing and expected future ranges of the waste heat temperature level. Additionally, large-scale metal hydrides storages are not to be expected in the future excludes in particular larger aircrafts from potential usage. At the same time, from a purely technical perspective, compressed hydrogen storage technology already being used in road transport today, is the least advantageous or suitable option compared to the other hydrogen storage options considered here.

Of the metal hydrides options studied, FeTi is the one that performs best across all applications. Disadvantageous is the high weight, which, however, is less relevant for train and ship traffic and the filling time, which is too low for vehicles with high relevance of it (railway and road).

From a technical perspective metal hydrides are a possible alternative to the hydrogen storage options considered so far, especially for shipping and rail transport. Metal hydride materials which seem to be worthy of more detailed investigation in future research for hydrogen storage applications are FeTi and the considered RHC showing suitable properties for low to medium temperature levels.

The necessary temperature level for the considered LOHCs and ammonia likely prohibits utilization of waste heat in most cases. Additionally, the comparably low volumetric energy density of compressed hydrogen makes it less technically advantageous as the discussed metal hydrides for various transportation applications. However, in the case of liquid hydrogen showing good evaluation results, the cryogenic temperature maintenance, the necessary insulation of the tank and the resulting boil-off losses are challenges not addressed here are worth to be mentioned.

Thus, it could make technical sense to increasingly advance the utilization of metal hydrides. Provided, the costs for the respective storage systems are sufficiently low, it can be expected, that in the future metal hydride storage tanks will be available for transportation applications up to the medium scale.

7.2. Stationary sector

Analogously to the transportation application areas, below examples in which metal hydride storage systems are already being used in stationary applications are outlined in order to subsequently perform an evaluation of the suitability of the different metal hydrides (and further hydrogen storage technologies) according to the use case parameters defined in the previous section.

7.2.1. Existing examples in stationary sector

In addition to the possibility of using metal hydrides as hydrogen storage, there are alternative forms of use, particularly in the stationary sector, as compressors and heat storage, in which the hydrogen is used as a working gas [4]. Since metal hydrides as a hydrogen storage option are investigated here, no examples of the other two applications are given below.

In contrast to the transportation sector, where lower temperature ranges are used, the stationary sector also allows the use of metal hydrides with desorption temperatures at several hundred degrees Celsius.

One example is the use of metal hydrides in off-grid systems. On a research scale, a stand-alone grid was simulated in [120] using a solid oxide fuel cell (SOFC) and a MgH₂ metal hydride. While this demonstrated the viability of such a system, the need for an efficient thermal management has been highlighted. This is due to the high operating temperatures of the system, which were between 300 and 350 °C for the metal hydride and over 700 °C for the fuel cell [120]. A similar concept was carried out in [148] with a PEM fuel cell and a LaNi_{4.8}Al_{0.2} metal

Table 12
Rating matrix for different hydrogen storage options and stationary use cases.

Storage option / Application case	Domestic (houses)	Domestic (districts)	Industry (chemical)	Industry (energy-intensive)	Power system (seasonal)	Power system (short-time)	Off-grid systems
TiFe	Storage capacity available today	●	●	●	●	●/●	●/●
	Storage capacity available 2040	●	●	●	-	-	●/-
	Temperature level	●	●	-	●	-	●
	Volume	●	●	●	●	●	-
	Weight	○	●	●	●	●	●
	Heat usage	●	●	●	●	●	●
LiBH₄	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	●	●	●	-	-	●/-
	Temperature level	●	●	-	●	-	○
	Volume	●	●	●	●	●	-
	Weight	●	●	●	●	●	●
	Heat usage	●	●	●	●	●	●
NaAlH₄	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	●	●	●	-	-	●/-
	Temperature level	●	●	-	●	-	●
	Volume	○	●	●	●	●	●/○
	Weight	○	●	●	●	●	●
	Heat usage	●	●	●	●	●	●
TiH₂	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	●	●	●	-	-	●/-
	Temperature level	●	●	-	●	-	○
	Volume	○	●	●	●	●	●/●
	Weight	○	●	●	●	●	●
	Heat usage	●	●	●	●	●	●
MgH₂	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	●	●	●	-	-	●/-
	Temperature level	●	●	-	●	-	●
	Volume	○	●	●	●	●	●/○
	Weight	○	●	●	●	●	●
	Heat usage	●	●	●	●	●	●
6Mg(NH₂)₂·9LiH·LiBH₄	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	●	●	●	-	-	●/-
	Temperature level	●	●	-	●	-	●
	Volume	○	●	●	●	●	●/○
	Weight	○	●	●	●	●	●
	Heat usage	●	●	●	●	●	●
Liquid Hydrogen	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	●	●	●	●	●	●
	Volume	○	●	●	●	●	●/○
	Weight	○	●	●	●	●	●
	Heat usage	●	●	●	●	●	●
Compressed Hydrogen	Storage capacity available today	●	●	●	○	○	●/○
	Storage capacity available 2040	●	●	●	●	●	●/●
	Volume	○	●	●	●	●	●/●
	Weight	○	●	●	●	●	●
	Heat usage	●	●	●	●	●	●
LOHC A	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	●	●	●	-	-	●/-
	Temperature level	●	●	-	●	-	●
	Volume	○	●	●	●	●	●/○
	Weight	○	●	●	●	●	●
LOHC B	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	●	●	●	-	-	●/-
	Temperature level	●	●	-	●	-	●
	Volume	○	●	●	●	●	●/○
	Weight	○	●	●	●	●	●
Ammonia	Storage capacity available today	●	●	●	●	●	●
	Storage capacity available 2040	●	●	●	●	●	●
	Temperature level	●	●	-	●	-	●
	Volume	●	●	●	●	●	-
	Weight	●	●	●	●	●	●
Heat usage	●	●	●	●	●	●	

hydride operating both at temperatures of $<100\text{ }^{\circ}\text{C}$ [148]. In addition, combinations with the supply of renewable electricity from photovoltaic systems, for example, are conceivable [149]. This was implemented, for example, in a demonstration building in which electricity was generated via photovoltaic (PV) systems, which was then converted into hydrogen. Subsequently, electricity could be reconverted via fuel cells if no electricity from PV systems was available. It was possible to store 120 kg of hydrogen [150]. A similar concept was implemented, in which 12 hotel rooms were supplied with electricity both from a photovoltaic system and via stored and subsequently reconverted hydrogen. The aim was to ensure operation throughout the year using renewable energies [151]. There is also the possibility of metal hydride storage usage for compensating fluctuations of renewable energies within a smart grid. Furthermore, simulations have been carried out for the stationary operation of high temperature metal hydride materials like NaAlH_4 or MgH_2 [152].

7.2.2. Comparison of application parameters and evaluation of hydrogen storage technologies

Analogously to Section 7, the results of the combination of different hydrogen storage concepts for stationary applications are compared and evaluated by combining the data from Tables 6, 7 and 8 based on the defined rating scheme. The results of the evaluation are shown in Table 12.

Typically, volume and weight limitations are of secondary importance for stationary applications. A significant influencing factor is the possibility of using waste heat and the usable temperature level. In addition, the area of the chemical industry is very broad; i.e., specific information on occurring temperature levels are not representative. While waste heat sources are available for all stationary use cases considered here (e.g., fuel cells or process heat), no seasonally available waste heat is applicable for the stationary case of seasonal storage because there is usually no direct coupling between the storage and the use of the stored or exhausted hydrogen, if a downstream use of the waste heat from, e.g., a fuel cell is not possible locally. For off-grid systems the volume relevance is varying, because of very different requirements in very different system. This takes into account the wide range of possible off-grid applications. In addition, higher temperature levels in stationary applications pose fewer challenges than in transportation applications.

From the above results it follows that metal hydride storage can, from a technical perspective, be considered for a wide range of stationary storage requirements. However, there may be a limitation in the available storage capacity. While there are currently only small- to medium-scale storage concepts for metal hydride storage, large-scale storage for liquid hydrogen is already available and cavern storage for pressurized hydrogen is planned to be implemented in the medium term [5,147].

8. Final considerations

The objective of this paper is to identify most suitable use cases for different hydrogen storage options. The main focus is on the metal hydride hydrogen storage technology, which is examined and evaluated regarding technical characteristics and requirements for transportation as well as for stationary use cases. The main results can be summarized as follows.

- The use of hydrogen storage tanks based on metal hydrides is unrealistic in road traffic and aviation due to the low achievable gravimetric energy densities and long filling time. Metal hydrides with higher gravimetric energy densities need in general desorption temperature levels that are too high for most transportation applications.
- A use of metal hydride hydrogen storage tanks in rail and ship transportation is conceivable due to the lower requirements for the

gravimetric energy density. In the context of larger ships, the use of high-temperature hydrides might also be a valid option.

- Land-based vehicles not being road-bound could benefit from the high volumetric energy density of metal hydride storage systems, since the gravimetric energy density is usually of secondary relevance for most of these vehicles.
- A truly promising metal hydride for transportation applications is TiFe , mainly because of low operating temperatures and sufficient available storage capacity.
- Since use cases within the transportation sector essentially involve small to medium-scale storage applications, it seems to be possible that by 2040 storage sizes could be available for all transportation applications under consideration.
- Due to the lower relevance of gravimetric energy density in stationary systems, the focus is on the usable temperature level and existing storage capacity today and in the midterm future.
- Metal hydrides with high volumetric energy density and low to medium temperature levels are of particular interest for small-scale systems (e.g., home storage systems). The metal hydrides FeTi , NaAlH_4 and the RHC ($6\text{Mg}(\text{NH}_2)_2\text{-}9\text{LiH-LiBH}_4$) appear to be promising candidates in this regard.
- Metal hydrides operating at higher temperatures could be a solution for large-scale industrial applications; one hurdle here could be the necessary increase in available storage system capacity, which is not available today and is most likely not expected by 2040. This is a disadvantage, especially in relation to liquid hydrogen and ammonia as a competing storage option technically much more advanced for the time being.

Further research is needed to develop specific concepts for the integration of metal hydride storage systems in the aforementioned applications. The focus should be on the dynamic behavior of a specific storage system to be developed, especially with regard to charging and discharging processes, in order to verify its real usability in the application cases which were under consideration here.

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Chris Drawer: Conceptualization, Formal analysis, Investigation, Methodology, Writing – original draft. **Jelto Lange:** Conceptualization, Methodology, Supervision, Writing – review & editing. **Martin Kaltschmitt:** Methodology, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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