

Hydrogen Storage for Automotive Applications: A Comparison

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Abstract

To abide by the Paris Agreement's goal of a maximum of a 1.5 °C increase in temperature within the world, there needs to be a greater transition towards sustainable energy with hydrogen, coupled with renewable energy sources such as solar and wind. Creating a carbon-neutral society requires the transportation sector, the largest source of greenhouse gas emissions in the United States, to transition to more environmentally friendly vehicles that use hydrogen fuel cells. The storage and transportation of hydrogen for hydrogen-powered vehicles requires great efficiency, a smaller environmental impact in manufacturing, and the right materials to prevent embrittlement and combustion. Although physical-based hydrogen storage is most developed, material-based hydrogen storage, such as nanomaterials and metal hydrides, present promise in the absorption and desorption of hydrogen. A comparison between physical (compression, cryogenic) and material (metal hydrides, LOHC, carbon-based) storage will be investigated to help determine what storage type fits different vehicles, such as sedans and buses. This investigation will bridge the gap between the hydrogen storage technology available as of 2023 and the feasibility of these different methods in powering various vehicles efficiently to help create a carbon-neutral society.

Introduction

99% of automobiles are powered by automobiles and diesel₁. On average, gasoline passenger vehicles emit around three-hundred fifty grams of carbon dioxide per mile driven, only 150 grams more than electric vehicles, assuming its power comes from fossil fuel based energy generation₂. Although electric vehicles are more climate-friendly, the emissions created from power plants (running on fossil fuels) that power EV batteries pose a problem to climate goals for 2050, with transportation being the leading sector for carbon emissions₃. Hydrogen fuel-cell powered vehicles represent an even better alternative to gasoline vehicles as the only emissions is water vapor, rather than carbon dioxide.

The worsening of climate change has prompted alternative fuels to be explored for a variety of uses. Although hydrogen has an energy yield of 122 kJ/g, larger than many hydrocarbon fuels, its low density has led to low power outputs when used in engines. Its poor efficiency has made it less attractive to customers, along with its high maintenance $cost_4$. Although the cost, and lack of availability of fuel, may deter the public from buying cars, the safety of the fuel does not factor into their decision to buy a hydrogen fuel cell vehicle. Hydrogen, as a fuel, is relatively safe. Because of its high diffusivity (or spreadability), its risk of combustion is small and the difficult process of its creation makes it more likely for a quick burning fire with little heat radiation₄.

Hydrogen production

Hydrogen, as one of the most prevalent elements on earth, comes from a variety of compounds. Three of the main types of hydrogen production are black-brown / gray hydrogen, blue hydrogen, and green hydrogen₅. These terms are defined based on their production source and environmental impact: black-brown hydrogen from natural gas, blue from carbon capture and storage, and green from clean energy sources. The most abundant hydrogen production



methods on Earth include the use of natural gas, which emits carbon dioxide in the air, unless it is captured . The use of natural gas leads to black-brown and gray hydrogen production, as shown in Table 1. Steam and natural gas are heated at a high temperature of 900 °C and a high pressure. With a nickel-based catalyst, the gasses create carbon monoxide and H2, which is then treated in a water-gas shift reaction to produce more hydrogen and carbon dioxide₅, as shown in Figure 1.

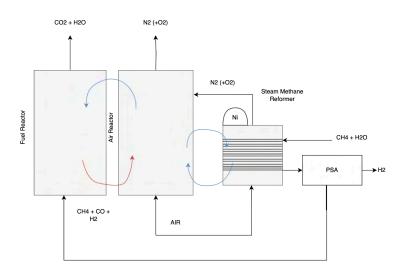


Figure 1. A steam methane reformer with a nickel catalyst producing black/brown hydrogen. With the help of a nickel catalyst, methane reacts with steam to produce hydrogen, carbon monoxide, and a small amount of carbon dioxide. This is an endothermic reaction, requiring heat to produce hydrogen. This steam is a substance made out of air (with nitrogen and oxygen)

Blue hydrogen is created by using Carbon Capture and Storage (CCS), which although produced with natural gas, minimizes the effect on the climate through the capturing of carbon (Table 1)₅. The carbon dioxide produced in black-brown / gray hydrogen is captured and stored. The storage of carbon is highly energy intensive. The energy needed to capture the carbon is also reduced by coupling endothermic and exothermic reactions. A liquid is used to chemically remove the carbon dioxide before it goes out into the smokestack. As shown in Figure 2, This captured CO2 is compressed until it becomes almost a liquid. It's then transported to a storage site, usually through a pipeline₆.

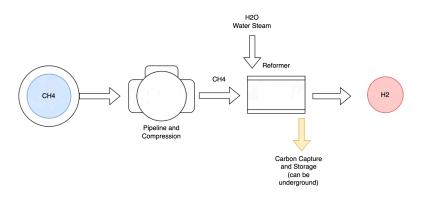




Figure 2. Blue hydrogen production process with methane. After pipeline and compression, methane and water are injected into a steam methane reformer to be separated into carbon dioxide and hydrogen.

The most climate effective hydrogen is green hydrogen, hydrogen produced from clean energy sources. Green hydrogen is produced through water electrolysis powered by clean renewable energy, such as solar and wind. Electrolysis occurs through alkaline (AWE), proton-exchange membrane (PEM), and solid-oxide steam, shown in Figure 3. Each electrolyzer uses a slightly different process, but they all have the relative same process. With an electrode stack separated by a membrane, high voltages and currents are replied to create an electric current in the water. The water thus breaks down into hydrogen and oxygen (Fig. 3). Hydrogen's production is difficult because of this current emphasis on clean production of hydrogen. Sustainable energy sources are inconsistent due to weather patterns, unlike fossil fuels. However, current electrolyzers are increasingly efficient and are declining in price, leading to a potential commercial feasibility₅.

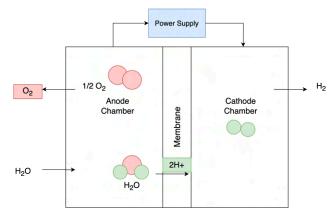


Figure 3. Electrolysis of water producing hydrogen. At the anode, water reacts to form oxygen and positively charged hydrogen ions. Electrons flow through an external circuit and the ions selectively move across to the cathode. At the cathode, the hydrogen ions/protons combine with electrons from the circuit to form hydrogen gas.

Table 1. Classification of hydrogen per generation method with strengths and weaknesses: Black, Brown, Gray, Blue and Green. Information taken from Noyan et al. 2023₅

Hydrogen Production	Strengths	Weaknesses
Black-brown / Gray hydrogen: produced through natural gas	 Common and mature technology (95% of hydrogen production) Cheap H2 concentrations > 65 mol% CO2 conversion rates > 80% 	 Endothermic reaction that requires a huge amount of heat supply Generates heat to produce extra steam Complex extraction of H2 with other outcomes that harm the environment (CO,



		CO2, CH4) • 2% of global annual CO2 emissions (by 2020)
Blue hydrogen: produced through natural gas with carbon capture and storage	 Minimizes Carbon emissions Cheaper than green hydrogen Systems with CCS would prioritize carbon-free fuels above electrification CO2 emissions decreased by 106% 	 Energy intensive More environmentally friendly options
Green hydrogen: produced through electrolysis through renewable energy sources	 Most effective in contributing to climate action goals Includes different electrolyzer options depending on the situation 4% of production 81% efficiency 	 Current prices are high (but will continue to fall as RE energy prices fall, \$0.70 - \$1.60 by 2050)

Hydrogen Storage

However, its main challenges remain in effective storage. Current storage methods are separated into two categories: physical and material. Physical based hydrogen storage relies on high pressure tanks or cryogenic tanks to create liquid hydrogen₇. The differences between the two is shown in Figure 4.



Physical-based storage (Type IV, single tank)	Material-based storage
700 bar \$15 ⁵ (\$500)	^b COST: \$/kWh (\$/kg H ₂) MH: \$43 (\$1430) S: \$15 (\$490) CH: \$17 (\$550)
Volumetric Density: 0.8 kWh/L (0.024 kgH ₂ /L system) Gravimetric density: 1.4 kWh/kg system (0.042 kgH ₂ /kg system)	Volumetric Density: kWh/L system (kgH_/L system) MH: 0.4 (0.012) S: 0.7 (0.021) CH: 1.3 (0.040) Gravimetric Density: kWh/kg system (kgH_/kg) MH: 0.4 (0.012) S: 1.3 (0.038) CH: 1.5 (0.046)

^B Cost projections at 500,000 units per year and reported in 2007\$ MH = metal hydride S = sorbet

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CH = Chemical hydrogen
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Figure 4. a side by side comparison of the costs, volumetric density, and gravimetric density requirements for the two types of storage systems.

Physical Storage

There are currently four types of hydrogen storage tanks used for high-pressured gaseous hydrogen storage (Figure 5).

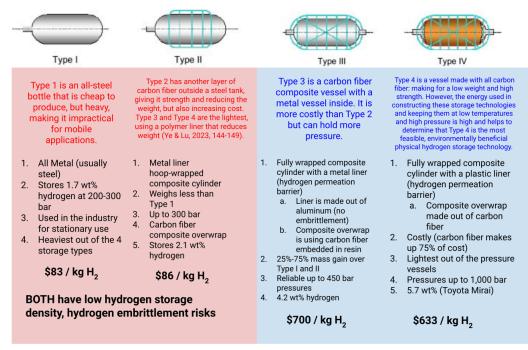


Figure 5. a side by side comparison of the four types of the compressed hydrogen storage, including their production materials, storage capabilities, and weight



Metal Hydrides

Metal hydrides represent possibilities in solid-state hydrogen storage. Through chemisorption (Figure 6), metal hydrides store hydrogen at a wide range of temperatures and pressures, and allow for high storage density per volume with low pressure. Lithium hydride, lithium aluminum, amine borane, and sodium borohydride are metal hydrides of interest₈.

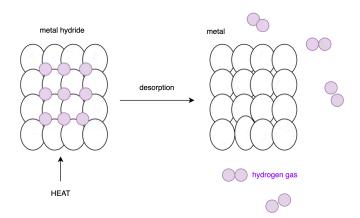


Figure 6. Metal hydrides store hydrogen very densely at low pressure, hydrogen is released by adding heat.

Nanomaterials

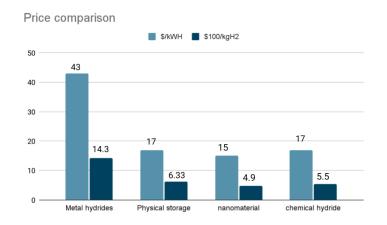
Carbon nanomaterial-based hydrogen storage also falls under the category of solid-state hydrogen storage₉. Porous nanomaterials have been explored to enhance hydrogen storage capabilities. These materials' sorption capacity, thermodynamic and cycling stability, and kinetics have been evaluated to review the material as an option for hydrogen storage₉.

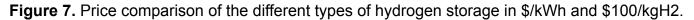
Liquid Hydrogen Carriers

Liquid organic hydrogen carriers offer low-cost storage options with minimal safety risk and high hydrogen storage capabilities for long periods of time₁₀. Liquid organic hydrogen carriers include chemicals such as ammonia, methanol, and formic acids. These chemical hydrides stay in a liquid state for a wide range of temperatures and already have established methods of transportation and storage. The technology of LOHCs is based on reversible hydrogen storage with release reactions₁₁. However, hydrogen combustion in an engine has negative impacts on the environment due to extra NOx emissions. Nitrogen oxide pollution is usually emitted by automobiles and other vehicles (construction equipment, boats). Damaging air quality, its strong oxidizing agent leads to reactions with volatile organic compounds that create smog₁₂.

Metal hydrides are currently \$43/kWH and \$1430/kgH2 while nanomaterials (sorbent storage systems) are \$15/kWH and \$490/kgH2. Chemical hydrides, or liquid organic carriers, sit in between at \$17/kwH and \$550/kgH2₁₃, as seen in Figure 7.







Long term, material based hydrogen storage presents greater possibilities in achieving DOE targets in volumetric and gravimetric capacity.

Applications of hydrogen storage

Compressed hydrogen storage systems have already been used in fuel cell vehicles. These vehicles are not mass produced due to its high cost and a lack of infrastructure for the widespread use of Hydrogen Fuel Cell Vehicles. Physical storage has not met the DOE₁₄ targets for onboard automobile storage and thus require significant improvement to compete against electric and gas vehicles.

Comparing physical based hydrogen storage and material based hydrogen storage for use in automotive applications will help in finding the best fit storage method for different automobiles. Based on past uses of hydrogen storage technology, the feasibility of certain storage methods for use in the transportation sector will be analyzed in terms of cost, efficiency, maintenance, and environmental impact to discover what storage methods are most realistic moving forward into a carbon-neutral society₁₅. This paper will first outline the methods used in finding data and how the data has been analyzed. The findings will be reported as results, with future implications discussed further in the next section.

Methodology

Due to the constant development of new hydrogen storage technology, only articles from 2015 onwards were considered. This includes problems such as scalability having been established as an issue and continues to be an issue in the research for more efficient, but more costly, technology.

To encompass greater amounts of information in searching, general searches were used in the start. These simple searches made finding storage standards, general opinions, and general information easy. From there, baseline information was established for more specific searches, including environmental effects, cost, maintenance, and vehicular use. Each search prompted hundreds of thousands to millions of results. The most recent articles were prioritized



while articles nearer to 2015 were used to discover general problems that continue to persist. Journal articles with tens of citations and diagrams were most used.

LIST OF RULES:

- 1. Article must be from 2015 and onward
- 2. Connection to the topic and date: For example, Schneemann's nanostructuring of metal hydrides for hydrogen storage
- 3. Must include diagrams regarding the topic (ex. structural diagrams of hydrogen fuel tanks)
- 4. Abstract must relate to current problems in the world regarding the change to a hydrogen society OR relate to current problems with the hydrogen storage option
- 5. Article must EITHER be specific to the topic searched or general about hydrogen overall
- 6. Quantitative data must be used to prove the article's point

The journal articles from more general searches were read in full while the articles taken from specific searches were used to find specific information regarding the search. Using the information compiled from the articles and information read, a table was created to track the type of article, publishing year, major approach, statistics, limitations, and results. Many articles described new processes conducted in labs, but are not yet ready for commercial use. This potential was considered but also realized to be a disadvantage for that storage system in the present.

In comparing storage systems, the most important determining factor in which was most appropriate for automotive applications was their comparison to DOE targets. Other considerations included current research and advances in reaching their targets, commercial feasibility, and whether they are fit for a car (need for a thermal management system, odd shape, heavy, etc.). Comparisons were made with traditional gasoline vehicles to understand consumer needs and how different hydrogen storage systems could fit within a traditional vehicle. These comparisons were also considered in whether storage systems were fit for automotive applications.

The costs of building hydrogen refueling stations were not considered in the costs for these storage systems. However, it was viewed as an advantage if infrastructure already existed and has been tested commercially for select hydrogen storage systems. Environmental impacts were considered but focused on the impact during the storage systems use and less on waste disposal and production. The most efficient technology was used to compare against DOE targets for the most fair comparison.

It's important to note that there are more variations of material-based storage in development. Only the most common were mentioned. This also applies to the comparison between current hydrogen fuel cell vehicles on the market. Only three were mentioned, but many other automotive companies have also produced these vehicles. This technology is continuously being researched and thus these points need to be considered overall, rather than just by the numbers.



Results and Discussion

Comparison to Gasoline-Powered Vehicles

Contrary to popular belief, gasoline vehicles operate with less efficiency than fuel cell systems. Gasolines operate with less than 20% efficiency in converting chemical energy in gas to power: only 12 to 30% of the energy from gasoline is used by a conventional vehicle to move it down the road₁₆. A hydrogen fuel cell system uses 60% of the fuel's energy: a \$50 reduction in fuel consumption compared to gasoline engines.

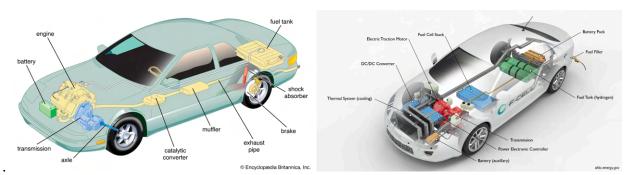


Figure 8. A side-by-side comparison of a gasoline vehicle (left) and a hydrogen fuel cell vehicle (right), showing the structural design similarities and differences between the two. As shown by the diagrams above, a hydrogen fuel cell vehicle needs a significant amount of space structurally to house all the technology it needs to function. Image on left from Encyclopedia Britannica (https://www.britannica.com/technology/automobile#/media/1/44957/120667). Image on the right from Alternative Fuels Data Center (Office of Energy Efficiency and Renewable Energy, https://afdc.energy.gov/vehicles/fuel_cell.html).

The placement of fuel tanks is similar for both hydrogen and gasoline powered vehicles, as seen in Figure 8. The cylindrical shape, as stated above, is inconvenient for vehicles. The unique molding of gasoline fuel tanks can make tanks fit without sacrificing space, while this still poses a problem for hydrogen vehicles. Environmentally, hydrogen vehicles exceed the climate standards in comparison to gasoline vehicles. Cost wise, gasoline vehicles are still cheaper.

Mechanically, hydrogen fuel cell cars are similar to electric cars, just with an on-board power generating device. These cars are simple, compared to gasoline cars and don't require the regular oil changes needed in the maintenance for a gasoline vehicle. Fuel cell cars use regenerative braking to help recharge the battery, so the wearing of the brake is less than on a traditional gas vehicle₁₇. Hydrogen fuel cells will have less maintenance costs, overall. Its storage tanks only add to its durability, with each method having failsafes or proven durability.

Costs can be lowered through the prevalence of the vehicle but also a reduction in the cost of hydrogen storage systems. Low pressure, adsorbent-based fuel systems can reduce cost of on-board tanks, yet research is still needed to look into materials with better adsorption properties (organic solvents are usually very expensive).



Physical-Based Storage

Description: Physical-based hydrogen storage is the most common method of storage in vehicles. It is composed of cryogenic hydrogen storage, compression based hydrogen storage, and cryo compressed hydrogen storage (see Table 6). Its developed technology makes it the easy choice for many automotive manufacturers. Paris's climate goals have led to technology needing to be developed and built rapidly in a growing world. People thus want to use existing industrial assets in the renewable world.

Table 2. DOE requirements for onboard hydrogen storage for automotive applications that are relevant to this paper. This table does not include the full list of DOE targets for onboard storage for light-duty automotive applications. These targets will be referenced to throughout this text. All data in this table from the Office of Energy Efficiency & Renewable Energy.

Storage Parameter	Units	2020	2025	Ultimate			
System Gravime	System Gravimetric Capacity						
Usable, specific energy from H ₂ (net useful energy/max system mass) ^b	kWh/kg (kg H ₂ / kg system)	1.5 (0.045)	1.8 (0.055)	2.2 (0.065)			
System Volumet	ric Capacity	1	1				
Usable Energy density from H ₂ (net useful energy/max system volume) ^b	kWh / L (kg H ₂ / L system)	1.0 (0.030)	1.3 (0.040)	1.7 (0.050)			
Storage System	Cost		<u>.</u>				
Storage system cost	\$ / kWh net (\$ / kg H ₂)	10 (333)	9 (300)	8 (266)			
Durability/Operability							
Operating ambient temperature ^d	°C	-40/60 (sun)	-40/60 (sun)	-40/60 (sun)			



Min/max delivery temperature	°C	-40/85	-40/85	-40/85	
Min delivery pressure from storage system	Bar (abs)	5	5	5	
Max delivery pressure from storage system	Bar (abs)	12	12	12	
Onboard efficiency ^e	%	90	90	90	
"Well" to power plant efficiency ^f	%	60	60	60	
Charging/Discharging Rates					
System fill time ⁹	min	3-5	3-5	3-5	

Transient response at operating temperature 10%-90% and 90% to 0% (based on full flow rate)	S	0.75	0.75	0.75	

Dormancy

Dormancy time target (minimum until first release from initial 95% usable capacity)	days	7	10	14
Boil-off loss target (max	%	10	10	10



reduction from initial 95% usable capacity after 30 days)					
Environmental Health and Safety					
Permeation and leakage ^h	-	Meet or exceed SAE J2579 for system safety			
Safety	-	Conduct and eval	uate failure analysi	S	

The high energy input and relatively heavy weight of physical based storage makes it a weaker alternative for future automobile applications when metal hydrides and MOFs, such as sodium borohydride and MOF-5, may be developed (based on current potential of developing technologies) to be. Low pressure liquid hydrogen systems, though available for transport, are too bulky for onboard automotive use₁₄. Compressed automobile hydrogen storage systems show the most promise between physical based storage options in being used in sedans due to its low amounts of energy usage, its commercial nature, and its longer dormancy state. This technology currently sits at around \$15/kWH and \$500/kgH2₁₄. Already proven to function in automobiles, future research and development into increasing storage capabilities will make this a safe, mature technology for transportation purposes.

Types of Compression-Based Hydrogen Storage: Physical-based hydrogen storage has five types. When considering the differences between the five in automotive applications, the factors to consider include the weight of the container, the environmental impact, the cost, and its storage capacity. The bulkiness and cost of Type 1 (though less expensive than Type 4) and 2 make them infeasible for use in automobiles ₇. Type 3 and 4 show the most promise due to its lightweight feel and cost, addressing many of the factors needed for storage systems in automotive applications. Type 4, however, is the most common type used in vehicles due to its lower environmental impact, its lower cost, and lighter material (no metal lining)₇. Type 4 also endures pressure up to 1000 bar, comparatively to Type 3's maximum pressure of 450 bar ₇. Hydrogen's volatility makes the greater withstanding of pressure valuable, preventing extra lead off that can occur from built up pressure. The majority (75%) of Type 4's cost is carbon-fiber₁₈. Type 4 still requires improvement in specific areas to reach DOE targets (Table 3).

Table 3. Type 4 hydrogen storage tank compared to DOE target (See Table 2). The current capabilities of this storage medium are compared to the requirements found in Table 2. An represents not meeting demands while a represents a meeting of demands

Type IV Storage



System Gravimetric Capacity (kwH / kg)	System Volumetric Capacity (kg/L)	Storage system cost (\$ / kg H2)	Durability/ Operability	Charging/ Discharging rates	Environme ntal Health and Safety	Dormancy
			(falls short by 5% for meeting Well to Tank efficiency target)			

This cost could be reduced with the development of new technology, the development of a Type 5 vessel for use in automobiles₁₈.

Type 5 vessels are the lightest of the five due to removal of a liner. Although the liner is light, it still weighs 8 kg. Currently, these vessels do not operate at a high enough pressure to store the adequate amount of hydrogen needed for non laboratory use₁₈. The further development of this technology could lead to a better alternative in comparison to the other four compressed hydrogen types. The removal of a liner also takes away the risk of liner blistering due to saturation and decompression₁₉. The safety concern is its inability to withstand a buildup of pressure which can lead to hydrogen leaking out₁₈.

Carbon fiber is corrosion resistant₁₈, leading to a greater lifeline in comparison to metal based storage methods. Its flexibility could prove to help support more innovative designs that account for the odd cylindrical shape in mobile applications.

Cryogenic Hydrogen Storage: Cryogenic hydrogen storage is more compact, due to its liquid state, compared to H2 in gaseous phase at ambient temperature. Hydrogen's boil off temperature is extremely low, at below $-253^{\circ}C_{18}$. This leads to high amounts of energy needed to turn hydrogen into a liquid state, encompassing between 25 to 40% of the energy content of hydrogen, losing a significantly greater amount of energy than in compressed hydrogen (which stays at 10%)₁₈. Safety concerns can also increase as time passes due to potential boil off losses if the vessel is confined for several days₁₈. Since hydrogen to become a gas again. This gas builds up over time, and for tanks that cannot hold the pressure, such as cryogenic tanks, this hydrogen must be let out through a fail safe mechanism. The fast process of vaporization and over-pressuring the storage tank, which is not built for storage, can damage the tank. The overall energy intensive process and thermal management needed for cryogenic hydrogen storage makes it inefficient and infeasible for use in vehicles₂₀. Environmentally, its high energy usage doesn't make it the best option in comparison to lower energy options. The high liquefaction cost itself, which does not meet DOE targets (Table 4), also makes it less appealing



to consumers. However, its kinetics are comparable to compressed storage and its compactness can help to increase gravimetric and volumetric density₂₀.

System Gravimetric Capacity (kwH / kg)	System Volumetric Capacity (kg/L)	Storage system cost (\$ / kg H2)	Durability/ Operability	Charging/ Discharging rates	Environme ntal Health and Safety	Dormancy
✓ 7.5 wt%	6.4 MJ/L = 1.78 kWh/L			comparable to compresse d hydrogen		

Table 4. a comparison of cryogenic storage vs. DOE targets (see Table 2).

Cryo Compressed hydrogen storage is still being further developed. This storage method combines both high pressure and cryogenic temperature vessels₂₁. This combination reduces safety risks, withstanding the pressure that can come from boiling off₂₁. The insulated vessel can extend the dormancy period while lower pressures can reduce the amount of carbon fiber used, reducing the cost. This vacuum structure makes it more safe than other physical-based storage methods₁₉. New research from Hosseini (et al.)₂₂ shows that this method can promise more storage density and safety than cryogenic storage, offering 80g/L volumetric density, meeting DOE targets for gravimetric and volumetric capacity $_{22}$, as shown in Table 5. Due to these factors, this storage method has shown greater promise than compressed systems, shown through computational analysis of FC buses₁₈.

Table 5. a comparison of cryo compressed hydrogen storage vs DOE targets (see Table 2)

System Gravimetric Capacity (kwH / kg)	System Volumetric Capacity (kg/L)	Storage system cost (\$ / kg H2)	Durability/ Operability	Charging/ Discharging rates	Environme ntal Health and Safety	Dormancy
5.4 wt%	80g/L = 0.08 kg//L					



Table 6. a summary	of the different	t types of physical	storage.	Includes pressure,	cost,
gravimetric density, mai	intenance, and in	nportant points to co	nsider		

Туре	Pressure (bar)	Cost (\$ / kg)	Gravimetric Density (wt%)	Maintenance	Points
I	300	83	1.7	Generally made out of steel, has a possibility of corrosion	Too heavy for automotive applications
11	200	86	2.1	Its metal liner may corrode	Heavy compared types III and IV
111	700	700	4.2	The carbon fiber overwrap is corrosion resistant, but the metal liner isn't	Heavier than Type 4
IV	700	633	5.7	Removal of a metal liner makes maintenance easier since carbon fiber is corrosion resistant	takes 4.1 wt% to compress hydrogen from 20 to 700 bar, another 1.6-3.6 wt% for pre-cooling of hydrogen to lessen the release of heat **no need for thermal management system
Cryogenic		167 (4300 kg)	100 L	To reduce	Applicable



		386 (100 L for automotive applications)		corrosion, the tank needs to be cleaned once a year	where high energy density is required and boil off isn't a big concern (e.g. commercial aircrafts), cost and energy to liquify is huge
Cryo Compressed	300 bar	390	5.4 (40 MG/L)	Needs to be cleaned once a year (partly cryogenic)	For mobile applications Moder ate temper ature (40-80 K) Greate r dorma ncy period

Material-Based Hydrogen Storage

The main material-based hydrogen storage methods include liquid organic hydrogen carriers (LOHCs) and metal frameworks. These vessels are still in development and will need extra management systems within a vehicle to make the technology feasible for use.

Liquid Organic Hydrogen Carriers (LOHCs): Liquid organic hydrogen carriers (LOHC) use existing technology and infrastructure to store and transport hydrogen (Figure 9).



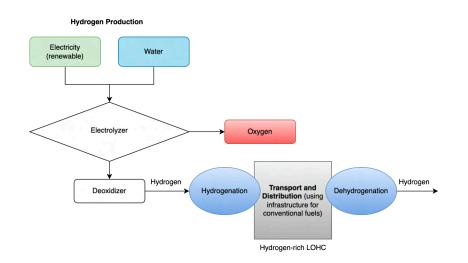


Figure 9. A simplified process for LOHC carriers from electrolysis to transport and distribution.

This makes refueling easy. In place of gas at refueling stations, these carriers would be quickly dispensed into cars₂₃. Despite the already developed technology for bulk transport, it may not be feasible for transporting hydrogen for immediate use in hydrogen-powered vehicles. To separate hydrogen from carriers like ammonia, extremely exothermic reactions are required₂₅. This leads to excess heat that isn't ideal for mobile applications. The separation of hydrogen also requires expensive catalysts that increase the cost of already expensive technology₂₄. Many metal-based catalysts cannot last long and involve critical platinum group metals, both limited and expensive. Its non reusability creates a costly maintenance endeavor.

LOHC's positives come from its ability to store large amounts of hydrogen in small volumes at ambient temperatures. LOHC technology focuses on direct use in vehicles combined with hydrogen combustion in internal combustion engines. LOHCs are thus focused more on stationary hydrogen and energy storage along with delivery of hydrogen to hydrogen fueling stations.

Although LOHCs may be fit for this type of role, Hynertech co. Ltd. has developed a fuel cell bus fueled by LOHC₂₅. LOHC has been popular for refueling ships and for use in heavy-duty-fuel-cell based trucks₂₅. Due to existing tank-ship technology, it can be adapted to distribute LOHC. Their lower volumetric density makes it more suitable for bigger vehicles with bigger tanks. Asian companies are interested in LOHCs and many of the major hydrogen fuel cell-vehicle producers are Asian companies, such as Toyota and Hyundai₂₅. There has been huge progress in the effectiveness of LOHC hydrogenation and dehydrogenation technology and progress in the concepts that deal with the heat generation and consumption needed for this technology. Its low pressure and ambient conditions could also provide the safety standards not yet realized with other storage methods. The progression has not been enough to reduce the energy for dehydrogenation and separation, adding to the environmental footprint in creating LOHCs. Other than needing improvements in scalability, carrier liquids have lower storage capacity and are not suited for the high demand of a hydrogen based society₁₉. The need for an additional thermal management system in addition to the storage tank for LOHCs will add extra cost and bulk to a compact vehicle.



Nevertheless, like many evolving hydrogen storage technologies, continuous research into catalysts have found cheaper alternatives. A catalyst, composed of nitrogen and carbon (metal-free), frees hydrogen, even at room temperature₂₆. The catalyst is able to take place at room temperature because of the structure nitrogen forms during the carbonization process₂₆. Further research into making this process more efficient will make the extraction of usable hydrogen from LOHC storage at a low cost and milder conditions than current technologies. The research's current focus on finding molecules that have a larger capacity will help to achieve DOE targets (Table 7) and thus provide power to vehicles over a greater distance.

Table 7 I OHC storage in com	parison to DOE targets (see Table 2)
Table 1. LOTIO Storage III con	parison to DOL targets (see Table 2)

System Gravimetric Capacity (kwH / kg)	System Volumetric Capacity (kg/L)	Storage system cost (\$ / kg H2)	Durability/ Operability	Charging/ Discharging rates	Environme ntal Health and Safety	Dormancy
2.06 (Perhydro Dibenzyltolu ene-Dibenz yltoluene)	0.77	137	<200 °C under 1 bar pressure >90% conversion possible, but 45% electrical efficiency with Solid Oxide Fuel Cell	Fast refueling time kinetics are lacking	Not classified as dangerous goods, BUT in need of better catalysts that don't emit harmful toxins	Meant for long hauls of transportati on

Metal Organic Framework (MOF) Storage, Sorbents: Metal Organic Frameworks (MOFs) have increasingly shown promise with the progression of new research and findings. Research has focused on ways to optimize the release of hydrogen through the increase of surface area or changes in temperature. MOFs are sorbents. Sorbents store hydrogen through physisorption₂₇. MOFs have the opposite flaw of metal hydrides: they have rapid adsorption and desorption rates (strong reversibility within seconds) but low capacity except at cryogenic temperatures₂₀, as shown in Table 9. Sorbents tend to bind hydrogen too weakly₂₇. Nevertheless, solid porous material and carbon nano-tubes are in early stages of development (Figure 10). They face material, volume, and weight problems that make it unrealistic for use in automobiles this present day₂₈. Climate change's imminent effects create an urgency that makes counting on new technology risky. In addition, new technology is expensive and hasn't been tested enough to gain consumers' trust, who are generally risk averse.



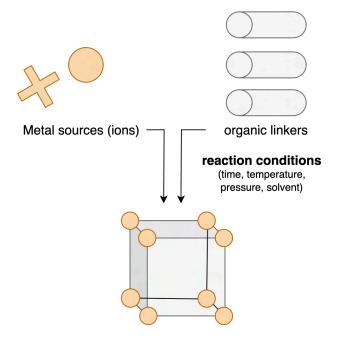


Figure 10. A simplified process to create metal organic frameworks. Through reactions involving solvents and specific temperatures and pressures, metal ions and organic linkers create metal hydrides.

Well known MOFs include UiO-66, MOF-5, and MOF-177, with recent advances includes a series of Pd-doped MIL-101 samples that affect hydrogen storage performances₂₉. MOF-5's main metal is $Zn4O_{30}$. Zinc is rarely found in sufficient amounts to be extracted economically, which makes bulk consumption and production hard. Palladium, which can help the storage method perform better, is also very rare and the most expensive out of platinum, and silver, with the exception of gold (Table 8)₃₁.

Table 8. a comparison of the costs between four different metals according to \$ / Troy ounce.

	Palladium	Platinum	Gold	Silver
Cost (\$/Troy Ounce)	959.50	917.50	2,018.71	22.80

These rare catalysts and the mining needed for them will not only endanger laborers but also impact the environment negatively₂₇.

Table 9. MOF (sorbents) storage in comparison with the DOE targets shown in Table 2



System Gravimetric Capacity (kwH / kg)	System Volumetric Capacity (kg/L)	Storage system cost (\$ / kg H2)	Durability/ Operability	Charging/ Discharging rates	Environme ntal Health and Safety	Dormancy

Metal Hydrides: Metal hydrides bind hydrogen chemically through van der Waal interactions₃₂. This leads to a meeting of DOE gravimetric and volumetric targets, but not the charging and discharging rates, capacity, and minimum delivery pressure. Metal hydrides bind hydrogen very strongly, making it a necessity to find better ways to facilitate the movement of hydrogen₂₇. Its limited reversibility slows hydrogen uptake and release kinetics (Table 11). Different metal hydride structures, such as nanostructuring metal hydrides, can help improve the lacking thermodynamics and kinetics₂₇. These nanostructures are shown in Figure 11.

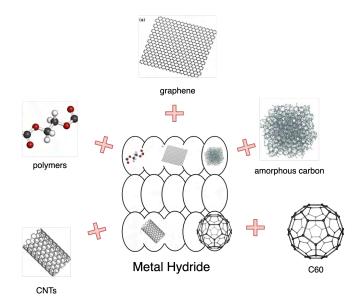


Figure 11. five different ways to nanostructure metal hydrides are shown above. These nanostructures are used to improve kinetics and thermodynamics.

Nanostructuring metal hydrides have proven to decrease the reaction energy needed for dehydrogenation by altering the hydrogenation and dehydrogenation reaction pathways. More research is needed to see how nanoscale and nano scaffolding affects metal hydrides, such as its effect on thermodynamics or on storage capacity. The most promising MOFs are high surface



area materials with metal cations or clusters connected through bridging organic ligands₃₃. Cations are included to help with a complete dehydrogenation of the hydride and help to bind the anions, occupying the pores of the structure₃₃. Hydrogenation can affect the volume, which can cause mechanical stress on a system, especially a system in a compact space like a car₂₇. This mechanical stress can then affect kinetics and thermodynamics, impacting reaction pathways that add to previous problems. Mechanical stresses are increased with its strong exothermic and endothermic reactions where heat cannot be reused. These high temperatures can lead to energy loss and inefficiency, meaning it does not meet DOE operability standards (Table 10). A need to keep heat trapped leads to bulky insulation in a compact vehicle. Metal's weight also adds enough problems in transportation, and its high reactivity makes safety a possible concern₂₈. The greatest concern deals with metal's high reactivity and volatility. One of the most promising metal hydrides includes the use of magnesium. This metal has an extremely high reactivity, especially to halogens, but the greater risk is when magnesium is exposed to the air. When magnesium reacts with oxygen, for example, the reaction is so bright and releases significant amounts of heat₃₄. This creates a possibility for dangerous explosions. However, the storage method's durability adds a layer of safety. Hydrogen can only be released when heat is added, so even if the container is damaged, the hydrogen will be contained₃₅. Hydrogen being chemically bonded prevents the hydrogen from leaking out, like in physical based storage options. Though greater progress could make metal hydrides more feasible in comparison to physical options, its inability to scale up production due to raw materials needed in its production could also prove to be a barrier in a more sustainable world₂₀.

Table 10. A c	comparison o	of hydrogen	chemically	absorbed vs.	DOE targets
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System Gravimetric Capacity (kwH / kg)	System Volumetric Capacity (kg/L)	Storage system cost (\$ / kg H2)	Durability/O perability	Charging/Di scharging rates	Environme ntal Health and Safety	Dormancy

Table 11. a ranking of metal hydrides, LOHCs, and sorbents in terms of hydrogenation, dehydrogenation, and refueling time. Rankings are 1 (easiest, fastest) to 3 (hardest, slowest)

	Metal Hydrides	LOHCs	Sorbents
Hydrogenation	3	2	1



Dehydrogenation	3	2	1
Refueling time	3	2	1

Kubas Type Hydrogen Storage: Newer technology on the market with potential to be used in automotive capacities include Kubas-Type Hydrogen Storage₂₀. This interaction is a low strength chemical bond (easier reversibility) occurring with transition metals (chemisorption). This means that there is no longer a need for high energy input to split bonds. Adsorption is triggered by pressure variation. This pressure isn't as high as 700 bar as in physical models. There is no need for a temperature management system (works at room temperature) and even has a potential to exceed DOE targets in gravimetric and volumetric density.

Current Models

Current Hydrogen Fuel Cell Vehicle Models: The most promising models of hydrogen-fuel powered passenger vehicles currently include the Toyota Mirai, the Hyundai NEXO, and the future BMW iX5 (Figure 12). BMW and Toyota have had a hydrogen-fuel powered vehicle on the market for over a decade, but improvements in efficiency of storage and the fuel cell have increased the mileage, getting closer to DOE targets. The BMW iX5 is a concept vehicle that will supply a 300 mile driving range with two hydrogen tanks with the combined capacity of 6 kg₃₇. The Toyota Mirai, comparatively, is estimated for a 402 mile range (Mirai XLE) and 357 mile range (Mirai Limited)₃₈. The Hyundai NEXO is a relatively newer vehicle on the market with an estimated 380 mile range, and became the first hydrogen-powered SUV₃₈. This vehicle reshapes hydrogen vehicles by incorporating three identical 13.7 gallon tanks instead of two, like the Toyota Mirai. All three use cylindrical shaped tanks, common with physical-based storage of hydrogen. Cylinders are a difficult shape to use for fuel storage, due to the traditional placement of storage of fuel tanks in cars and physical-based hydrogen storage is still developing technology, which doesn't meet DOE targets yet₂₀. The unlikelihood of fulfilling the 300-mile driving range that consumers would like to see in passenger vehicles with compression without compromising traditional free space in cars is still a work in progress. However, Hyundai's NEXO solves this problem with the use of three identical tanks, freeing up more space behind the rear seat₃₀. Storage tanks are usually found in the rear, and the design of three smaller tanks makes it easier to configure in other places (two under seat, one under cargo floor) other than just two big tanks in the back of the car.

Hydrogen vehicles are currently the most widely available in California, due to the access to hydrogen fueling stations in the area₄₀. There are a limited number of test vehicles available from specific organizations with access to hydrogen fueling stations₄₀. Hydrogen buses and tractors are in development and in testing. Its development and feasibility will rely on the number of fueling stations, not those just supporting light duty vehicles, that can be built.



There were only 59 fueling stations in the US in 2023, most of which were located in California₄₀. There are other private stations to support cars but mostly for development and research. The number of these stations is set to increase as the demand for hydrogen vehicles expands.



Figure 12. A comparison between three current hydrogen fuel cell vehicle models

Compression based storage is the most mature technology, making it the easy choice for automobile producers, who rely on customer happiness to succeed. The greatest challenge with getting newer technologies into the market is the development of a framework with a low manufacturing cost. Metal hydride material production at a low cost is a huge barrier in its competitiveness with the gas separation industry₄₁. Production costs are dependent on the cost of its inputs, target profits, manufacturing costs, quality control, and other related costs. For metal hydrides, manufacturing costs depend significantly on the energy demanded in its production, its multi-step process to produce the desired results, and the expensive catalysts needed to improve storage capabilities₂₄. There is a great measure of difficulty in scaling such a delicate process up to commercial scale and making it affordable to all.

Material costs of MOFs in comparison to physical based hydrogen storage: The material costs of metal hydrides are dependent on four parts: solvents, metal sources, organic ligands, and acid/base catalyst ₄₁.

Table 12. The table below shows the percentage breakdowns of the costs of the four main components in creating an MOF.

	Catalyst	Solvent	Linker	Metal
MOF-5	-	78.6%	6.4%	15.0%
MOF-74	-	1%	77.8%	21.2%
Uio-66	0.5%	71.1%	4.3%	24.1%



For example, to make MOF-5, you need: 81.30 L of DMF (\$414), 1.03 kg of terephthalic acid (\$34), 3.45 kg of zinc acetate dihydrate (\$79)₄₂. This leads to a total cost of \$527. Because solvents cannot be recycled, this is the cost for every new hydride. The solvent makes up the majority of the cost in the production of MOF-5 (Table 12). MOF synthesis usually involves a reaction of hydrated metal salts with bridging organic ligands in expensive organic solvents between $100^{\circ}C$ and $150^{\circ}C_{32}$. These accumulated costs make it hard to scale-up the production of these components, jacking up their market price for commercial suppliers₄₂. This low economic feasibility continues to turn developers away from this technology in the present.

There is potential for the production of MOFs to be produced for less than \$10/kg by reducing solvent usage with alternative synthesis processes, such as aqueous synthesis₃₂.

Although these vehicles, and most vehicles on the hydrogen vehicle market, consistently use physical-based hydrogen storage, more promise can be shown with other developing technologies. The cost and extra needs of specific storage methods may sway developers back to simply improving physical-based technology, proved to be effective already.

Discussion

The environmental burdens of industrial development for hydrogen are significant. There is not an adequate amount of big supply chains for clean hydrogen storage and transportation, along with a lack of, more uncommon, storage and transportation infrastructure. The use of existing technology plays a huge role in deciding the path to pursue in a hydrogen energy world. The technology for liquid organic hydrogen carriers and compression-based hydrogen is prevalent, leading to a bias being presented towards them. It is, however, necessary to consider the future potential of technologies to determine their potential in mitigating climate change and consumers' well-being.

Tank Technology: It is very important to consider redesigning tanks and elements to make market-available products more effective and to allow sufficient time for the development of more promising technologies for future use. LOHC and physical-based storage methods, particularly Type 4 and 5 tanks, present the most feasible storage methods.

- These storage methods currently have the most existing infrastructure available, making the transition into a hydrogen society easier.
- These storage methods have already been proven to work and need to be adjusted better to automotive applications.

In comparison, metal hydrides and MOFs still require research to reach the development stages LOHC and compressed hydrogen storage methods have already surpassed. LOHC has a greater environmental impact since it involves compounds that generally contain carbon and whose production can harm the environment. Certain compounds, such as benzene, are too toxic while others are too volatile for practical relevance.

Mobile Applications: Both storage types have shown to be feasible in mobile applications. It could be possible that LOHCs are better fitted for larger vehicles, such as trucks, with larger spaces for storage tanks. Construction sites generally have large-stationary diesel tanks, and hydrogen fuel (in the form of LOHC) could be an easy transition. If engineered correctly,



diesel-powered construction equipment, housing huge tanks, can be powered by hydrogen fuel. There are certain sites far from urban areas which could make it difficult to use hydrogen fuel for construction equipment due to a lack of charging sites, but for those near urban areas, hydrogen fuel is presenting as a good alternative especially due to the large tank size and already-heavy weight of construction equipment, which is not expected to be light.

The lack of charging stations makes it difficult for hydrogen fuel, and other electricity-based options, to be used in remote areas. There is an added layer of difficulty with hydrogen cars needing to be charged more often than gasoline vehicles being filled up. It may also be more feasible to use the existing infrastructure for LOHC storage for bulk transport to fueling stations and focus on other technologies that may be more environmentally protective for sedans and passenger cars. LOHC compounds are commonly transported for a variety of reasons, but the main source of emissions with LOHC is dehydrogenation. Economically, LOHC is most reasonable, especially with promising developments with new catalysts.

Future decision factors:

Environmental Impacts: While moving forward, it is important to conduct hazard assessments in early research stages to address the end of the technology life cycle (the environmental problems after its use) to predict the hazards each chemical used could cause. Many catalysts currently used for dehydrogenation processes lead to emissions and energy usage from the storage system in order to operate. The disposability of these chemicals and lifelines of catalysts must be considered when constructing storage systems. Chemical waste has specific guidelines and not following them can disrupt ecosystems and humans' own way of life.

Safety: When determining the right choice for smaller vehicles, one of the most important factors to consider is safety. The main concern is hydrogen boil-off and possible combustion from a hydrogen leak. Compression based storage methods account for this, but as hydrogen becomes more mainstream, it is important to figure out regular maintenance struggles to ensure efficient operation. Current technologies, both material and physical, are relatively safe; minimizing accidents will come from better efficiency in holding hydrogen and better structural integrity.

Rare Materials: The most effective catalysts are made up of rare materials, such as rhodium and palladium. These materials are both expensive and require replacements after they reach their maximum number of cycles (dehydrogenation, rehydrogenation). In a hydrogen-based society, competition over rare materials and costs will be difficult.

Economics: The cost factor is always important to consider, as a consumer and producer. Current climate goals will require everyone to take action, meaning sustainable technology needs to be affordable. Building new infrastructure, and investing in R&D takes time and money, in addition to other pressing matters. In this way, it is important to figure out how to optimize production and use existing infrastructure to ease the transition. A total and sudden turnover to a hydrogen society will be extremely expensive (costs can be spread out in the long run) and hard to adjust to in a short time.

Technology Development: Metal hydrides are still in need of major development until they reach commercial use and thus cannot be considered as a viable alternative at this moment. Although its potential fulfilling gravimetric and volumetric density are its main advantages, its



expensive materials, expensive synthesis process, and difficult scalability do not make it a great choice in the present. Many of the processes needed to chemically separate bonds are also highly endothermic and exothermic. For automotive applications, bulky insulation and a thermal management system are needed to manage high-temperature reactions. Cryo-compressed hydrogen storage also falls into this category. Its potential will fulfill most, and maybe all, DOE targets, but its cost and current development don't make it the right choice for the present.

Overall compressed hydrogen storage is still the most viable option at this moment. It has the most developed technology, current hydrogen FCEVs use compressed storage, and compressed gas tanks are a more common technology. There is a huge potential for cryo-compressed hydrogen storage once technology fully develops will help to develop a more efficient hydrogen economy, but existing infrastructure and the already commercial realization of compressed hydrogen storage make it the right choice to step off into fulfilling Paris climate goals. The development of hydrogen FCEVs that account for the cylindrical tanks make Type IV hydrogen storage already fit for vehicles.

Policy: Gains in design will help in achieving hydrogen goals. The US Department of Energy has created a strategic framework to achieve widespread use of hydrogen. Its breakdown of 2030, 2040, and 2050 help in guiding development. These policies keep in mind world goals, and what is needed to meet them. The next steps in terms of policy are to set specific monetary investment goals needed to achieve goals set in place and where this money should come from. These guidelines are great, but they should be adapted every few years to adjust for the new progress of R&D. Developers should not be pigeonholed in just reaching the guidelines; they should go above and beyond.

Limitations: This research does not account for the end-of life part of life cycle analysis. This includes disposability of chemicals, materials, and catalysts. Production is touched upon but not emphasized.

Future Research: Further research still needs to be conducted to realize the full potential of hydrogen storage technologies in order the make the best choice for cars in the future. New catalysts are in testing, and work needs to be adjusted to commercial demands. This research must keep in mind all stages of the life cycle (production, usage, disposability) to see which storage methods are most fit for the environment, even after their usage is over. Disposability has not been a huge matter of discussion in this article but should be when investigating hydrogen storage technologies further. Depending on changes to policy, the timeline of current research, and the money invested in technology, any current opinions on the right storage type for vehicles can change. Future research must be into what can adapt these current technologies to commercial infrastructure already used in the world (such as natural gas pipelines adapted to hydrogen) and to commercial manufacturing. In addition to understanding the need to fill DOE targets, future research must consider society's own ability to accept the cost of a hydrogen society and how to establish consumers' trust in the capabilities of these new vehicles.

Conclusion

The development of more efficient, environmentally friendly, and cost-effective hydrogen storage options is essential to help make a hydrogen society a reality. In comparing both physical and



material methods of hydrogen storage, the safest option and the ones with the greatest potential are those most developed, such as LOHC and compressed storage techniques. Through systematic review, the question regarding what hydrogen storage method is most fit (efficiency, costs, environmental impact wise) and feasible for automobiles of different sizes has been tackled. The comparison of these methods to current DOE standards allows for the most objective comparison of different methods in their current state, without the difficulty of comparing the potentials of technology. Recognizing the growing potential of material storage methods, greater research is needed to make reactions more efficient to create less stress on the system, in the form of heat and pressure, so that material storage methods can be reevaluated for future use in automobiles. Currently, the most effective methods are LOHC and compression based. They are the ones best fitted to automotive applications, without the bulk of thermal management systems and fear of system failure, but further research and development into compressed physical storage and LOHCs can increase their use in the automotive world, decrease their environmental impact, and pave the way for a transition to widespread use of hydrogen-fueled transportation.

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