REVIEW

Energy Technology
Generation, Conversion, Storage, Distribution
www.entechnol.de

Large-Scale H_2 Storage and Transport with Liquid Organic Hydrogen Carrier Technology: Insights into Current Project Developments and the Future Outlook

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The green hydrogen economy is evolving rapidly, accompanied by the need to establish trading routes on a global scale. Currently, several technologies are competing for a leadership role in future hydrogen value chains. Within this context, liquid organic hydrogen carrier (LOHC) technology represents an excellent solution for large-scale storage and safe transportation of hydrogen. This article presents LOHC technology, recent progress, as well as further potential of this technology with focus on benzyltoluene as the carrier material. Furthermore, this contribution offers an insight into previous and ongoing project development activities led by Hydrogenious LOHC Technologies together with an evaluation of the economic viability and an overview of the regulatory aspects of LOHC technology.

1. Introduction

Mitigating climate change is one of the most important challenges of the 21st century. Negative effects induced by climate change, such as extreme weather events, are causing massive economic as well as irreversible ecological damage. As the emission of greenhouse gases (GHGs), especially CO_2, is mainly responsible for the anthropogenic change of climate, it is essential to reduce it. To achieve this, fossil fuel consumption must decline significantly, which in turn requires energy efficiency improvements and energy substitutes based on renewable resources. However, the potential, as well as the demand for renewable energy is not equally distributed with respect to geographical regions. Therefore, large amounts of clean energy will be produced, stored, and distributed around the globe. For that, trading routes for green energy will need to be established on similar lines as with fossil-based fuels. Many forecasts on a global scale predict green hydrogen will become one of the major energy commodities in the future because of its various end-use scenarios.[1,2] However, due to its physical properties, the storage and transportation of molecular hydrogen is unfavorable for large-scale and long-distance trade routes. Several technologies for the efficient handling of hydrogen have been established and are still evolving. To achieve sufficient gravimetric energy densities, which are necessary for the hydrogen storage and transport, molecular hydrogen at atmospheric conditions needs to be processed, for example, compressed, liquefied, or chemically transformed into synthetic hydrogen derivatives.[3] The latter, the transformation of hydrogen into synthetic hydrogen derivatives, is conducted via a chemical reaction between molecular hydrogen and a suitable counterpart forming a hydrogen rich carrier molecule that enables easy and efficient storage and transport.

Chemically bound hydrogen can be generally categorized into carbon-based and noncarbon-based approaches. For the latter, noncarbon-based approaches, ammonia and derivatives thereof are considered as well-suited hydrogen carrier materials mainly due to the established Haber Bosch production process (currently relying on fossil hydrogen), the relatively high H_2 storage capacity, and the abundance of the feedstock nitrogen (78% N_2 in air). The reconversion of ammonia to molecular hydrogen is, however, like with carbon-based hydrogen carrier, not yet developed at industrially relevant scales and remains therefore a topic of research and development.[4–6] Due to the high toxicity of ammonia, strict safety standards need to be followed to guarantee safe handling of this nitrogen-based hydrogen carrier. For carbon-based approaches on the other hand, molecules based on CO_2 and H_2 as a feedstock represent promising hydrogen carrier candidates as they are fully compatible with the existing fossil fuel infrastructure. Molecules such as methane, methanol, synthetic fuels, etc. are prominent examples of this category. Although this carbon-based concept is carbon neutral in total, limitations in some applications must be considered due to local occurring CO_2 emissions at the energy demand site as well as the requirement for sustainable CO_2 sources at the energy supply site. Another carbon-based type of hydrogen carrier, the liquid

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The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/ente.202301042.

DOI: 10.1002/ente.202301042

organic hydrogen carriers (LOHCs),[7] circumvents the dependency on CO_2 by neither emitting nor relying on CO_2. Key to this technology is the LOHC carbon backbone that stays intact during storage and release of hydrogen. LOHCs are characterized as cyclic hydrocarbons that can be used several hundred times to store and release hydrogen in addition to an excellent compatibility with use of current liquid fossil fuel infrastructure. The company Hydrogenious LOHC Technologies GmbH together with academic partners has significantly contributed to the development of LOHC technology with the focus on benzyltoluene as the carrier molecule. Thus, the technology has reached a level of maturity that is at the stage of an implementation at industrial scale. Similarly, LOHC technologies using carrier molecules other than benzyltoluene have been developed by other players, for example, Chiyoda who focuses on toluene as an LOHC. This article presents LOHC technology giving an insight into current projects and future prospects.

2. LOHC Technology

In general, the word "hydrogen carrier" describes materials that store hydrogen via physical or chemical principles. In the case of physical binding, hydrogen is bound (stored) in a sorption mechanism to suitable materials via attractive interactions between molecular hydrogen and the carrier material. Desorption (release) of the hydrogen is initiated by an adaption of the environment, in particular by variations of temperature and/or pressure.

In contrast, chemical principles rely on an actual chemical reaction between hydrogen and a suitable reaction partner (unloaded carrier precursor), generating the respective loaded chemical hydrogen carrier. As a result, hydrogen is stored as a newly formed chemical compound with no molecular hydrogen being existent. Depending on the application, the hydrogen carrier can either be used directly in further processes or the hydrogen carrier can be converted back to molecular hydrogen and the unloaded carrier. In the following sections, only chemical hydrogen binding principles will be described with a focus on LOHCs.

Per definition, the hydrogen-loaded materials are called "hydrogen carriers", whereas unloaded materials are referred to as "hydrogen carrier precursors." In addition, the terminology of loaded/unloaded, charged/discharged, and hydrogenated/dehydrogenated (hydrogen) carrier is in use, specifying the hydrogen-rich or hydrogen-lean carrier material, respectively. In the context of LOHCs, the latter terminology loaded/unloaded, charged/discharged and hydrogenated/dehydrogenated is preferred and will be used within this publication.

On a conceptual level, a differentiation between reversible and nonreversible as well as cyclic and noncyclic approaches must be made (**Figure 1**). In nonreversible concepts, the hydrogen carrier itself is the target component, which is used directly either in electrochemical or in thermal conversion processes or as a feedstock in the chemical industry. Examples include carbon-based molecules like methane, methanol, synthetic fuels, as well as ammonia. In contrast, reversible concepts describe systems in which the hydrogen can be reversibly bound to and released from the carrier material in a dedicated process step targeting molecular hydrogen as the desired end-product. In this case, the carrier material is used only as a transport medium and is not involved in the production or the final use of hydrogen. If the unloaded carrier is later discarded or used for alternative purposes, the concept can be described as reversible noncyclic. The cracking of ammonia represents an example during which hydrogen is obtained next to the carrier precursor nitrogen. The nitrogen is typically discarded, for example, released into the atmosphere.

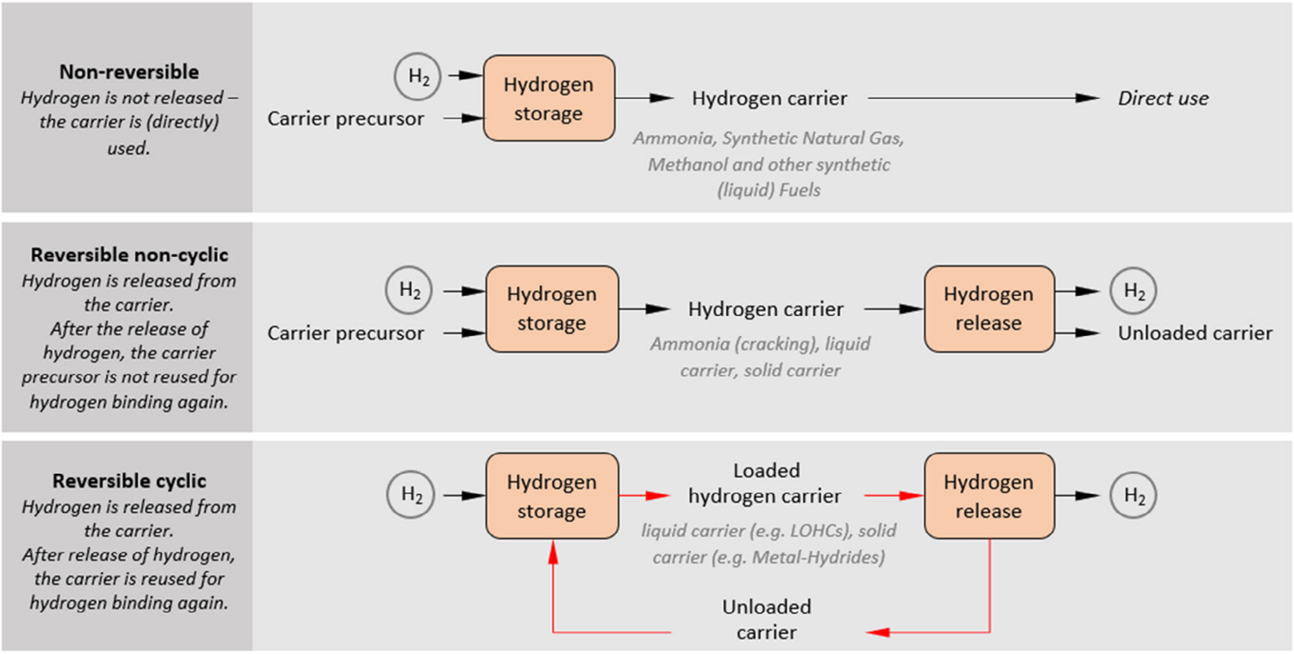


Figure 1. Overview of hydrogen carrier categories.

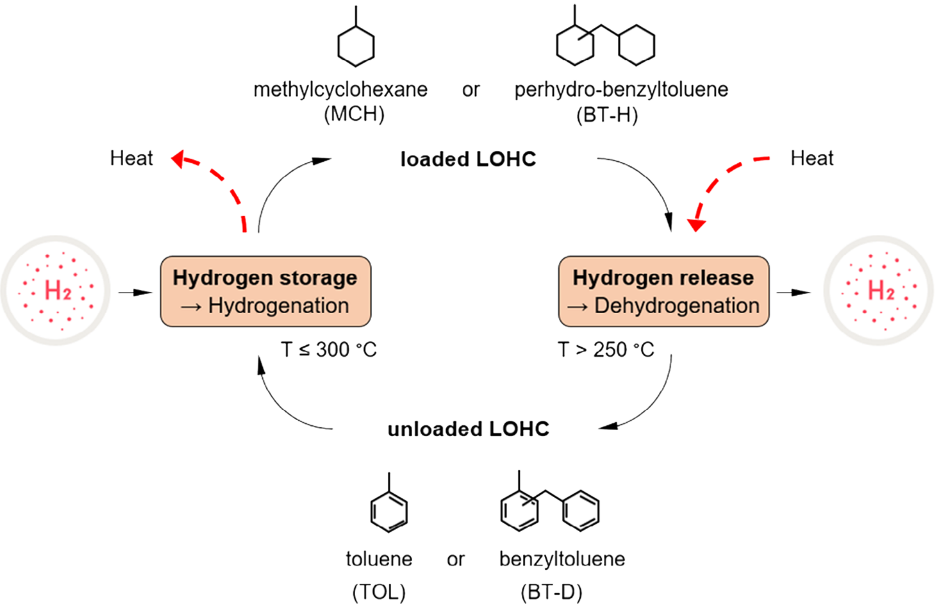
Another example is given by the hydrogen carrier methylcyclohexane, which is reconverted to hydrogen and toluene in a dehydrogenation reaction. In a noncyclic scenario, the toluene will not be transported back to the hydrogen providing location, which means that it will no longer serve as a hydrogen carrier. Instead, the toluene can be repurposed as a feedstock for the chemical industry. However, if the unloaded carrier is reused as a hydrogen carrier, the concept is reversible and cyclic. This represents a circular economy where the carrier is repeatedly loaded and unloaded with hydrogen. In other words, the carrier only serves as a means of transporting hydrogen. Examples are LOHCs such as toluene or benzyltoluene that are cyclically charged and discharged with hydrogen (**Scheme 1**).[8]

In general, the key decision criteria for reversible cyclic versus reversible noncyclic concept are the market need and the economic value of the unloaded carrier after hydrogen release. Depending on that, the unloaded carrier is either used as a hydrogen carrier again or reused for alternative purposes. The previously mentioned hydrogen carrier system based on toluene, for example, is discussed in both reversible cyclic and noncyclic scenarios.

Early demonstration projects use ammonia in nonreversible or reversible noncyclic approaches, whereas reversible cyclic applications are predominantly performed with liquid and solid carriers. Within the liquid carrier category, the focus lies on the reversible cyclic LOHC concept, which has been under intensive investigation since the beginning of the 2000s.[9] As a result, LOHCs have especially evolved over the past 10 years as most suitable for industrial demand, in particular the LOHC pairs toluene/methylcyclohexane as well as benzyltoluene/perhydro-benzyltoluene. In both cases, the carrier material toluene and benzyltoluene are commercially available in large quantities and exist in a liquid state at temperatures relevant for logistics (−25–60 °C). Another LOHC material is dibenzyltoluene (DBT), which has already demonstrated its eligibility as LOHC in previous projects. However, the focus has shifted to use preferably BT due to superior properties of BT (e.g., viscosity). The hydrogen storage (hydrogenation) release (dehydrogenation) from the LOHCs are catalytic reactions without the necessity of additional cofeeds and without the formation of coproducts. Within the context of LOHCs, benzyltoluene is often abbreviated as BT-D (**B**enzyl**T**oluene-**D**ehydrogenated) and perhydro-benzyltoluene as BT-H (**B**enzyl**T**oluene-**H**ydrogenated).[8] LOHC technology based on benzyltoluene and the LOHC material itself can generally be described by the term LOHC-BT.

Regarding thermodynamics, storage of hydrogen via LOHC is an exothermal process and the reverse reaction, release of hydrogen, is an endothermal process (Scheme 1). Both variants are catalytic reactions and depending on the reaction conditions, the process is either pushed toward hydrogenation or dehydrogenation. Particularly, the catalyst composition, the reaction pressure, and the reaction temperature determine the type of reaction (hydrogenation vs. dehydrogenation). LOHC dehydrogenation prefers elevated temperatures and low pressures, whereas for hydrogenation the opposite is true. For example, the dehydrogenation of perhydro-benzyltoluene requires a reaction enthalpy of 63.5 kJ $(\text{mol}_{H_2})^{-1}$ (8.8 kWh$(\text{kg}_{H_2})^{-1}$)[10] at a temperature level of at least 250 °C to release relevant amounts of hydrogen under atmospheric pressure.[11,12] Since higher temperatures improve the kinetics of the dehydrogenation reaction and shift the thermodynamic equilibrium, temperatures of ≈300 °C are preferred to allow an efficient hydrogen release process.[13]

The gravimetric hydrogen storage density is 6.1 wt% for methylcyclohexane and 6.2 wt% for perhydro-benzyltoluene,[14] whereas the volumetric values correspond to 47 $\text{kg}_{H_2}\,(\text{m}_{\text{LOHC}})^{-3}$ for methylcyclohexane[14] and 54 $\text{kg}_{H_2}\,(\text{m}_{\text{LOHC}})^{-3}$, for perhydro-benzyltoluene.[15] Hence, hydrogen-loaded benzyltoluene (BT-H) offers favored properties with respect to gravimetric and



Scheme 1. Reversible, cyclic hydrogen carrier concept represented by the LOHC pairs toluene/methylcyclohexane (TOL/MCH) and benzyltoluene/perhydro-benzyltoluene (BT-D/BT-H).

volumetric hydrogen densities compared to methylcyclohexane. Furthermore, the enthalpy of reaction for dehydrogenation of methylcyclohexane is ≈9.4 and 8.8 $\text{kWh}(\text{kg}_{\text{H}_2})^{-1}$ for perhydro-benzyltoluene.[14] The boiling point of both hydrogen carriers differs significantly, 101 °C for methylcyclohexane[16] and 278–282 °C for perhydro-benzyltoluene.[17] A higher LOHC boiling point is a desired property that facilitates the separation of hydrogen gas from the unloaded carrier after the dehydrogenation reaction. Furthermore, methylcyclohexane is, in contrast to perhydro-benzyltoluene, highly flammable, which represents a risk potential especially if bulk amounts are handled. From that perspective, the properties of perhydro-benzyltoluene are superior to those of methylcyclohexane. Nevertheless, the pair toluene/methylcyclohexane plays an important role as an LOHC, mainly due to low costs for the LOHC material itself as well as due to existent infrastructure and higher volumes of traded commodity nowadays in particular in the chemical industry.

While hydrogenation of cyclic aromatics is well known in the chemical industry and adapted to the LOHC technology, dehydrogenation aiming for high-purity H_2 with minimal carrier degradation has specifically been developed for LOHC applications.

As a midstream industry, clean hydrogen transportation interacts by definition, with both upstream (hydrogen producing) and downstream (hydrogen off-taking) segments. The formation and establishment of an LOHC-based ecosystem relies on the superior compatibility of this technology with currently existing industrial chemical processes, infrastructure, and logistics. On the upstream side, the LOHC technology interfaces with hydrogen production technologies. For instance, low-carbon hydrogen (fossil-based) is produced via steam methane reforming (SMR) or autothermal reforming (ATR) with carbon dioxide capture and storage (CCS), known as blue hydrogen. Renewable hydrogen, denominated as green, is generated through diverse electrolysis technologies, such as alkaline, proton-exchange membrane (PEM), solid-oxide electrolyzer cell (SOEC), and anion-exchange membrane (AEM). The Hydrogenious storage plant, in which the LOHC benzyltoluene is charged with hydrogen (hydrogenation), can cope with hydrogen streams in very broad purity and pressure ranges. With regard to pressure, a minimum of 10 barg is required for the hydrogenation reactor, which covers all blue and most green hydrogen production cases without the need for prior hydrogen compression. Examples of currently established pressure levels for various hydrogen producing technologies are shown in **Table 1**.

Table 1. Compatibility of Hydrogenious storage plant with hydrogen feed streams at different pressure levels.

	Outlet pressure in barg	H_2 compression required upstream of hydrogenation
SMR/ATR with CCS	24[67]	No
PEM	30[68]	No
AEM	35[69]	No
Alkaline electrolysis	0.3[70]	Yes
SOEC	0[20]	Yes

With regard to hydrogen purity levels, the conventional storage plant design of Hydrogenious tolerates considerable amounts of the most common trace substances present in green and blue hydrogen product streams. For instance, the process is compatible to water (order of 10^3 ppm), methane (order of %), and carbon dioxide (order of %) without any relevant detriment to the catalyst activity and process robustness. Ongoing R&D investigations at Hydrogenious partners Friedrich-Alexander University Erlangen-Nuremberg and Helmholtz Institute Erlangen-Nuremberg for Renewable Energy indicate that the hydrogenation process can even use hydrogen-rich gas mixtures as feedstock. These could contain higher concentrations of impurities, which offer an opportunity to simplify the H_2 purification process and minimize losses in the scope of the hydrogenation plant.[18,19] Critical impurities, however, such as sulfur, chlorine, or ammonia that act as catalyst poison, must be removed before the hydrogen stream is used for LOHC-BT hydrogenation.

The exothermal LOHC-BT hydrogenation reaction is carried out in a continuous operation mode at a flexible temperature between 150 and 300 °C and can modulate the production quickly from the turndown point to the nominal capacity, which is beneficial if the feedstock is a renewable, fluctuating hydrogen source. The exothermal reaction enthalpy (8.8 $\text{kWh}(\text{kg}_{\text{H}_2})^{-1}$) at temperatures around 150–300 °C is suitable for several off-takers, for example, in the form of medium-pressure steam. For instance, thermal water desalination processes, refining and petrochemical unit operations or electricity generation represent potential use cases. In addition, it is possible to export steam as feed to a solid oxide electrolyzer cell. As an example, the SUNFIRE-HYLINK SOEC system requires steam at 3.5–5.5 barg at 150–200 °C.[20] Such steam qualities can easily be provided as a byproduct of the Hydrogenious storage plant. Based on the reaction stoichiometry and the latent heat of water, the SOEC requires ≈5.6 $\text{kWh}(\text{kg}_{\text{H}_2})^{-1}$ for the vaporization of water, which can be fully covered by the 8.8 $\text{kWh}(\text{kg}_{\text{H}_2})^{-1}$ reaction heat of the hydrogenation process.

The produced perhydro-benzyltoluene is regarded as a safe and easy-to-handle hydrogen carrier, owing to the physical and chemical properties (**Table 2**). Key advantages of LOHC-BT are reflected by the high flash point (hardly flammable), low toxicity potential to humans, and the possibility to store BT-D/BT-H under ambient conditions. Therefore, BT-D/BT-H can be safely stored for a long time in conventional tanks. This allows for rapid and low-cost repurposing of current fossil fuel terminals to serve the hydrogen economy.

Next, benzyltoluene loaded with hydrogen (BT-H) has to be delivered from the hydrogenation site (storage plant) to the dehydrogenation site (release plant), where the hydrogen is unloaded in a chemical reaction. The unloaded carrier BT-D is then transported back to the hydrogenation site, to be reloaded with hydrogen. Both hydrogenation and dehydrogenation are therefore part of the hydrogen transport chain.

In continental supply chains, trucks, which can carry up to 27 tons of LOHC-BT ($\hat{=}$1.5 $\text{tonnes}_{\text{H}_2}$), may be preferred for shorter transport distances and hydrogen throughput. With increasing hydrogen demand, they may be replaced with trains, which can carry roughly up to 1000 tons of LOHC-BT ($\hat{=}$54 $\text{tonnes}_{\text{H}_2}$). Alternatively, crude and refined product

Table 2. Physical and chemical properties warranting benzyltoluene and perhydro-benzyltoluenes' status of a safe and easy-to-handle LOHC pair. Additional information about the properties of LOHCs is listed in the DIN SPEC 91437.[8]

	Unloaded carrier: Benzyltoluene[71]	Loaded hydrogen carrier: Perhydro-benzyltoluene[17]
Melting point (1 atm) [°C]	−80–−70	−70
Boiling point (1 atm) [°C]	280–290	278–282
Flash point [°C]	137	130
Vapor pressure (20 °C) [hPa]	<0.01	<0.01
Density (20 °C) [g cm^{-3}]	0.995	0.996–0.876
Dynamic viscosity (20 °C) [mPa s]	/	4.3–6.6
Kinematic viscosity (20 °C) [mm^2 s^{-1}]	4.0	4.4–7.6
Solubility in water (20 °C) [mg l^{-1}]	< 0.1	<0.1
Autoignition temperature [°C]	510	510

pipelines can be coutilized or even fully repurposed to transport hydrogen in the form of LOHC. Benzyltoluene-based LOHC is compatible to existing pipelines due to the similar fluid dynamic properties of the loaded BT-H and unloaded BT-D to fossil energy carriers. As for long-haul maritime shipping, LOHC can be transported with chemical tankers while the capacity of the H_2 via LOHC supply chain is ramping up. Chemical tankers are easily available and can be readily used on demand. In the future, as the LOHC ecosystem expands, bulk carriers will become the preferred choice for shipping.

Flexibility and intermodal transport using existing infrastructure are remarkable features of the LOHC technology, which can facilitate hydrogen supply chains of different size and nature. For instance, benzyltoluene can be hydrogenated in a large-scale storage plant, for example, in the Middle East. Subsequently, it can be shipped in a chemical tanker to hydrogen demand centers such as Central Europe (**Figure 2**). At the destination, hydrogen is released in a centralized large-scale release plant that converts most of the landed LOHC feeding hydrogen into a transnational gas pipeline system. A certain part of loaded LOHC could also be transferred into smaller river barges that distribute the LOHC to off-takers with a specific high demand for hydrogen, known as the 1:*n* principle (one storage plant supplies LOHC to several release plants). Local dehydrogenation units could supply hydrogen directly to off-takers such as the steel or chemical industry. Furthermore, last-mile truck deliveries through densely populated urban centers, for example, to an LOHC-based hydrogen refueling station, are feasible due to the low hazard potential of the LOHC benzyltoluene/perhydro-benzyltoluene. As a reference, even the strict regulations of the Agreement concerning the International Carriage of Dangerous Goods by Road (ADR) do not impose limitations on road transport of LOHC-BT in the European Union, as opposed to other hydrogen carriers, such as ammonia.

As the hydrogen storage plant is required to interact with upstream technologies, the dehydrogenation plant will interface with downstream technologies that consume hydrogen. LOHC-BT dehydrogenation is an endothermic reaction, which takes place at temperatures around 300 °C and is performed in a continuous operation mode. The reactor heating can be accomplished with different concepts, for example, using high-pressure steam or a synthetic heat transfer fluid at low pressure or via direct electrical heating. The most obvious heat sources for the LOHC dehydrogenation reaction are electricity, gas-fired heating, or heat integration with industrial heat sources. Sources of origin for the latter might be waste incineration power plants, steel furnaces, or cement plants. If a self-sustaining hydrogen release system is desired, a fraction of the released hydrogen can be combusted to provide the dehydrogenation reaction heat in a similar manner as most ammonia cracking processes work. However, the ammonia cracking reaction

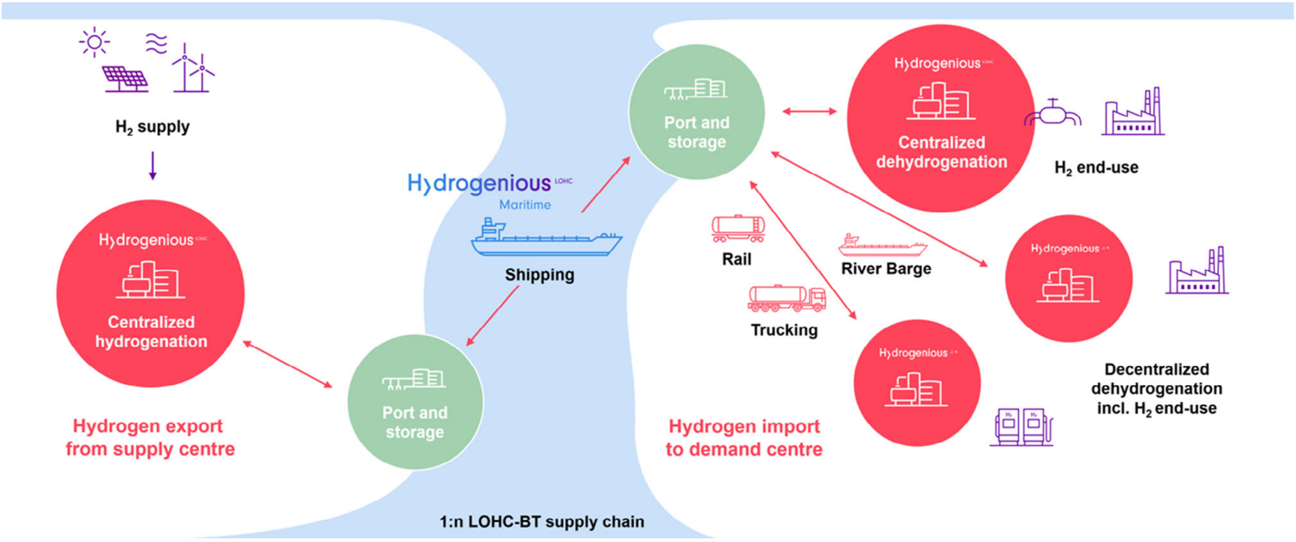


Figure 2. Overview of hydrogen supply chain based on LOHC and its 1:*n* distribution principle.

requires heat at temperatures above 600 °C, which can only be reached with a standalone gas-fired heater design.[21–23]

The crude hydrogen as released from BT-H dehydrogenation has a purity higher than 99.9%, which is enough for most applications in the power, heating, and in most industrial sectors. By adding an additional purification unit, superior hydrogen qualities, for example, fuel cell grade (ISO 14687, Grade D), can be met, with only minor hydrogen losses.

There are several companies offering LOHC technologies in their portfolio, among which Hydrogenious LOHC Technologies GmbH ("Hydrogenious") arises as a very prominent player. Hydrogenious was founded as a spin-off company of the Friedrich-Alexander University of Erlangen-Nuremberg in 2013 and has grown rapidly with currently more than 200 employees. Furthermore, due to market and project developments, Hydrogenious has formed joint ventures with partners, inter alia, in the Middle East and in Norway. The core technology of the German company features the storage and release of hydrogen using the LOHC pair benzyltoluene/perhydrobenzyltoluene to facilitate the storage and transport of hydrogen. Since 2015, Hydrogenious has demonstrated its technology in multiple projects, based on containerized systems.

After several successful project activities, for example, the EU-funded HySTOC project in Finland,[24] the first complete benzyltoluene-based LOHC supply chain was established in 2022 within the H2Sektor pilot project that was subsidized by the Bavarian State Ministry for Economic Affairs and Energy.[25] The core of the project represents a hydrogen refueling station in Erlangen (Germany), built and operated by H2 MOBILITY, Germany, that was supplied with hydrogen not only in gaseous form but also via LOHC-BT. For the LOHC-BT supply chain, green hydrogen is produced via a PEM electrolysis fed by photovoltaics and bound to the LOHC in a hydrogenation reactor at the Hydrogenious headquarters. The hydrogen-loaded LOHC is transported to the publicly accessible hydrogen refueling station (**Figure 3**), where the hydrogen is released from the LOHC in a dehydrogenation unit and supplied in fuel cell quality at 45 barg to the station's pressurized buffer tank. The dehydrogenated BT-D is transported back to the Hydrogenious headquarter, where the next cycle of LOHC-BT usage starts. The dehydrogenation unit with a maximum hydrogen release capacity of up to 9 tons of H_2 per year is not only used as a hydrogen supplier to the refueling station but also as a long-term test facility, which has operated for several thousand hours since its commissioning.

Several state-of-the-art LOHC hydrogenation and dehydrogenation units based on benzyltoluene, like the one in Erlangen (Figure 3) have already been built and are in operation. These demonstration units were designed to process volumes of hydrogen in the order of up to 8–70 tons of hydrogen per year. The next steps in scaling up the LOHC-BT technology are hydrogenation and dehydrogenation plants capable of processing hydrogen in the order of kilotonnes per year. Hydrogenious is currently very active in building these plants to scale up the benzyltoluene-based LOHC technology to an industrially relevant order of magnitude, with ranges upward of hundred tons of hydrogen per year per supply chain. For an overview and an outlook of the upcoming scaling of hydrogen transport using LOHC-BT, a selection of current projects developments within Hydrogenious LOHC Technologies is presented.

The first step toward scaling-up is the building of a storage plant with a capacity of up to 1.8 kilotons of H_2 per year, which is expected to be commissioned in 2025.[26] The project with the name "Hector" is funded by the Ministry of Economic Affairs, Innovation, Digitalization and Energy of the State of North Rhine-Westphalia. It connects the CHEMPARK Dormagen, where renewable hydrogen will be produced as an industrial byproduct, with an LOHC storage plant that enables distribution of hydrogen to locations outside the CHEMPARK Dormagen. Together with the Dutch co-investor Vopak, the establishment of a green hydrogen supply chain between the CHEMPARK Dormagen and Rotterdam will be achieved. A release plant with a capacity of 0.5 kilotonnes of H_2 per year will be located in

Figure 3. Dehydrogenation unit of Hydrogenious at the hydrogen refueling station of H2 MOBILITY Germany in Erlangen.[63]

Rotterdam acting as the counterpart to the storage plant in Dormagen.

To further scale up LOHC supply chains based on benzyltoluene, Hydrogenious focuses on project sizes with capacities ranging from 4 to 8 kilotonnes of H_2 per year. Several publicly funded projects support these efforts to speed up and derisk commercialization. For instance, the German flagship project TransHyDE,[27] which is part of the largest funding initiative of the Federal Ministry of Education and Research (BMBF) around the hydrogen economy, plays a crucial role in designing the up-scaled processes. The TransHyDE project "Heligoland" investigates the hydrogen supply chain from Heligoland to Hamburg by means of LOHC-BT. Within the project, a storage plant with a capacity of 8 kilotons of H_2 per year is designed with a special focus on high dynamics and low partial load to be well compatible to fluctuating renewable energies. Additionally, concepts for waste heat utilization of the storage plant like sea water desalination or power generation are being investigated. Furthermore, logistic-related topics, such as the International Maritime Organization (IMO) classification for LOHC-BT in bulk transport via ship, the possible usage of existing transport assets, as well as the retrofit of existing tank farms are elaborated. On the potential dehydrogenation site in Hamburg, a 4 kilotons of H_2 per year release plant is being planned. To further improve the dehydrogenation process, alternative sources for heat integration will be investigated and concepts for space-optimized systems as well as control strategies will be developed. Further upscaling of the hydrogenation and dehydrogenation processes to 100 kilotonnes of H_2 per year per supply chain is an important objective within the TransHyDE Heligoland project to meet the fast-growing market demand for large-scale intercontinental H_2 trading.

Results derived from the TransHyDE project will merge with other project streams from Hydrogenious such as the ones within the context of the "Important Projects of Common European Interest", a funding scheme by the EU commission and EU member states. Hydrogenious has currently two projects placed in the Hy2Infra wave, the so-called "Northern Green Crane"[28] and the "GreenHyrogen@BlueDanube"[29] project. Within the Northern Green Crane project a supply chain of 8 kilotons of H_2 per year is planned. Together with industry partners Vopak and the Get H2 initiative, the construction of one storage plant with a capacity of 8 kilotonnes of H_2 per year and two release plants, with a release capacity of 4 kilotonnes of H_2 per year each, is envisioned. This project is expected to become one of the first industrial-scale green hydrogen import projects based on LOHC-BT.

A similar-sized LOHC-BT supply chain of around 10 kilotonnes of H_2 per year is being investigated in a joint study by ADNOC, Uniper, Jera, and Hydrogenious to establish an LOHC transport route between the United Arab Emirates and Germany.[30] After setting proof points with first midscale plants, the natural step is to scale-up toward larger industrial plant capacities.

The next scale-up step toward industrial plants in the order of 100 kilotonnes of H_2 per year are conducted within the H2A platform, which was founded by the partners Hydrogenious, Evos, and Port of Amsterdam.[31] The H2A platform focuses on the development of green hydrogen supply chains through imports via the port of Amsterdam. In particular, an industrial-scale release plant with a capacity of 30–175 kilotonnes of H_2 per year as well as surrounding infrastructure will be developed. As the port of Amsterdam is located in a densely populated area, LOHC-BT is well suited for that application and regarded as the preferred carrier technology.

As of March 2023, UK-based fuel distributor Greenergy and Hydrogenious have announced they will be undertaking a joint feasibility study to investigate LOHC-based green hydrogen supply from Canada to the UK[32] The study will analyze the economics of constructing hydrogenation and dehydrogenation assets to import hydrogen in the order of at least 30 kilotonnes of H_2 per year into the UK based on Hydrogenious LOHC-BT technology. Greenergy's access to existing bulk liquid terminals is ideal for the import, release, and distribution of hydrogen using the LOHC-BT technology.

In May 2023, the renewable energy development company CWP Global and Hydrogenious agreed to conduct a feasibility study to explore a green hydrogen transport chain from Morocco to Europe.[33] The project is called 15GW AMUN and serves as a base for model considerations investigating a 175 kilotons of H_2 per year LOHC-BT-based supply chain.

Next to Hydrogenious, there are several other companies who are very active in the development of industrial LOHC solutions. While the LOHC pair benzyltoluene/perhydro-benzyltoluene is exclusive to Hydrogenious, toluene/methylcyclohexane LOHC concepts are offered by other companies. Due to the advantageous properties of LOHC technology, renowned companies have also joined the LOHC market. Chiyoda Corporation, for example, began in 2015 using the LOHC pair toluene/methylcyclohexane to demonstrate a hydrogen supply chain from Darussalam (BN) to Kawasaki (JP) based on tank containers. The project showed that the storage and transport of over 100 tons of hydrogen was feasible in a period of ten months.[34] A further development of their LOHC technology was announced in 2021 in a joint project study of Chiyoda with KOOLE terminals in the Port of Rotterdam.[35] Recently, the company Axens, which has more than 50 years of experience in the hydrogenation of aromatics, signed a joined commercial cooperation agreement with Chiyoda to concentrate their LOHC activities. Together, the expertise of Axens' toluene hydrogenation technology and Chiyoda's methylcyclohexane dehydrogenation technology is combined allowing fast track approaches for project implementation and creating a single point of contact for potential customers.[36] Independently, the company Honeywell announced in April 2023 to use the LOHC toluene/methylcyclohexane as carrier system as well. Their approach is based on the hydrogenation/dehydrogenation technology of the subsidiary Honeywell UOP.[37] Furthermore, AirProducts holds patents dating back to 2003 covering important aspects of the LOHC approach including heteroaromatic compounds such as carbazole-based hydrogen carriers.[9,38] Unfortunately, to the best of our knowledge, the latest activity related to commercialization of LOHC has been reported by AirProducts in 2012 in form of the final project report.[39]

3. Economic Viability

LOHC-BT technology offers several benefits in terms of safety and ease of handling that are complex to quantify but, play a crucial role in project implementation. In particular, its economic competitiveness is proven by various independent studies based on comparative levelized cost of hydrogen calculations from renowned agencies and institutes.[40–45] **Figure 4** shows comparisons that LOHC is highly competitive when compared to other hydrogen transport methods and the differences are considered to be accurate when taking into account the character of such high-level studies.

As soon as a detailed technoeconomic analysis of a certain project setup is prepared, the access to the correct process parameters is indispensable in order to generate meaningful results. The following tables summarize some of the key parameters of the LOHC-BT technology to assess economic viability of a specific H_2 supply chain. Although they might vary according to project-specific circumstances, the values (**Table 3–5**) provide a good reference.

Though a full financial model will require a myriad of economic input parameters, the viability of projects involving LOHC-BT technology is determined essentially by three factors: 1) cost of benzyltoluene; 2) synergies with pre-existent infrastructure, for example, port and storage facilities, reducing overall capital expenditure; and 3) cost of thermal energy for release plant at H_2 import location.

In addition to the technological aspects, shipping and logistical costs also play a substantial role in the economic viability of the full H_2 supply chain based on LOHC-BT. Due to the cyclic reversible nature of the LOHC technology BT-H will be transported to the energy demanding site and subsequently the hydrogen-unloaded BT-D has to be transported back to the energy providing site. The additional back transportation is however, from an economic perspective, not stringently disadvantageous due to the fact that tanker ships for energy import usually travel back to the energy-providing site anyway. Whether the return travel is conducted with (BT-D) or without freight represents only a minor economical difference. In general, the shipping and logistical costs are governed by the following four aspects: 1) charter rates of chemical tankers or bulk carriers; 2) fuel costs and carbon emission fees; 3) voyage-related fees, for example, toll and port fees; and 4) buffer tankage capacity of LOHC-BT.

Table 3. Key parameters of Hydrogenious LOHC-BT technology.

Parameter	Value	Unit
Theoretical gravimetric hydrogen density in BT-H	6.2	wt%
Optimal reaction swing	$\geq$90	%
BT degradation	0.05–0.10	wt%/cycle

Table 4. Key technical parameters of the Hydrogenious hydrogenation process.

Parameter	Value	Unit
Specific electricity consumption (balance of plant, w/o H_2 compression)	$\leq$0.6	$kWh(kg_{H_2})^{-1}$
Hydrogen loss	$\leq$0.1	%
Heat utilization potential (exothermal process)	−8.8	$kWh(kg_{H_2})^{-1}$

Table 5. Key technical parameters of the Hydrogenious dehydrogenation process.

Parameter	Value	Unit
Specific thermal energy demand (endothermal process)	11	$kWh(kg_{H_2})^{-1}$
Specific electricity consumption (balance of plant, w/o H_2 compression)	$\leq$1.6	$kWh(kg_{H_2})^{-1}$
Specific electricity consumption for H_2 compression to 50 barg	$\leq$2.25	$kWh(kg_{H_2})^{-1}$
Hydrogen loss at quality > 99.9% H_2	0	%

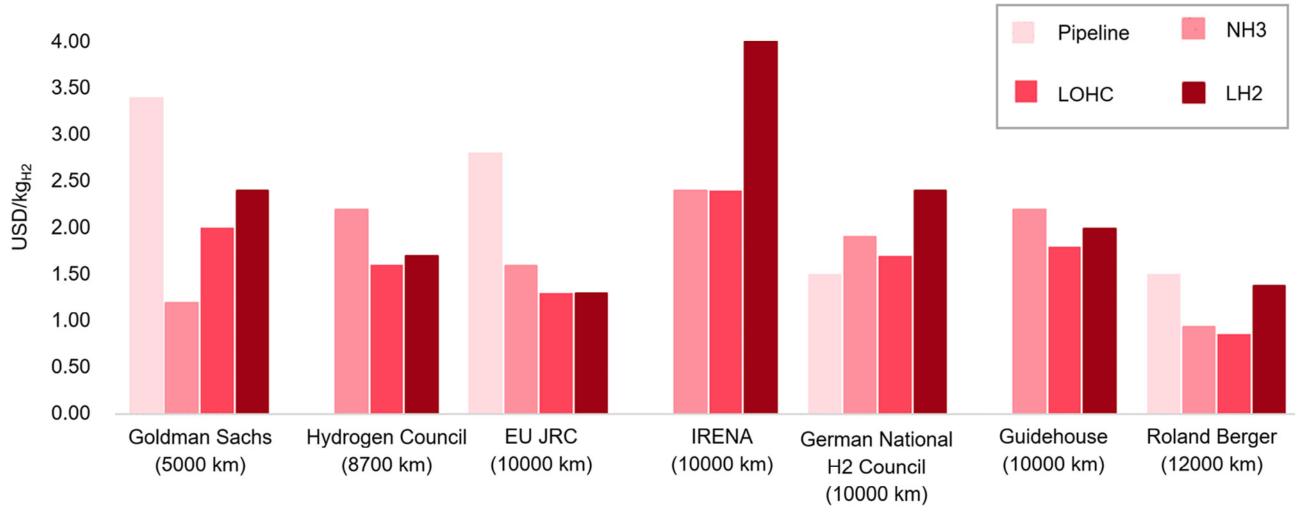


Figure 4. Comparison of total cost of ownership (TCO) of hydrogen transport in USD/kg_{H2} via different large-scale transport technologies published by diverse entities.[40–45] Depending on the individual studies, LOHC is either toluene, benzyltoluene, or dibenzyltoluene.[8] Note: H_2 production costs are not included in these TCO studies. Assumptions for all conversions: 1 EUR = 1.15 USD; 1 EUR MWh^{-1} = 0.033 $EUR(kg_{H_2})^{-1}$. For further studies comparing different LOHCs with each other, see refs. [64–66].

The highlighted factors need to be carefully considered and weighed up during project development.

As a generic example, the following 175 kilotons of hydrogen per year LOHC-BT supply chain, where hydrogenation and dehydrogenation plants are ≈1500 nautical miles (≈3000 km) apart from one another, is given. A transport radius of ≈3000 km was chosen because this distance includes potential hydrogen supply routes within Europe as well as from the Middle East and North Africa region to Southern Europe. Depending on the transport distance, the proportion of the LOHC-BT supply chain on the levelized cost of hydrogen in relation to H_2 feedstock costs has to be adopted. Longer transport routes will increase and shorter transport routes decrease the proportion of the costs for the LOHC-BT supply chain, respectively. Based on the parameters in Table 3–5, Hydrogenious estimates the following breakdown of relative levelized cost of hydrogen (**Figure 5**).

Figure 5 reveals, compared to the studies in Figure 4, the contribution of each step of the value chain to the levelized costs of hydrogen across the whole H_2 supply chain based on LOHC-BT. Hereby, the green hydrogen production itself is responsible for almost half of the levelized costs. Due to the transport distance of ≈3000 km, the portion of logistics and BT inventory contributes to around 17% of the levelized costs. Both values, logistics and BT inventory, are sensitive to the transport distance and need to be adopted for other transport routes accordingly. The storage of hydrogen in LOHC-BT contributes, in contrast to other hydrogen storage and transport technologies, only to about 8% of the levelized costs in this scenario. The complementary process, the H_2 release is responsible for slightly more than one-fourth of the levelized costs, which is reflected mainly by the energy demand of the dehydrogenation reaction. The requirement to provide energy for the dehydrogenation reaction, however, is an intrinsic factor that is valid for all hydrogen carrier technologies, for example, also for ammonia backcracking. In order to reduce dehydrogenation costs at the LOHC-BT release plant, innovative concepts are under development, which are presented in the following section.

4. Future Potential of the Technology

In general, hydrogen storage using LOHCs is characterized by a high level of safety, which is based on the intrinsic LOHC property that no spontaneous release of the chemically bound hydrogen occurs. When releasing hydrogen from the LOHC, energy in the form of heat needs to be applied in the presence of a catalyst. This energy input at a temperature level >250 °C requires solutions that guarantee technical feasibility and favorable economics. Other hydrogen carriers such as ammonia require similar amounts of energy, however, to some extent at much higher temperatures (e.g., >600 °C) if conversion back to molecular hydrogen is targeted.[21–23,46]

The heat supply for the release plant correlates with CO_2 emissions; hence, low emissions are of utmost importance to guarantee low-carbon hydrogen delivered to the customer. This becomes more challenging, the larger the capacity of the dehydrogenation process. Thanks to the moderate temperature level that is required, a favored solution is the combination with existing heat sources from contiguous processes. A first and rather innovative solution constitutes process integration with a solid oxide fuel cell (SOFC) as investigated for example by Preuster et al.[47] Since SOFCs are operated at a temperature level around 700–1,000 °C and the LOHC dehydrogenation requires temperatures around 300 °C, an efficient heat transfer from the exothermal SOFC operation to the dehydrogenation reactor is possible. As a result of the combination, a total electric efficiency of up to 45% in relation to the lower heating value of the LOHC-bound hydrogen is expected. The first steps toward the realization of such a combined LOHC-BT and SOFC system is currently pursued in the project Multi-SOFC.[48] The companies Hydrogenious LOHC NRW GmbH and Robert Bosch

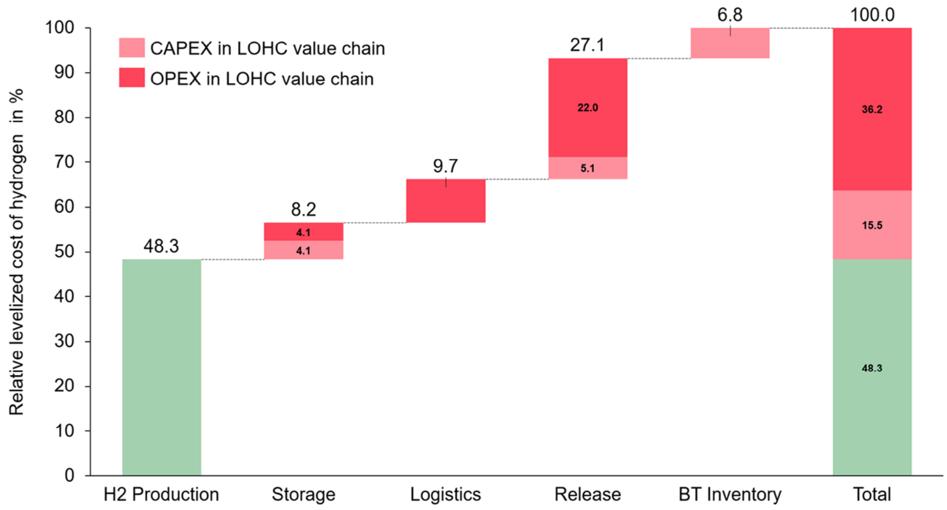


Figure 5. Levelized cost of hydrogen breakdown for a 175 kilotonnes of H_2 per year LOHC-BT-based supply chain (≈3000 km). Note: H_2 production costs are included in contrast to Figure 4. Green: Cost for hydrogen feedstock; dark red: operational expenditure for the LOHC-BT value chain; light red: capital expenditure for the LOHC-BT value chain.

GmbH provide a heat and power-generating SOFC unit to the Hermann Josef Hospital in Erkelenz that can be powered by H_2 that is locally released from BT-H. Furthermore, the utilization of a coupled dehydrogenation and SOFC unit is in particular well suited for on-board applications. In the project Ship-aH2oy by Hydrogenious LOHC Maritime AS, a zero-emission ship propulsion system will be developed based on the combined use of BT-H dehydrogenation and SOFC as a powertrain.[49] The application will be demonstrated on a Commissioning/Service Operation Vessel for offshore wind turbines and will be constructed over a project period of 5 years based on funding by Horizon Europe.

Another approach, suitable for larger energy demand, is to connect a dehydrogenation plant to an existing thermal process with large thermal power in range of several ten to hundred megawatts, such as power plants, steel furnaces, or cement plants. Hydrogenious recently concluded a feasibility study investigating heat integration possibilities of the dehydrogenation process with power plants. Obviously, this causes a minor reduction in electricity production of the power plant as a share of the heat is used elsewhere. Nevertheless, the reduced electricity output of the power plant is counteracted by superior process characteristics. Compared to two stand-alone processes, the efficiency of H_2 supply, costs, and CO_2 emissions is highly favorable for the overall combined process. In summary, the study showed a high economic benefit of a combined power and dehydrogenation plant setup, although the heat integration requires adaptions, especially regarding the design of the steam turbine.

Another concept to cover the heat demand during the dehydrogenation step is investigated by Hydrogenious and academic partners, whereby the hydrogen unloaded benzyltoluene is further used in a selective oxidation reaction to methylbenzophenone.[50] Due to the exothermic character of this additional step, an autothermal dehydrogenation is envisioned: At first, perhydro-benzyltoluene is dehydrogenated in a first-reaction zone releasing the hydrogen. Afterwards, the dehydrogenated LOHC product BT-D is forwarded into a second-reaction zone making contact with air. The following catalytic reaction is highly exothermic, and water is formed as a coproduct. The released heat can be used to cover the heat demand of the dehydrogenation reaction. The concept can still be seen as reversible cyclic because the formed methylbenzophenone is fully regenerated in a hydrogenation step back to perhydro-benzyltoluene. Ongoing investigations focus on the oxidation catalyst development, the heat integration concepts, and the handling of benzyltoluene/methylbenzophenone mixtures.

Another concept to overcome the energy demand for hydrogen release is the (direct) electrochemical utilization of chemically bound hydrogen in LOHCs. The so-called direct-LOHC fuel cell converts the chemical potential directly into electricity without the release of molecular hydrogen.[51] Hydrogenious established a Joint Development with the academic partner Helmholtz Institute Erlangen-Nuremberg for Renewable Energy and industrial partner Schaeffler to further develop this innovative approach.[52] In addition, the achievements of the company Energy 18 H should be mentioned: The scientists demonstrated a direct-LOHC-Fuel-Cell on the basis of perhydro-dibenzyltoluol.[53]

5. Regulatory Framework for Green H_2 Imported to EU via LOHC

Hydrogen regulation for international trade and import is currently in the process of development in the EU as well as in other countries. Due to the importance of the EU as a hydrogen import market, this article concentrate on regulatory aspects applying to the EU. Import of hydrogen in the EU depends on the determined hydrogen targets and compliance with EU regulation.

The main targets for the hydrogen import in the EU are defined in the REPowerEU Plan[54] by the European Commission in May 2022. Herein, the domestic production of 10 million tons of renewable hydrogen and additionally the same amount of imported renewable hydrogen is targeted by 2030. Furthermore, the need for an accelerated deployment of hydrogen infrastructure is defined as a goal in the REPowerEU Plan. LOHC is not specifically mentioned in the renewable hydrogen targets of the REPowerEU Plan, presumably due to its characteristic being a hydrogen transportation technology.

However, hydrogen carriers such as LOHCs are already mentioned in the EU hydrogen strategy published in 2020.[55] According to the EU hydrogen strategy, hydrogen can be transported in pure gaseous form, in liquid form or chemically bound, for example, as LOHC, to facilitate the transport. This highlights the recognition of LOHC as a form of hydrogen transportation under the EU strategic documents.

At the same time, transportation of hydrogen is not possible without development of the dedicated infrastructure. The regulatory framework for the development of hydrogen infrastructure, including for the use of LOHC, is based on1) TEN-E regulation[56] or regulation on guidelines for trans-European energy infrastructure that was revised in 2022 and 2) hydrogen and decarbonized gas market package[57] that should be adopted in 2023.

TEN-E defines EU rules for crossborder energy infrastructure that also includes hydrogen infrastructure for the LOHC technology. LOHC assets, such as the Hydrogenious storage and release plants, are covered by the TEN-E regulation as “reception, storage and regasification or decompression facilities for hydrogen embedded in other chemical substances with the objective of injecting the hydrogen, where applicable, into the grid.”

The hydrogen and decarbonized gas market package supports technology neutrality and includes LOHC as an option for hydrogen transportation. At the same time, the LOHC infrastructure is not subject to regulation by the hydrogen and decarbonized gas market package that provides a certain advantage. Therefore, LOHC infrastructure might be used without the need to fulfill requirements such as third-party access. The reasoning behind this is that LOHC can use a multitude of existing liquid fuel terminals whereas other technologies such as ammonia and liquified hydrogen need dedicated (new) infrastructure.

Besides the need for a hydrogen infrastructure, the import of hydrogen depends on hydrogen certification and compliance of renewable and low-carbon hydrogen with determined emissions thresholds ($kg_{CO_2eq}(kg_{H2})^{-1}$). It should be noted that different countries have different requirements for renewable hydrogen thresholds, for instance, for the USA 4.0 $kg_{CO_2eq}(kg_{H2})^{-1}$ and for the UK 2.4 $kg_{CO_2eq}(kg_{H2})^{-1}$ apply.[58]

Main requirements regarding emission thresholds for hydrogen in the EU will be determined: 1) for the renewable hydrogen, in the Renewable Energy Directive[59] and the delegated acts on conditions for hydrogen as "renewable fuels of non-biological origin" (RFNBO) (Delegated Act RED II Art. 27[3]) and the methodology for calculating life-cycle GHG emissions (Delegated Act RED II Art. 25[2] and 28[5])[60] and 2) for the low-carbon hydrogen, in the hydrogen and decarbonized gas market package, Article 8 of the Gas Directive.[57]

The Renewable Energy Directive and the Hydrogen and decarbonized gas market package are in the process of development but based on the available drafts of these regulations, it is expected that the main threshold for renewable and low-carbon hydrogen will be 70% GHG emissions savings which corresponds to emissions of $3.4\,\mathrm{kg}_{\mathrm{CO_2eq}}(\mathrm{kg}_{\mathrm{H2}})^{-1}$. Furthermore, standard CEN - EN 16325 is under revision and aims for establishing a scheme of guarantee of origins also for renewable hydrogen.

The use of LOHC for hydrogen transportation shall be taken into account in the calculation of GHG emissions. It should be noted that GHG emissions are subject to calculations through the whole supply chain from the point of production to the point of use.[61] However, the GHG emissions from the use of LOHC depend on different conditions and should be calculated on a case-by-case basis. A comprehensive approach to calculate the corresponding CO_2 footprint of hydrogen will be based on the Delegated Act on GHG methodology. As of now, there is no international specific standard in place that defines in detail how GHG emissions have to be calculated.[62] However, ISO 19870 'Methodology for determining the GHG emissions associated with the production and transport of hydrogen' is developed covering explicitly LOHC (and other carriers) but still it is not mentioned in European legislation.

6. Outlook

The presented overview of LOHC-BT technology underlines its potential as a storage and transport vector for large-scale H_2-to-H_2 value chains that will be indispensable in future clean energy systems. However, the viability of the addressed aspects, parameters, and boundaries of LOHC-BT technology is strongly dependent on the emerging clean hydrogen market, which will remain highly dynamic in the near future. Parallel to the operation of first industrial-scale pilot plants that cover the full LOHC-BT H_2-to-H_2 value chain, the following fundamental aspects need to be clarified to accelerate the hydrogen economy even further: 1) price for renewable hydrogen; 2) implementation of regulatory framework; and 3) stimulating programs to invest in the hydrogen economy.

Obviously, all aspects are currently intensively developed and politics as well as industry push to make initial investments in the necessary infrastructure. Nevertheless, targets to enter a new era of our global energy system, by the year 2030, are ambitious and therefore full commitment of all involved parties is mandatory to reach this goal.

Acknowledgements

This work was funded by the TransHyDE-Projekt Helgoland Förderkennzeichen 03HY208 A-J.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

benzyltoluene, hydrogen storages, hydrogen transportation, LOHC

Received: August 29, 2023
Revised: November 9, 2023

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