



Review article

Low-carbon energy storage via liquid organic hydrogen carriers: From new developments to global initiatives



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ABSTRACT

Liquid organic hydrogen carriers (LOHCs) are a promising approach for long-term, low-carbon energy storage via hydrogen, due to their safety profile and compatibility with existing infrastructure. These properties also make it possible for them to efficiently transport and distribute large-scale hydrogen over long distances. This review explores recent advancements in LOHC technology, focusing on innovations that improve hydrogen storage capacity, techno-economics, and environmental impacts of LOHCs. Additionally, it examines global initiatives aimed at scaling LOHC systems for large-scale hydrogen storage and transport whilst highlighting collaborations, policy frameworks, and investment trends that support the integration of LOHC into renewable energy infrastructure. LOHC has the potential to play a critical role in achieving global decarbonization goals by facilitating the safe and reusable storage of low-carbon hydrogen. However, technological challenges persist, and more research is needed to maximize their overall thermal efficiency and market adoption.

1. Introduction

Energy is the cornerstone of global socio-economic development, yet the current energy landscape remains largely reliant on fossil fuels, despite their environmental impact and finite availability [1,2]. This situation has led to a growing consensus that achieving climate change mitigation goals and ensuring energy security requires not only enhanced energy efficiency but also a shift towards clean, renewable energy systems [3,4]. The intermittent and geographically variable nature of renewable sources like wind and solar power, however,

necessitates the development of robust energy storage solutions to facilitate their widespread adoption and integration into existing power grids [5,6]. The concept of a hydrogen-based economy has gained traction as a potential solution for achieving a sustainable, low-carbon energy future [7]. Moreover, hydrogen's abundance and widespread distribution align with the United Nations' Sustainable Development Goal 7, which aims to provide universal access to affordable, reliable, sustainable, and modern energy [8].

Hydrogen, particularly green hydrogen, has emerged as a promising clean energy carrier [9]. Its high gravimetric energy density (120 MJ/

Abbreviations: ADR, Agreement Concerning the International Carriage of Dangerous Goods by Road; ASME, American Society of Mechanical Engineers; AEMEL, Anion Exchange Membrane Electrolyzer; AEL, Alkaline Water Electrolyzer; AI, Artificial intelligence; BT, Benzyltoluene; CCUS, Carbon Capture, Utilization, and Storage; CEN, European Committee for Standardization; CH₂, Compressed hydrogen; DBT, Dibenzyltoluene; DESNZ, Department for Energy Security and Net Zero; DII, Desertec Industrial Initiative; DoH, Degree of hydrogenation; ETC, Energy Transition Commission; EU, European Union; GHG, Greenhouse gas; GHS, Globally Harmonized System of Classification and Labeling of Chemicals; GWP, Global warming potential; HPTSU, Hydrogen Production, Transportation, Storage, and Usage; HSC, Hydrogen Storage Capacity; HYS2, Low Hydrogen Supply 2; IPCEI, Important Projects of Common European Interest; ISO, International Organization for Standardization; LCHP, Low-carbon Hydrogen Production; LCOH, Levelized cost of hydrogen; LOHC, Liquid Organic Hydrogen Carriers; MCHRs, Microchannel reactors; NECZ, N-ethylcarbazole; NFPA2, National Fire Protection Association 2 Hydrogen Technologies Code; OSHA, Occupational Safety and Health Administration; PEMEL, Proton Exchange Membrane Electrolyzer; PEMFC, Proton Exchange Membrane Fuel Cells; PV, Photovoltaic; R&D, Research and development; REACH, Registration, Evaluation, Authorization, and Restriction of Chemicals; RID, Regulations concerning the international transport of dangerous goods by rail; SDG, Sustainable Development Goal; SMR, Steam Methane Reforming; SOEL, Solid Oxide Electrolyzer; SOFC, Solid Oxide Fuel Cells; TBR, Trickle bed reactor; TRL, Technological readiness

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kg or 33.3 kWh/kg) and clean oxidation process, which produces only water as a by-product, make it an attractive option for various applications, including stationary, mobile, and transportation sectors [10,11]. Due to its capacity for energy storage and transmission, hydrogen serves as a versatile energy carrier, enabling the integration of renewable energy sources and supporting the decarbonization of challenging sectors such as power generation, transportation, and industry [12]. The importance of carbon-neutral hydrogen production cannot be overemphasized in the context of the energy transition. Conventional hydrogen production methods, such as steam methane reforming (SMR), rely on fossil fuels and result in significant carbon dioxide emissions [13]. To truly harness the environmental benefits of hydrogen, it is crucial to develop and deploy technologies that enable the production of hydrogen from renewable sources or with minimal carbon footprint [14,15]. Various pathways for carbon-neutral hydrogen production have been explored, including water electrolysis powered by renewable electricity, biomass gasification, and methane pyrolysis [15,16]. These methods aim to decouple hydrogen production from fossil fuel dependence and contribute to the development of a sustainable hydrogen economy. Hydrogen can be converted or distributed in pure form by many pathways, ranging from compressed gas hydrogen in pipes to liquid hydrogen transported by ships, trains, or trucks, to synthesized fuel pathways. The main drawback of hydrogen compression is its low volumetric storage density, which involves high-pressure containment systems, though some energy is required to compress hydrogen. Liquefaction is considerably more energy-intensive due to the extreme cooling to cryogenic temperatures.

Water electrolysis powered by renewable electricity represents a promising pathway for sustainable H_2 production, as illustrated in Fig. 1 [18–21]. The generated hydrogen can be efficiently converted into stable chemical carriers, facilitating energy storage and transportation across varying distances [22]. Among various hydrogen carriers, including metal hydrides [23] and ammonia [24–26], liquid organic hydrogen carriers (LOHCs) have emerged as a practical solution due to their high storage capacity, low toxicity, and simplified handling requirements [21,23–25]. LOHCs represent an innovative approach to hydrogen storage and transportation, operating through a reversible catalytic hydrogenation/dehydrogenation cycle [27,28]. The technology encompasses 2 fundamental processes: hydrogenation, where hydrogen gas bonds chemically to LOHC molecules under catalytic conditions, to form hydrogenated LOHC (H-LOHC); and dehydrogenation, where hydrogen is released from H-LOHC for end-use applications [29,30]. This closed-loop system maintains the structural integrity of LOHC molecules throughout multiple cycles, enabling their continuous reuse as storage media [31]. Unlike conventional storage methods, LOHCs offer distinct advantages, including ambient condition storage capabilities and enhanced volumetric energy density compared to

compressed or liquefied hydrogen [32,33]. The system's compatibility with existing liquid fuel infrastructure significantly reduces implementation costs and safety concerns associated with high-pressure systems [34]. Furthermore, LOHCs demonstrate remarkable stability during long-term storage without energy loss and minimal boil-off compared to liquefied hydrogen [35]. The technology's efficiency is underpinned by rapid, reversible reactions that theoretically yield pure hydrogen during dehydrogenation, while the carrier molecules remain unchanged [36]. As illustrated in Fig. 2, this closed-loop material cycle enhances system sustainability and operational efficiency, positioning LOHCs as a promising solution for large-scale hydrogen storage and transportation applications [32,33].

Given the multifaceted nature of LOHCs in enabling a carbon-neutral hydrogen economy, existing reviews tend to adopt a narrower scope, concentrating specifically on singular aspects of LOHC technology rather than providing a comprehensive overview. These specific publications focus on either the advancements in LOHC materials and catalyst development [22,36–41], or examine only implementation projects [42,43] and a comparative analysis against alternative hydrogen storage technologies [27,34,44,45], though 2 elaborated reviews were identified and are differentiated from ours in Table 1. Li et al. [43] discuss the various LOHCs instituted in various countries across the globe, with more emphasis on the funding sources, transport modalities, supply chains, and implemented technologies. The paper concludes that a detailed understanding and evaluation of these projects would open doors for large-scale production and long-distance transportation of hydrogen. In another review [46], the author discusses extensively the various existing hydrogen storage alternatives and compares them with LOHCs. They also reported findings from selected studies wherein various catalyst performances were investigated, and suggest that further studies are needed to promote LOHCs to hydrogen storage industrial standards. Other important considerations, such as the environmental implications of LOHCs implementation, were not emphasized in their study. More recently, Munyentwali et al. [47] presented some updates in the LOHCs technology, such as renewable LOHCs from lignocellulosic biomass and LOHCs production from plastic waste recycling.

As opposed to previous works, this article is designed as a comprehensive, integrative resource that not only discusses materials and catalytic performance but also expands the lens to include reactor designs, policy frameworks, national and international initiatives, environmental and economic impacts, innovation gaps, and proposed solutions to facilitate large-scale deployment. This broader perspective is intended to guide the development of LOHC technology while serving the needs of both new and established researchers in the field. Moreover, the field of LOHC has been developing rapidly in recent years, driven by the global push toward decarbonization, the need for

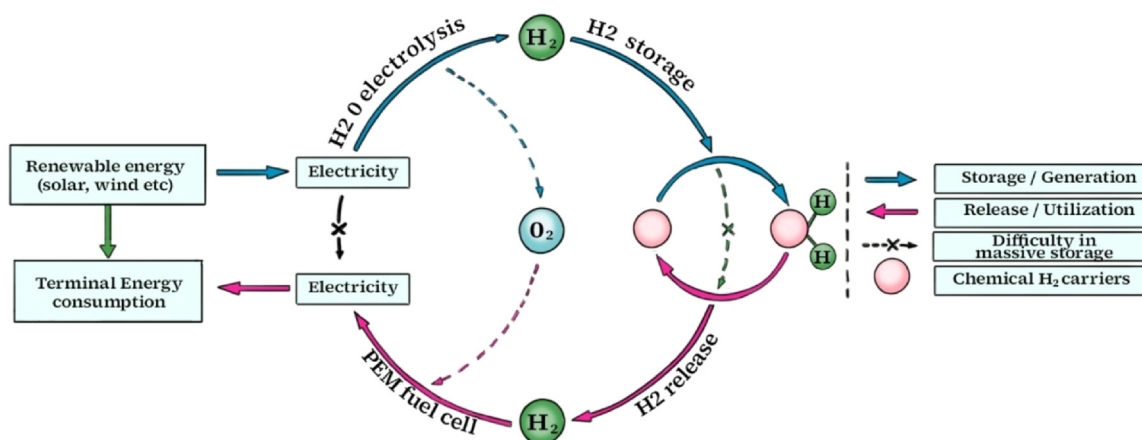


Fig. 1. Renewable energy storage and utilization via hydrogen and hydrogen carriers [17].

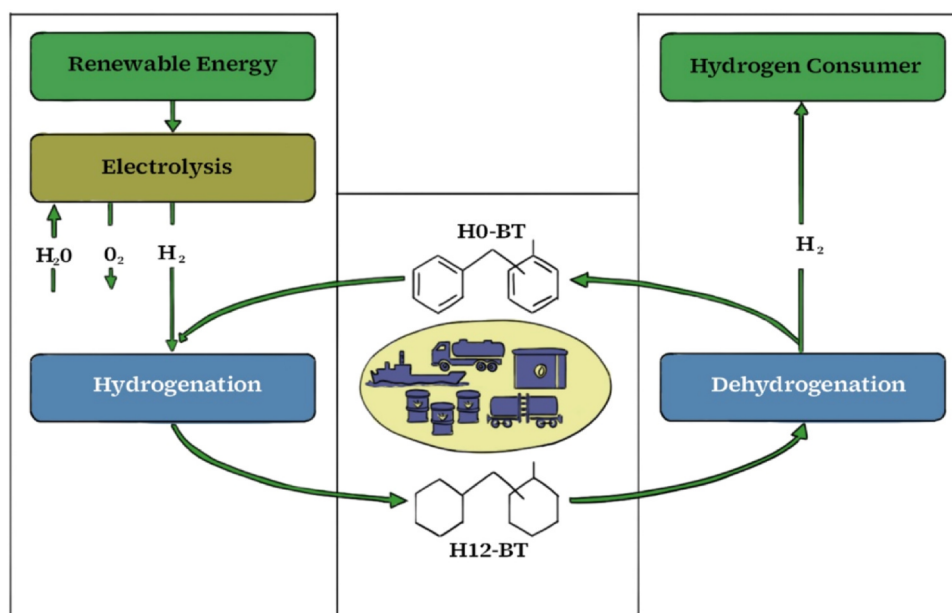


Fig. 2. Schematic representation of the LOHC cycle for renewable energy storage and distribution. LOHC = liquid organic hydrogen carrier.

scalable energy storage solutions, and growing investment in clean energy infrastructure.

2. Principle of LOHCs technology

LOHCs allow for hydrogen storage in a liquid form at ambient conditions through reversible hydrogenation and dehydrogenation reactions. For storage purposes, newly produced hydrogen chemically bonds to the LOHC molecules in a hydrogenation reaction. Later, when the stored hydrogen is required for use, the dehydrogenation process breaks these chemical bonds between the hydrogen and LOHC molecules to release pure hydrogen gas while regenerating the original LOHC compound for future storage cycles. Fig. 3 displays the schematic of the hydrogenation-dehydrogenation cycle for hydrogen storage using LOHC materials. During hydrogenation, hydrogen gas binds ($+xH$) to LOHC through a catalytic reaction at moderate temperatures and elevated pressure, releasing heat as a byproduct. In dehydrogenation, heat input is required to break the hydrogen-LOHC (Hx -LOHC) bonds, releasing hydrogen ($-xH$) for the required applications while the hydrogen-lean LOHC is recycled.

2.1. Hydrogenation process

Hydrogenation in the context of hydrogen storage using LOHCs involves the addition of hydrogen molecules to unsaturated organic compounds to form a stable, hydrogen-rich product. This process begins with the introduction of hydrogen gas to a carefully selected unsaturated organic compound that serves as the LOHC. The reaction is exothermic and takes place in the presence of a catalyst, commonly transition metals such as nickel, platinum, palladium [49]. The catalyst works by adsorbing the hydrogen molecules, where the H-H bond is cleaved at the active metal site, leading to the formation of 2 hydrogen atoms ($2H^*$) bound to the surface of the catalyst. An unsaturated LOHC molecule is also adsorbed onto the catalyst surface. The combined steps involve the addition of 1 hydrogen atom to one of the carbon atoms of the unsaturated bond, forming a half-hydrogenated intermediate. This step is often reversible and can lead to isomerization in certain cases. The second hydrogen addition then completes the hydrogenation, saturating the bond and forming the product molecule. For more complex organic compounds, such as aromatics or polycyclic structures, the mechanism can involve multiple steps and intermediates [50].

Advancements in computational chemistry and in-situ characterization techniques have allowed for more detailed mechanistic studies to improve understanding of these hydrogenation mechanisms. These have revealed subtleties in the reaction pathways, such as the influence of LOHC compound structure and orientation on the catalyst surface [51–57] and the role of surface defects or additives [58–61].

Sotoodeh et al. [62] reported that the hydrogenation of N-ethylcarbazole (NEC) proceeds through a complex reaction mechanism involving 2 main parallel pathways: one leading to the formation of tetrahydro-N-ethylcarbazole, quickly converting to hexahydro-N-ethylcarbazole, and the other directly forming octahydro-N-ethylcarbazole. This parallel pathway is a characteristic mechanism in hydrogenation reactions similar to those observed in the hydrogenation of aromatic compounds. The hydrogenation mechanism of NEC has been extensively investigated due to its relevance as a benchmark LOHC system. Fig. 4 schematically illustrates the reaction pathway for NEC hydrogenation over Ru-based catalysts, highlighting the formation of tetrahydro- and hexahydro-NEC intermediates and their subsequent conversion to fully hydrogenated octahydro-NEC through parallel hydrogenation routes.

In addition to meeting the requirements of reversible thermodynamics, fast kinetics of hydrogenation is also important for LOHC [63]. The efficiency and rate of the hydrogenation process can be influenced by the type of LOHC used, the properties of the catalyst, and reaction conditions such as temperature and pressure [64]. The impacts of temperature, pressure, degree of hydrogenation (DoH), and catalyst amount on the hydrogenation rate of dibenzyltoluene (DBT) over a Ru/ Al_2O_3 catalyst are illustrated in Fig. 5.

Fig. 5(a) shows that increasing the catalyst mass in the reactor leads to an increase in the reaction rate, but only up to a certain point. This is because the hydrogenation reaction occurs on the catalyst surface, so more catalyst provides more reaction sites. However, the effect plateaus at catalyst mass ratios of 3% or more, where the reaction rate stabilizes between 59.9 and 60.8. Moreover, the hydrogenation reaction is exothermic, and an excessive amount of catalyst could generate more heat than can be efficiently dissipated. This localized temperature increase may lead to thermodynamic limitations, shifting the reaction equilibrium unfavorably towards the reactants and reducing overall conversion efficiency. Such thermal effects could be particularly pronounced in reactors with limited heat exchange capabilities. Another potential factor for the catalyst behavior could be mass transport limitations. As

Table 1
Comparison of this review with recent LOHC literature

Refs.	Focus area	Policy & global strategy	Life-cycle assessment (LCA)	Economics	Comparative Scope
[46]	LOHC development and infrastructure, including economics	No	No	Moderate	Medium
[48]	LOHCs practical uses and developments in biomass-based LOHCs	No	No	No	Narrow
This Work	Comprehensive (Techno-economic, LCA, policy, global initiatives)	Yes	No	Extensive	Wide

LOHC = liquid organic hydrogen carrier.

catalyst concentration increases, diffusion effects could start to dominate, restricting the availability of hydrogen at the catalyst surface. This could lead to an observed plateau in reaction rate not solely due to site saturation but also due to constraints in hydrogen supply. Fig. 5(b) and (c) demonstrate that the reaction rate decreases as hydrogenation progresses and DoH increases. This is due to a decrease in the driving force of the reaction. As hydrogenation progresses, the difference between the DBT concentration and the equilibrium concentration decreases. This reduction in concentration gradient slows down the hydrogenation reaction rate. Fig. 5(b) and (c) also show that higher pressures and increasing temperature up to the range studied (130–170 °C) lead to faster reaction rates at the same DoH. However, it is important to note that above certain temperatures, the reverse reaction, dehydrogenation, becomes thermodynamically favored [66]. This is because hydrogenation is exothermic and favored at lower temperatures, while dehydrogenation is endothermic and becomes more favorable at elevated temperatures.

2.2. Dehydrogenation process

During the dehydrogenation process, hydrogen is released from the hydrogen-rich LOHC in the presence of a catalyst. For instance, the dehydrogenation of LOHCs over heterogeneous catalysts follows a stepwise mechanism that begins with the adsorption of the hydrogen-rich LOHC molecule onto the active metal sites of the catalyst. Yang et al. [67] described the dehydrogenation of perhydro-N-ethylcarbazole (12H-NECZ), over various catalysts, including Pd, Pt, Ru, and Rh. The reaction proceeds via 2 intermediates: octahydro-N-ethylcarbazole (8H-NECZ) and tetrahydro-N-ethylcarbazole (4H-NECZ). Initially, 12H-NECZ releases 2 hydrogen molecules to form octahydro-N-ethylcarbazole (8H-NECZ). Subsequently, 8H-NECZ undergoes further dehydrogenation, releasing 2 additional hydrogen molecules to generate tetrahydro-N-ethylcarbazole (4H-NECZ). In the final phase, 4H-NECZ releases the remaining 2 hydrogen molecules to produce the fully dehydrogenated NECZ. Under optimized reaction conditions, this stepwise dehydrogenation can proceed to completion, with the intermediate species (8H-NECZ and 4H-NECZ) being consumed as the reaction progresses toward the final product. These reaction conditions and catalyst choice significantly influence the relative concentrations of these intermediates during the dehydrogenation process. The dehydrogenation of hydrogen-rich LOHCs proceeds through a well-defined stepwise mechanism governed by catalyst type and reaction conditions. Fig. 6 illustrates the sequential dehydrogenation pathway of perhydro-N-ethylcarbazole (12H-NECZ), showing the progressive release of hydrogen via octahydro- (8H-NECZ) and tetrahydro-N-ethylcarbazole (4H-NECZ) intermediates prior to regeneration of N-ethylcarbazole.

Naseem et al. [69] studied the effect of different reaction conditions, including temperature and pressure on the dehydrogenation performance of different LOHC materials. They found that lower pressures enhance dehydrogenation by shifting the equilibrium favorably. Fig. 7(a) shows the equilibrium conversions across 50–400 °C at varying pressures, for the dehydrogenation of perhydrodibenzyltoluene. Their research also demonstrated that while higher temperatures favor this endothermic dehydrogenation process, exceeding optimal temperature conditions results in increased energy usage, as seen in Fig. 7(b).

2.3. LOHC materials

The efficiency of hydrogen storage in LOHC systems depends on the properties of the LOHC materials used. The choice of LOHC material directly affects the hydrogen capacity – how much hydrogen can be stored per unit of the LOHC, reaction conditions, and the energy consumed during the hydrogenation-dehydrogenation processes. LOHC systems should remain liquid over a wide temperature range, characterized by a low melting point (so they stay liquid at low temperatures) and a high boiling point (to avoid vaporization at high

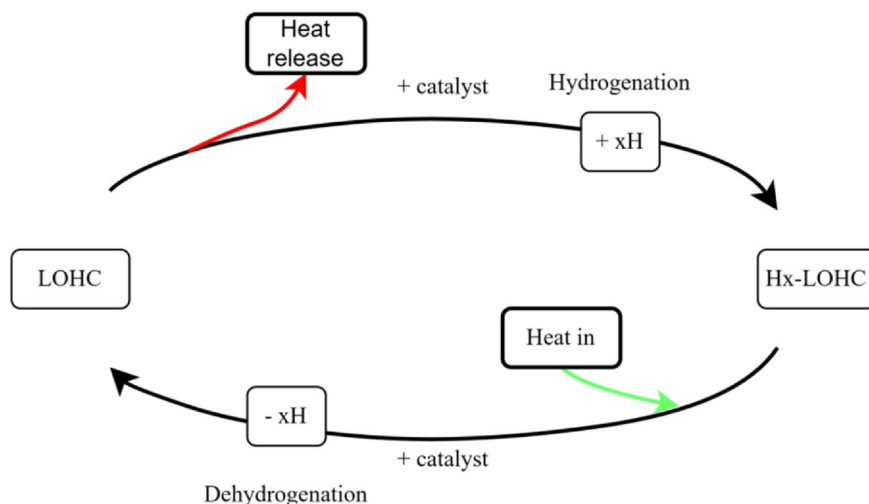


Fig. 3. Hydrogenation-dehydrogenation reaction steps in LOHCs technology. LOHCs = liquid organic hydrogen carriers.

temperatures) [63]. Based on their chemical structure and functionalities, LOHC compounds have been broadly categorized in this paper into: homocyclic compounds, heterocyclic compounds, and functional group-based compounds such as nitrogen-based (nitriles, imides, amides, amine-boranes), oxygen-containing (ketone/alcohol pairs, ester/alcohol systems), and silicon-based (silane/alcohol pairs, organosilanes) systems. The carbonyl groups present in some of these carriers exhibit varying hydrogenation tendencies based on their polarizability, which impacts how easily they accept electrons during the reaction. [70]. This reactivity follows a trend from aldehydes being the most easily hydrogenated to ureas being the least (aldehyde < ketone < anhydride < imide < ester < amide < carbamate < urea). For dehydrogenation, substituting C atoms with B and N atoms has been reported to enhance dehydrogenation efficiency by weakening the adjacent C-H bond through electronic effects and changes in molecular

orbital energies, leading to reduced activation barriers for hydrogen release [71].

Aldehydes, with less steric hindrance and higher electron deficiency, are more reactive than ketones. Recently, Schwaab et al. [72] investigated the dehydrogenation behavior and thermal stability of the LOHC pair benzaldehyde/cyclohexylmethanol on a platinum (Pt(111)) catalyst. Using in situ synchrotron radiation photoelectron spectroscopy and temperature-programmed desorption, they observed that this LOHC system stores hydrogen in a cyclohexyl group and an alcohol functional group, achieving a hydrogen storage capacity (HSC) of 7.0%. However, during the dehydrogenation of the cyclohexyl group (between 250 and 350 K), the stability issues arose as the resultant aldehyde decomposed. Comparing cyclohexylmethanol to a related compound, 1-cyclohexylethanol (a secondary alcohol with 6.3% hydrogen storage), they found that 1-cyclohexylethanol displayed better thermal stability.

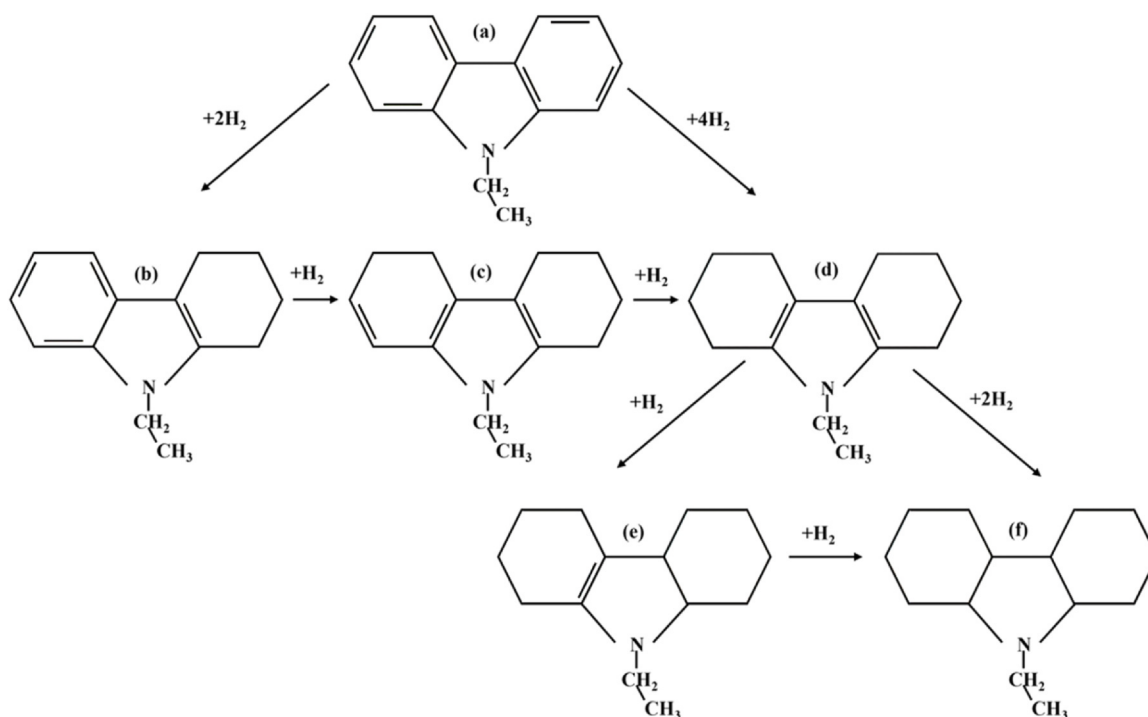


Fig. 4. Reaction pathway for the hydrogenation of N-ethylcarbazole (NEC) over Ru-based catalysts [62].

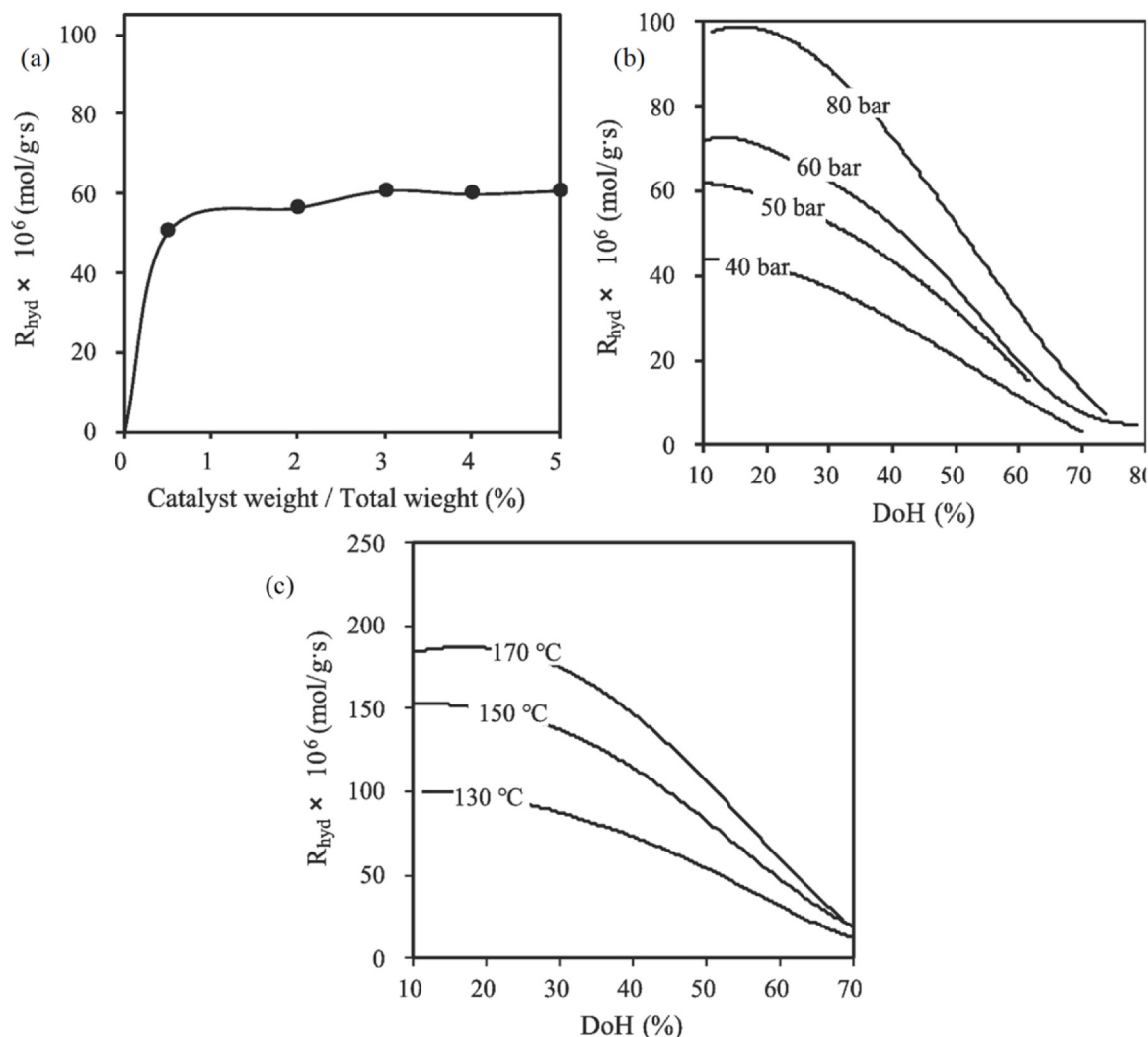


Fig. 5. Hydrogenation rate of dibenzyltoluene over a Ru/Al₂O₃ catalyst at varying (a) catalyst weights (130 °C, 50 bar), (b) pressures and DoH (130 °C), and (c) temperatures and DoH (80 bar) [65].

Conversely, anhydrides and imides are high up the reactive trend due to the electron-withdrawing effect of adjacent carbonyl groups. Moving towards esters and amides, electron-donating groups like oxygen or nitrogen reduce the electrophilicity of the carbonyl carbon, making hydrogenation increasingly difficult. The hydrogenation of esters yields alcohols, while amides can yield either amines or more complex nitrogen-containing compounds. Ventura-Espinosa et al. [73] evaluated the primary amine/nitrile pair as a potential LOHC, with ruthenium complexes catalyzing the dehydrogenation of primary amines to nitriles, releasing hydrogen under mild conditions without additives. Though dehydrogenation of amines to nitriles has been shown in experimental works, the reaction was usually carried out under a nitrogen atmosphere. This arrangement essentially reduces the hydrogen partial pressure and thus mimics vacuum conditions thermodynamically conducive for hydrogen release. But in industrially

applicable conditions, where hydrogen is accumulated in the reaction medium, the process ceases to be as favorable. As highlighted in recent studies [74], this limitation makes the technical application of hydrogen release from nitrile-type LOHCs on a large scale very challenging. Therefore, while this reaction type is an interesting field of study, its use as a viable LOHC system is questionable. A graphene-supported ruthenium catalyst showed strong stability and reusability, remaining active over 10 cycles without significant loss of performance, making it promising for practical applications. A major challenge is benzylamine dehydrogenation selectivity, as transamination can lead to imine by-products. Although this issue was addressed by using long-chain amines like dodecylamine, which improved selectivity.

Imides can lead to several possible products upon hydrogenation, depending on the specific pathways facilitated by the catalyst. This variability demands precise control over the hydrogenation process

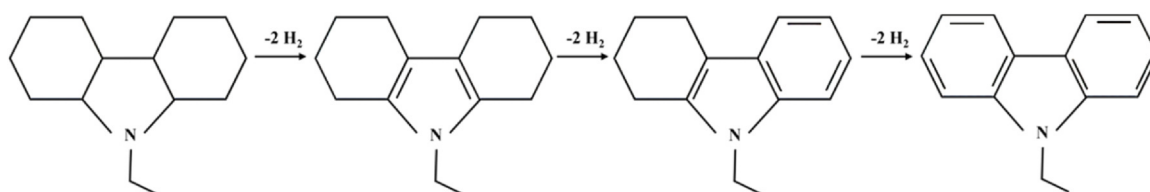


Fig. 6. Stepwise dehydrogenation mechanism of perhydro-N-ethylcarbazole (12H-NECZ) showing sequential hydrogen release via 8H- and 4H-NECZ intermediates [68].

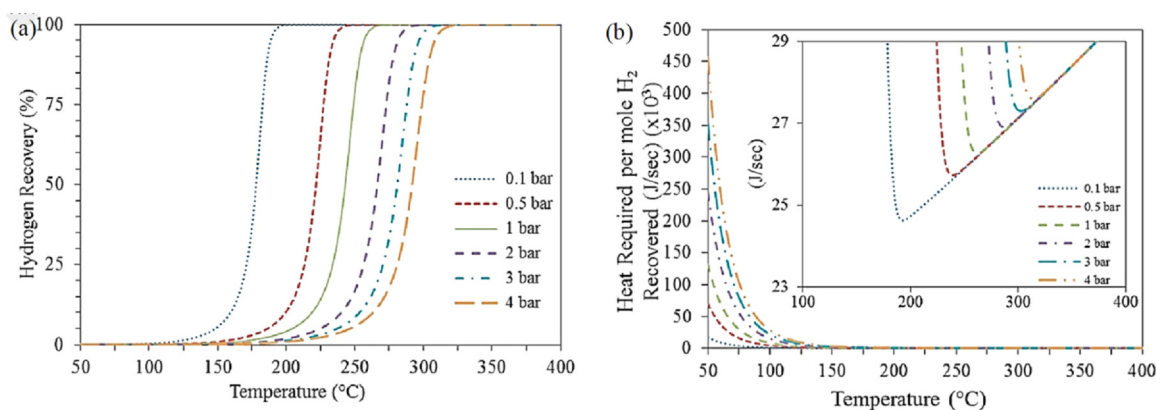


Fig. 7. (a) The equilibrium conversion of perhydridibenzyltoluene dehydrogenation at different pressures and temperatures. The results represent the maximum thermodynamically achievable hydrogen recovery under the given conditions, not the experimentally determined hydrogen recovery. (b) Heat energy required per second to release 1 mol of hydrogen during the dehydrogenation of perhydridibenzyltoluene over Pt/Al₂O₃ catalyst [69].

[75]. Thus, to achieve selective hydrogenation of imides, it is important to choose catalyst complexes with specific properties that guide the reaction toward the desired product [76–79]. Dehydrogenating these compounds often demands high energy due to the stability of the reduced bonds, especially with imides and amides, which are characterized by strong carbon-nitrogen bonds. This stability, combined with potential side reactions that can occur during dehydrogenation, makes it difficult to efficiently release stored hydrogen.

Carbamates and ureas are the most resistant to hydrogenation due to strong resonance and electron-donating effects from their nitrogen atoms. Balaraman et al. [80] reported the first-ever catalytic hydrogenation of urea derivatives to their corresponding amines and methanol. Using a bipyridine-based PNN Ru(II) pincer complex, they achieved a successful catalytic conversion of ureas to amines and methanol under relatively mild conditions (110 °C, 10–13.6 atm H₂). Their system worked effectively across a broad substrate scope, from simple alkyl ureas to more challenging aryl and sterically hindered variants, and revealed that ureas represent the most challenging carbonyl-containing compounds for hydrogenation, due to strong resonance stabilization. Despite this difficulty, they achieved yields of up to 90% methanol and 95% amine yields for compounds like 1,3-diphenylurea. Through mechanistic studies, they demonstrated a stepwise hydrogenation pathway proceeding through formamide intermediates. Table 2 presents the properties of the different conventional and novel LOHC materials reported in the literature.

2.4. Catalysts for LOHCs

In LOHC systems, catalysts are required both for hydrogenation and dehydrogenation processes. These catalysts need to offer high activity, selectivity, and stability while operating under mild conditions. LOHC catalysts can be homogeneous or heterogeneous, with each having its own strengths and limitations.

Homogeneous catalysts operate in the same phase as the LOHC reactants and have been widely explored for dehydrogenation reactions in research. However, their practical application is often limited by challenges in catalyst separation and recovery, as well as thermal stability issues. Novel approaches, such as catalyst immobilization in biphasic systems, have been investigated to address these limitations, but widespread technical application remains a challenge. Transition metal complexes such as ruthenium, rhodium, and iridium are highly active homogeneous catalysts employed in LOHC systems. Ruthenium-based complexes have been well investigated for the hydrogenation of ester/alcohol, aromatic ketones, and nitrile-based compounds used in LOHCs [126–130]. Rhodium-based complexes, although more expensive, can operate at milder conditions and have shown catalytic activity in the hydrogenation of esters and ketones [131–133]. Iridium-based

complexes have also been explored. The Jensen group [134] established iridium pincer complexes as highly effective catalysts for the dehydrogenation of numerous potential LOHCs, including perhydro-dibenzofuran, perhydro-indole, N-methyl perhydro-indole, 4,4'-bipiperidine, 4-amino-methylpiperidine, and aminomethylcyclohexane. These catalysts are characterized by a tridentate pincer ligand coordinated to the iridium center, which provides stability while maintaining high activity. Their effectiveness stems from the careful balance of electronic and steric properties provided by the pincer ligands, which can be fine-tuned through ligand modification. Other Iridium-type complexes with different ligand sets have also been developed and reported to show excellent activity with indoles, amine-boranes, ketones, N-heterocyclic LOHCs like quinolines [135–139].

Homogeneous catalysts face practical limitations in LOHC applications, including challenges with catalyst separation and recovery, and low thermal stability [140]. More recently, Søgaaard et al. [141] proposed a novel approach to enhance catalyst stability and separation in LOHC systems by immobilizing cationic iridium complexes within a molten salt-liquid biphasic system. The Crabtree-type catalyst, [Ir(cod)(py)(PCy₃)]PF₆, was immobilized in a molten salt medium—tetraphenylphosphonium bis(trifluoromethylsulfonyle)imide ([PPh₄][NTf₂])—and employed for hydrogenation/dehydrogenation of the 2-methylindole/2-methylindoline LOHC pair. To facilitate phase separation, they incorporated dibutyl ether into their design. Their system operates at relatively mild temperatures (below 140 °C), just above the melting point of the salt, with the presence of the ionic catalyst and stabilizing ligand further reducing the melting point to below 120 °C. The system they developed enables continuous operation and easy catalyst/LOHC separation, allowing for the release of substantial quantities of hydrogen while using minimal amounts of precious metal catalyst.

Heterogeneous catalysts involve metal particles on a solid support and offer advantages in terms of stability, ease of separation, and reusability. Ruthenium, palladium, platinum, nickel, and cobalt are common metals used in heterogeneous catalysts. These metals are often supported on materials like alumina, silica, or carbon to maximize surface area and catalytic efficiency. Supported Ru, Pd, and Pt-based catalysts are among the most studied for LOHC systems hydrogenation/dehydrogenation reactions, due to their efficiency as well as their reactivity and stability over repeated cycles [142]. Eblagon et al. [143] examined the hydrogenation of 9-ethylcarbazole (NEC) in its molten state over various noble metal and nickel-based catalysts. Their experiments highlighted that the rate of NEC conversion and the selectivity of the products were influenced by both the metal type and the catalyst support material. On an activated carbon support, the metals ranked in catalytic activity as follows: Ru was the most effective, followed by Pd, Pt, and Ni. Among the Ru-based catalysts tested with

Table 2
Summary of physical properties of conventional and potential LOHC materials investigated in literature

H ₂ -lean LOHC	H ₂ -rich LOHC	Theoretical HSC (wt%)	Melting point (°C) at atm (H ₂ -lean)	Melting point (°C) at atm (H ₂ -rich)	Boiling point (°C) at atm (H ₂ -lean)	Boiling point (°C) at atm (H ₂ -rich)	Dehydrogenation enthalpy at standard conditions (298.15 K, 1 atm) (kJ/mol H ₂)	Refs.
Benzene	Cyclohexane	7.19	5.53	6.47	80.1	80.74	65.3	[81]
Naphthalene	Decalin	7.29	79	-30.4 (trans), -42.9 (cis)	218	187 (trans), 196 (cis)	63.9	[38]
1-(N-phenylethyl) naphthalene (PEN)	Perhydro-PEN	6.49	NA	NA	350 >	339.7	62.4 (cis)/ 64.2 (trans)	[82]
Toluene derivatives								
Toluene	Methylcyclohexane (MCH)	6.16	-95	-126	111	101	68.3	[83]
Benzyltoluene	perhydro benzyltoluene (H12-BT)	6.20	-30	-80-70	277-290	264-272	63.5	[83]
Dibenzyltoluene	perhydro dibenzyltoluene (H18-DBT)	6.29	-34	< -50	392	371	65.4	[83]
Phenyl derivatives								
Biphenyl	Bicyclohexyl	7.27	69-72	4	255	227	371.14	[84], [71]
BN-biphenyl	BN-bicyclohexyl	7.20	NA	NA	NA	NA	176.45	[71]
Terphenyl	Perhydroterphenyl (PHT)	7.2	58-213	16-48 (cis), 47-164 (trans)	337-389	300-350	66.1	[85-87]
Carbazole derivatives								
N-ethylcarbazole (NEC)	Dodecahydro-N-ethylcarbazole	5.8	70	-85	166	ca. 295	50.6	[88]
N-Carbazole	Dodecahydrocarbazole	6.7	246.3	NA	354.69	272.9 ± 8	51	[89]
Quinoxaline derivatives								
Phenazine	Tetradecahydrophenazine	7.3	176	NA	360	316.4 ± 10	61.3	[22]
Quinoxaline	Tetrahydroquinoxaline	7.2	55	92 - 98	237.6	289	59.9	[90,91]
2,6-dimethyl-1,5-naphthyridine	2,6-dimethyldecahydro-1,5-naphthyridine	6.0	NA	NA	NA	NA	58	[92,93]
2,5-dimethylpyrazine	2,5-dimethylpiperazine	5.3	15	113-118	155	162-165	55	[93,94]
Quinoline derivatives								
Quinoline (QL)	Decahydroquinoline	7.24	-40	48	234	201	61.9	[95]
2-MethylQL	2-Methyl tetrahydroquinoline (THQ)	6.6	-9	NA	247.6	NA	59.5	[96,97]
4-MethylQL	4-MethylTHQ	< 7.2	10	NA	261 - 263	NA	60.2	[98]
Pyridine derivatives								
Pyridine	Piperidine	7.1	-42	-7	115	106	61.8	[99]
2-Methylpyridine	2-methylpiperidine	6.1	-70	-5	129	119	59.9	[100]
2,6-dimethylpyridine	2,6-dimethylpiperidine	5.3	-6	-63	144	126-139	55.8	[100]
2-(N-methylbenzyl)pyridine (MBP)	2-[(n-methylcyclohexyl)-methyl]piperidine isomers (H12-MBP)	6.15	-50.1 to -46.2	-19.3 to -18.0	291-293	291-293	66.7	[101]
Pyrrole derivatives								
Butyl pyrrole	Butyl pyrrolidine	3.14	-23	NA	170.50	154.5-157.3	88.9	[102]
Indole derivatives								
Indole	Octahydroindole/ Octahydroindoline	4.7-6.7	51 - 54	NA	253	NA	48.6-57.5	[103]
2-methylindole	Octahydro-2-methylindoline	6.4	56-90	NA	273	NA	57.5	[104]
1-methylindole	Octahydro-1-methylindole	5.8	NA	NA	237 - 239	NA	53.5	[104]
Amides								
Glycine anhydride (Cyclic dipeptide)	2-aminoethanol (AE)	6.56	300	10	573.6 ± 43	170	56.7	[105]
N,N'-(ethane-1,2-diyl)bis(N-methylformamide)	Methanol + N,N'-dimethylethylenediamine (DMEDA)	5.26	NA	NA	365.4 ± 25	NA	NA	[106,107]
N,N'-(ethane-1,2-diyl)-diformamide	Ethylenediamine (EDA) + Methanol	6.45	NA	NA	478.7 ± 28	NA	NA	[108]
N,N'-(ethane-1,2-diyl)diacetamide	EDA + Ethanol	5.26	175	NA	438.7 ± 28	NA	NA	[109]
Ethylene Urea	EDA + Methanol	6.52	129 - 132	NA	358.7 ± 11	NA	NA	[110]

(Continued on next page)

Table 2 (continued)

H ₂ -lean LOHC	H ₂ -rich LOHC	Theoretical HSC (wt%)	Melting point (°C) at atm (H ₂ -lean)	Melting point (°C) at atm (H ₂ -rich)	Boiling point (°C) at atm (H ₂ -lean)	Boiling point (°C) at atm (H ₂ -rich)	Dehydrogenation enthalpy at standard conditions (298.15 K, 1 atm) (kJ/mol H ₂)	Refs.
Imides								
Bis-cyclic imide	1,4-butanediol + EDA	6.66	NA	NA	NA	NA	NA	[111]
Nitriles								
Benzonitrile	Benzylamine	> 7	-13	10	191	185	> 67	[112,113]
n-Octanenitrile	n-Octylamine	> 7	-45	-5 – -1	198 – 200	175 – 177	> 67	[114,115]
Cyclic amine-boranes								
Aniline-borane trimer	1,2-BN-cyclohexane	9.4	NA	63	NA	NA	NA	[116]
Aniline-borane trimer	BN-methylcyclopentane	4.7 (demonstrated)	28–30	-18	378	335	NA	[117]
Carbamates								
Octyl Phenylcarbamate	Aniline + Methanol + Octanol	ca. 5.3	NA	NA	317.6 ± 11	NA	NA	[118]
Benzyl carbamate	Benzylamine + Methanol + Benzylalcohol	ca. 5.3	85–89	NA	318.7 ± 21	NA	NA	[119]
Organosilanes								
Organosilane	Silane + Methanol	5.0	NA	NA	NA	NA	NA	[120]
Organosilane	1,4-disilabutane + methanol	4.3	NA	NA	NA	NA	NA	[121]
Organosilane	Disilamide + Methanol	4.8	NA	NA	NA	NA	NA	[122]
Ketones								
Acetophenone	1-Cyclohexylethanol	6.3	20.2	NA	202	189	65	[123]
Benzophenone	Dicyclohexylmethanol	7.19	47 – 51	58–64	305	318.3	67.6	[124]
Esters								
Liquid oligoesters	Ethylene glycol	6.5	NA	NA	NA	NA	76.99–116	[125]

HSC = hydrogen storage capacity; LOHC = liquid organic hydrogen carrier.

different supports, a catalyst consisting of 5 wt% Ru on alumina (Al_2O_3) exhibited the highest activity.

Pd-based catalysts have also demonstrated excellent performance, particularly in the dehydrogenation of heteroaromatic LOHCs under mild temperatures [144]. However, in the dehydrogenation of homocyclic compounds such as perhydrodibenzyltoluene (H18-DBT), Pd-based $\text{Pd}/\text{Al}_2\text{O}_3$ showed relatively poor performance. Modisha et al. [145] evaluated the catalyst activity of the dehydrogenation of H18-DBT using $\text{Pt}/\text{Al}_2\text{O}_3$, $\text{Pd}/\text{Al}_2\text{O}_3$, and $\text{Pt-Pd}/\text{Al}_2\text{O}_3$ catalysts. At 320°C, the 1 wt% $\text{Pt}/\text{Al}_2\text{O}_3$ achieved 90% degree of dehydrogenation, while increasing the Pt loading to 2 wt% improved performance to 96% DoD. In contrast, Pd-based catalysts showed poor performance, with $\text{Pd}/\text{Al}_2\text{O}_3$ reaching only 11% DoD. Surprisingly, the bimetallic Pt-Pd combination performed even worse, achieving just 6% DoD, demonstrating that palladium hindered the reaction rather than enhancing it.

Recent research is focused on developing bi/trimetallic catalysts, where the addition of a second metal can enhance activity and stability. Ru-Pd bimetallic catalysts have shown improved performance in the hydrogenation/dehydrogenation of N-heterocyclic compounds compared to monometallic alternatives [146,147]. Conversely, the role of the support material extends beyond simply providing a high surface area for metal dispersion. The properties of these supports can significantly influence catalyst performance. For instance, the addition of basic promoters to alumina supports can enhance the dehydrogenation activity of Pt catalysts by facilitating the removal of hydrogen from the carrier molecule. A clear ranking of catalyst support efficacy has been reported: Al_2O_3 performed best, followed by TiO_2 , $\text{SiO}_2\text{-Al}_2\text{O}_3$, zeolite, graphite, and activated carbon [148]. Advantages of heterogeneous catalysts include easy separation from reaction products and excellent

thermal stability at high operating temperatures [149]. However, they may have lower selectivity compared to homogeneous catalysts, which can lead to reduced efficiency and unwanted byproducts.

Table 3 presents the catalytic performance of hydrogenation and dehydrogenation reactions in various LOHC systems. For dehydrogenation, $\text{Pt}/\text{Al}_2\text{O}_3$ and Pd-based catalysts demonstrate high conversion efficiencies, with NEC achieving 100% conversion at 180°C using $\text{Pd}/\text{Al}_2\text{O}_3$. Bimetallic catalysts (Pd-Ni, Pd-Au, Pt-WOx) enhance selectivity and reduce activation energy. For hydrogenation, Ru-based catalysts ($\text{Ru}/\text{Al}_2\text{O}_3$, Ru/TiO_2) show high yields at lower temperatures (130–170°C). Nickel catalysts (Ni/AlSiO-1, Raney-Ni) offer cost-effective alternatives. The results highlight the role of catalyst composition, reaction temperature, and pressure in optimizing LOHC performance, which is essential for scalable hydrogen storage solutions.

2.5. Reactor design

Hydrogenation/dehydrogenation in LOHCs are catalytically driven process; therefore, the design of reactors is important in ensuring reactant contact and, invariably, the system's performance, scalability, and operational costs. In this paper, these reactors for LOHC systems have been divided into concepts based on the mode of operation – batch or continuous, and operational strategy – process intensification.

2.5.1. Batch catalytic reactors

Batch reactors have served as the primary experimental apparatus for studying LOHC (de)hydrogenation reactions, as they provide precise control over reaction conditions and detailed kinetic analysis [161–164]. These batch reactors are usually equipped with stirring

Table 3
Performance of catalyst systems in (de)hydrogenation of some leading LOHCs

H_2 -lean LOHC	Catalysts	T (°C)	P (MPa)	Time (h)	LOHCs Conv (%)	Yield (%)	Refs.
Dehydrogenation performance							
Dibenzyltoluene (DBT)	1 wt% $\text{Pt}/\text{Al}_2\text{O}_3$	290	0.1	14	-	91	[30]
	5 wt% $\text{Pt}/\text{Al}_2\text{O}_3$	290		7	-	90.2	[150]
	0.3 wt% $\text{Pt}/\text{Al}_2\text{O}_3$	240	0.1	4	-	58.2	[151]
	$\text{Pt}/\text{Al}_2\text{O}_3$	315	0.1	-	-	100	[152]
	1 wt% Pt-1 wt% $\text{Pd}/\text{Al}_2\text{O}_3$	320	-	1.33	-	10	[153]
N-ethylcarbazole (NEC)	5 wt% $\text{Pt}/\text{Al}_2\text{O}_3$ (doped with WO_x)	250	-	3.5	-	75.9	[154]
	5 wt% Pd/NGC	180	0.1	10	100	98.72	[149]
	2.5 wt% $\text{Pt}/\text{SiO}_2\text{-TiO(OH)}_2$	180	0.1	7	100	97.9	[149]
	5 wt% $\text{Pd}/\text{Al}_2\text{O}_3$	180	0.1	4	100	100	[149]
	2.5 wt% $\text{Pd}/\text{rGO-EG}$	170	0.1	12	100	84.61	[149]
	5 wt% Pt/TiO_2	180	0.1	6	100	79	[149]
	4 wt% Pd/SiO_2	170	-	1.6	100	100	[149]
	Pd_3 (3.75 wt%)- Ni_1/SiO_2	180	0.1	8	100	91.1	[149]
	Pd_3 (3.75 wt%)- Au_1/SiO_2	180	0.1	8	100	94.9	[149]
	0.58 mol% $\text{Pd}_{1.3}$ -0.42 mol% Ru_1/rGO	180	0.1	4	100	84.11	[149]
	2.5 wt% $\text{Pt}_{0.8}\text{Ir}_{0.2}/\text{Mg-Al-O}$	350	-	1.6	91.1	99.9	[149]
	Pt-Cu/S-1	400	0.1	6	92.26	-	[149]
Monobenzyl toluene (MBT)	Pt-Pd/ Al_2O_3	250	0.1	6	-	99	[155]
Hydrogenation performance							
MBT	0.5 wt% $\text{Ru}/\text{Al}_2\text{O}_3$	175	10	0.104	-	53	[156]
DBT	5 wt% $\text{Ru}/\text{Al}_2\text{O}_3$	170	5	0.772	-	100	[157]
	0.5 wt% Ru/MgO	150	5	3.33	-	100	[158]
	Raney-Ni	170	0.8	18	-	-	[159]
	$\text{Ni}_{70}/\text{AlSiO-1/1}$	150	7	1.5	100	100	[149]
	Mg_2NiH_4	280	6	20	-	-	[160]
NEC	1.3 wt% Ru/YH_3	130	7	2.5	100	100	[149]
	$\text{Ru}/\text{pg-BC}$	130	6	1.17	100	99.41	[149]
	1.5 wt% $\text{Ru-Ni}_1\text{Al}_2\text{-LDO}$	150	8	1	100	100	[149]
	5 wt% Ru/TiO_2	130	7	-	-	95	[149]
	$\text{Ru}/\text{anatase}$	150	7	24	100	95.7	[149]
	$\text{Ru}_{0.7}\text{Ni}_{0.3}/\text{SBA15}$	100	5	1.33	100	99.82	[149]
	Pt (MP)/ $\text{CeO}_2\text{-A-400}$	100	0.5	3	90.8	90.8	[149]
Toluene	Ni nanoflowers	140	5	0.5	100	100	[149]
	6 wt% $\text{Pt}_1\text{Pd}_1/\text{HBEA}$	150	712	2	100	100	[149]
	Pt-Rh/MWNTs	20	1	3	100	100	[149]

LOHCs = liquid organic hydrogen carriers.

mechanisms to ensure uniform mixing of the LOHC liquid, and thermal management systems such as heating jackets to maintain optimal reaction temperatures, particularly crucial for endothermic dehydrogenation processes.

Kiermaier et al. [165] demonstrated successful dehydrogenation using a 100 mL 3-neck flask fitted with an intensive condenser. Their experimental setup incorporated a heating jacket for temperature control and a magnetic stirrer for mixing. The reactor was charged with H12-NEC and catalyst, then purged with argon to establish an inert atmosphere. To obtain accurate kinetic data, they deliberately avoided using solvents or continuous inert gas flows, which could have altered the hydrogen partial pressure within the reactor and compromised the kinetic measurements. Similarly, Huang et al. [166] successfully performed (de)hydrogenation reactions in a 100 mL stainless steel autoclave reactor. The reaction mixture consisted of 10 g of N-ethylcarbazole dissolved in 40 mL of decahydronaphthalene along with a predetermined quantity of catalyst. After sealing, the autoclave underwent 5 consecutive nitrogen purge cycles followed by leak testing. Once verified leak-free, the nitrogen was evacuated, and the reactor was heated to the target temperature with continuous stirring maintained at 600 rpm. Upon reaction completion, the mixture was cooled to room temperature, and the products were immediately separated via filtration.

Whilst batch reactors offer lower initial capital investment and setup complexity, their operational efficiency is limited by significant downtime during startup, shutdown, and product separation cycles. These drawbacks have driven increasing interest in continuous flow reactor systems for LOHC applications.

2.5.2. Continuous catalytic reactors

Fixed-bed reactors have emerged as the dominant technology for LOHC (de)hydrogenation processes, due to their simple design, efficient catalyst utilization, and favorable heat and mass transfer characteristics [30,167–174]. These systems enable continuous operation with simplified product separation and can be configured in both horizontal and vertical orientations, though each configuration requires careful optimization of pressure gradients and thermal management across the catalyst bed.

Heublein et al. [175] conducted a comprehensive comparison of H12-NEC dehydrogenation across 5 various packed-bed reactor configurations displayed in Fig. 8. Their investigation revealed that vertical arrangements provided superior heat transfer efficiency compared to horizontal setups. Whilst both orientations achieved comparable maximum power densities, vertical configurations offered obvious advantages: they enabled counter-current flow between the liquid carrier and evolved hydrogen gas, and demonstrated significantly enhanced radial heat transfer. However, the researchers noted that vertical configurations exhibited decreased catalyst efficiency and operational stability at elevated void fractions. System performance could be optimized through careful pressure control and the implementation of intermediate gas separation stages.

Trickle bed reactor (TBR) has also been considered for continuous operation of LOHC (de)hydrogenation [176–178]. Shen et al. [179] developed a co-current downflow TBR system operating up to 450 °C and 40 bar, consisting of liquid recirculation and continuous gas feed. The setup comprised a 500 mL receiver vessel with a metering pump for liquid circulation, while gas flow was controlled via a mass flow controller and distributed through a spray nozzle system. A pressure control valve manages reactor exit flow and gas separation. The packed bed reactor (25.4 mm internal diameter) contained 5 wt% Pd/Al₂O₃ pellets with 3 mm inert silica support for optimized flow distribution. System initialization involved nitrogen pressure testing followed by catalyst bed wetting using a mixture of 3 g tetrahydrocarbazole in 300 mL mesitylene, circulated at 196 mL/min liquid and 100 mL/min gas flow rates under ambient conditions. Temperature-controlled feed lines and continuous monitoring of process parameters ensured stable operation.

2.5.3. Process intensified reactors

Process intensification has led to innovative reactor designs that address conventional limitations. More recently, Tan et al. [180] introduced an innovative intensified reactor design that combines the TBR with a gas-liquid micro dispersion module, which marks a departure from traditional TBR designs. The design's key characteristics is its specialized gas-liquid contacting system, which employs a stainless-steel microfiltration membrane as the dispersion medium. This novel configuration achieves enhanced hydrogen-liquid mass transfer efficiency while maintaining compact reactor volumes. This intensification strategy addresses one of the fundamental limitations in LOHC (de)hydrogenation processes – the gas-liquid mass transfer resistance – while simultaneously offering the advantages of reduced equipment footprint.

Traditional LOHC setups use 2 separate reactors: one for hydrogenation and the other for dehydrogenation, where each reactor operates at distinct temperature and pressure conditions. However, this conventional 2-reactor setup requires significant energy and time to alternate between the 2 processes. Heating the dehydrogenation reactor from standby to operating temperatures for each hydrogen release step demands a substantial amount of energy. This heat, once generated, is often lost as the reactor cools down when not in use, leading to inefficiencies, especially in systems where hydrogen release timing needs to be controlled. Jorschick et al. [181] developed the one-reactor concept for LOHC systems to address the energy inefficiencies inherent in conventional 2-reactor setups, using a hot pressure swing reactor. In this system, hydrogenation and dehydrogenation reactions are carried out sequentially but within the same heated environment. They further refined the process by adjusting the hydrogenation reaction to occur at a slightly higher temperature than dehydrogenation. This approach allowed the heat generated during the hydrogenation step to be stored within a heat storage system and then reused for the subsequent dehydrogenation step. The transition from hydrogenation to dehydrogenation mode within the single reactor was managed through pressure adjustments at the back pressure regulator. However, this system will require a catalyst that could effectively drive both hydrogenation and dehydrogenation reactions with sufficient activity and selectivity under the same conditions.

Geiling et al. [182] demonstrated a practical implementation of this one-reactor concept for an LOHC-based energy storage system at > 10 kW scale, as shown in Fig. 9. Their system integrated a single bidirectional reactor (oneReactor) with proton exchange membrane fuel cell (PEMFC) and electrolyzer technologies, using DBT as the carrier molecule. The key findings focused on operational insights: they determined that up-flow operation was optimal for dehydrogenation when discharging both liquid and gas products together, and proved the system could handle variable hydrogen input during hydrogenation from the electrolyzer. After extensive testing at 725 h, temperatures above 150 °C, and approximately 7 hydrogenation/dehydrogenation cycles, both the catalyst and LOHC material showed no significant degradation.

Structured reactors, such as microchannel reactors (MCHRs), provide enhanced mass and heat transfer characteristics while maintaining high surface area for catalytic reactions. These intensified designs can significantly improve reaction rates and energy efficiency, though they often come with higher manufacturing costs and complexity. Ali et al. [183] conducted a study using continuous flow MCHRs to examine the efficiency of dehydrogenating H18-DBT, while comparing its performance against a conventional stirred tank reactor. MCHRs are small, tightly packed reactors with channel sizes usually under 1 mm in diameter. Their MCHR achieved relatively higher hydrogen yields (58%) compared to the stirred tank reactor (12%) under identical conditions (290 °C, 20 h), and eventually reached 88% hydrogen release after 40 h. They also characterized the reaction kinetics, finding first-order behavior with an activation energy of 13.79 kJ/mol, while using less catalyst than the stirred tank reactor.

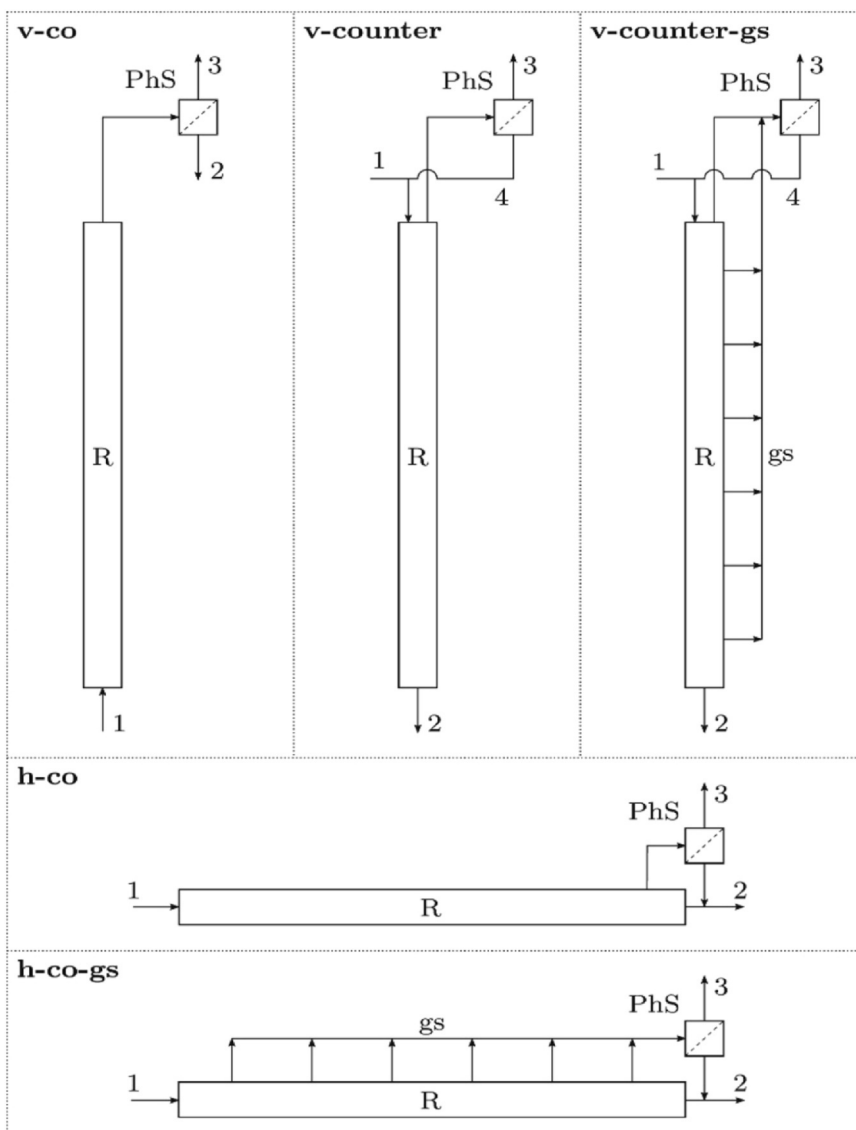


Fig. 8. Different fixed bed arrangement studied by Heublein et al. [175]. v-counter and v-co represents counter-flow and co-flow, respectively, with the liquid feed (1) entering the reactor at the bottom and both the liquid product (2) and the purified hydrogen gas (3) leaving the reactor at the top.

Membrane technology has been integrated with this microreactor concept in a hybrid design. The dehydrogenation of LOHC systems is constrained by both thermodynamic equilibrium and kinetic limitations. When hydrogen accumulates in the reaction mixture, it creates 2 limiting effects. Firstly, according to Le Chatelier's principle, the high hydrogen partial pressure thermodynamically shifts the equilibrium

toward the hydrogenated state, reducing the maximum achievable conversion. Secondly, from a kinetic perspective, the elevated hydrogen concentration can inhibit the catalytic reaction rate by competing for active sites on the catalyst surface and impeding the desorption of newly formed hydrogen. Membrane reactor integration represents an advancement that combines reaction and separation in a single unit,

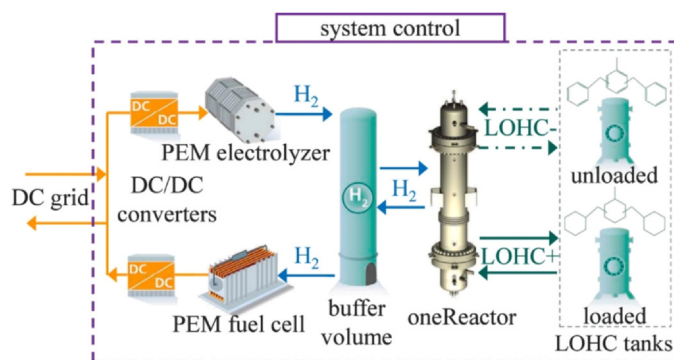


Fig. 9. Process flow diagram of a one-reactor concept for LOHC energy storage system integrated with PEM fuel cell and electrolyzer [182]. LOHC = liquid organic hydrogen carrier; PEM = proton exchange membrane.

and is particularly beneficial for shifting equilibrium-limited dehydrogenation reactions. For effective hydrogen separation, the partial pressure of H_2 on the permeate side must be lower than within the reactor. This can be achieved either by reducing absolute pressure on the permeate side (e.g., near-vacuum conditions) or by using a stripping gas. While stripping gas is commonly employed in lab-scale studies due to its ease of implementation, it is not always a technically reasonable solution for large-scale applications due to process inefficiencies and additional operational complexities. Reactive distillation also provides a similar capability, where it functions by exploiting the difference in volatility between hydrogen and the LOHC compounds. By continuously removing hydrogen as it forms, this technique also drives the reaction beyond its normal equilibrium limitations, while enabling heat integration.

Wunsch et al. [184] developed a micro-structured multistage reactor with integrated membrane separation for H18-DBT dehydrogenation. The reactor consists of several stages, each with its own reaction zone. In each reactor stage, the reactants enter at the center of the chamber and move outward radially, where the products are collected in a ring-shaped outlet. Also, within each stage, the reactor has hexagonally arranged pins spaced 1.2 mm apart that stabilize the catalyst bed and improve heat transfer. Catalyst particles, sized between 200 and 300 μm , are distributed throughout this structure to form a compact bed. The reactor also incorporates a PdAg membrane module with 17 microchannels, designed to allow for intermediate hydrogen removal. They reported an improvement in the degree of dehydrogenation with an increased number of separation stages.

Geißelbrecht et al. [180] employed a novel approach to enhancing hydrogen release from H12-BT using a reactive distillation column, as displayed in Fig. 10. They demonstrated that dehydrogenation of H12-BT can achieve 85% conversion at 210 °C and 200 mbar, a significant improvement compared to ambient pressure conditions. The system's effectiveness stems from 4 synergistic mechanisms. Firstly, boiling conditions reduce hydrogen partial pressure, thermodynamically favoring dehydrogenation. Secondly, the endothermic reaction induces local cooling, causing H12-BT vapor to condense directly in the catalyst bed, efficiently recycling heat where needed. Thirdly, the distillation process concentrates unreacted H12-BT at the catalyst location while removing dehydrogenated product (H0-BT), enhancing reaction rates by over 50% compared to conventional slurry reactions. Finally, continuous removal of H0-BT from the catalyst zone minimizes unwanted side reactions and catalyst coking.

Recently, a new swirling reactor concept for dehydrogenating H18-DBT was reported by Hoecke et al. [185]. The designed reactor is a vertically oriented cylindrical vessel with reaction, expansion, and disengagement zones. The reaction zone is located at the bottom of the reactor, with a diameter of 150 mm and a height of 150 mm, and contains the catalyst bed where dehydrogenation occurs. The expansion zone is located above the reaction zone, with a diameter increased to 300 mm to reduce velocity, allowing solids to fall back into the reaction zone. The disengagement zone is located at the top of the reactor, with a rounded top section, and separates gas from liquid/solid phases. Liquid is fed tangentially into the reaction zone via inlet pipes located around the circumference. This generates a swirling liquid flow pattern within the reactor. The swirling liquid motion fluidizes and keeps the catalyst particles in motion for improved mass/heat transfer and concentrates the gas phase formed during dehydrogenation towards the center of the reactor, separating it from the liquid/solid phases. This reactor was also reported to show a separation effect that improves the removal of H_2 gas from the liquid flow, shifting the reaction equilibrium toward dehydrogenation. To consolidate the comparison of current reactor strategies for LOHC systems, Table 4 summarizes key characteristics of representative reactor types in terms of operational temperature, scalability, thermal performance, hydrogen release behavior, and design challenges.

The selection of an appropriate reactor type is critical to optimizing hydrogen release from LOHC systems, as it directly affects heat transfer, reaction kinetics, and scalability. Table 4 compares key reactor types used in LOHC dehydrogenation across several performance metrics. Fixed-bed reactors are the most commonly adopted design in pilot and demonstration systems due to their simplicity and scalability. However, they often suffer from non-uniform temperature distribution and catalyst degradation at hot spots. Trickle-bed reactors, by enabling better heat dissipation through liquid–solid–gas interactions, offer improved thermal control and moderate H_2 release rates, though their performance may be limited by internal mass transfer resistance. Batch reactors are generally used for laboratory-scale studies because of their ease of operation, but are unsuitable for continuous processing. In contrast, MCHRs have emerged as promising alternatives due to their high surface-to-volume ratio and excellent heat management, supporting faster reaction rates. Nevertheless, challenges related to fabrication complexity and clogging persist. Membrane reactors, by combining dehydrogenation and in-situ hydrogen separation, offer a compact and efficient route for LOHC processing. Despite their promise,

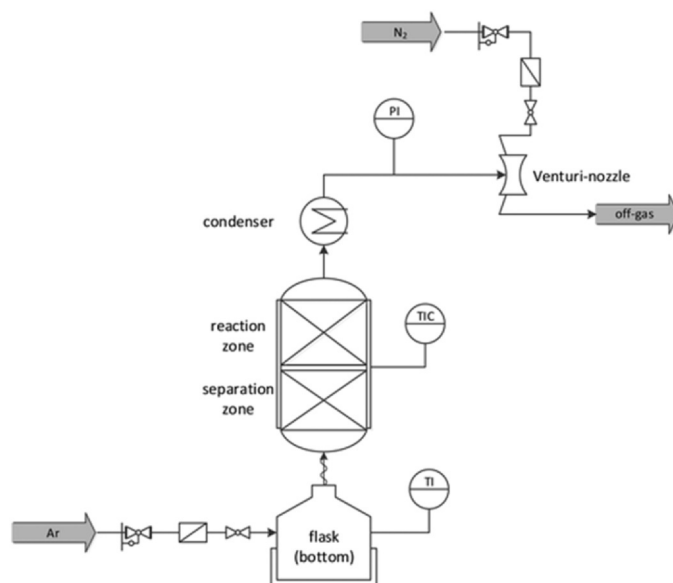


Fig. 10. Reactive distillation reactor set-up used for the dehydrogenation of H12-BT. [180].

Table 4
Comparison of reactor types for LOHC dehydrogenation

Reactor type	Temp. range (°C)	Scalability	Heat management	H ₂ release rate	Advantages	Limitations	Refs.
Batch reactor	150–300	Low	Poor	Low	Simple setup, good for lab-scale tests	Poor control, not suitable for continuous flow	[166]
Fixed-bed reactor	200–320	High	Moderate	Moderate	Scalable, widely used in demo systems	Thermal gradients, catalyst hotspots	[81,166]
Trickle-bed reactor	180–300	Medium-High	Moderate-Good	Moderate-High	Liquid–solid–gas contact, better heat control	Mass transfer limitations	[81,166]
Microchannel reactor	150–250	Low-Medium	Excellent	High	High surface-to-volume ratio, fast kinetics	Complex fabrication, clogging risk	[186,187]
Membrane reactor	200–280	Medium	Moderate	Moderate	H ₂ separation and reaction integration	Membrane durability, cost	[188,189]

LOHC = liquid organic hydrogen carrier.

membrane cost, selectivity, and long-term durability remain obstacles to industrial application [188,189]. The trade-offs among reactor types underscore the importance of tailoring reactor configurations to specific LOHC scenarios, depending on application scale, energy constraints, and system integration needs. A clear understanding of these parameters is essential for advancing LOHC-based hydrogen storage toward commercial viability.

2.6. Role of LOHCs in carbon-neutral hydrogen supply chains

LOHCs have emerged as a transformative technology for hydrogen storage and transport, offering solutions to critical challenges in hydrogen supply chains. They provide a flexible platform compatible with both green and blue hydrogen production, fostering the scalable adoption of carbon-neutral hydrogen. This technology is particularly advantageous for long-term storage and long-distance transport, demonstrating significant economic benefits over compressed hydrogen (CH₂) for storage periods exceeding 60 days, making it ideal for large-scale, extended storage requirements [36]. This makes LOHCs especially valuable for transporting hydrogen produced in renewable-rich, low-demand areas to high-demand regions via existing infrastructure [22].

LOHCs also offer several critical advantages for hydrogen storage and transport, making them a highly effective solution for large-scale applications. Designed with a high HSC, LOHCs facilitate efficient storage and transportation, enabling the handling of hydrogen on a broad scale. They also provide enhanced safety and environmental benefits, as they are nonvolatile and nontoxic, positioning them as a safer and more eco-friendly alternative to gaseous hydrogen. Unlike hydrogen gas, which requires high-pressure storage, LOHCs remain in a stable liquid state across a wide temperature range, simplifying storage, handling, and transport processes. In addition, LOHCs can leverage existing transport and storage infrastructure but still require dedicated dehydrogenation facilities at the point of hydrogen use [36]. This versatility makes LOHCs not only efficient for long-distance and long-term applications but also practical and cost-effective, particularly in inter-continental hydrogen transport scenarios. By leveraging conventional gasoline and diesel delivery and storage systems, LOHC technology ensures hydrogen can be transported over long distances, stored for extended periods, and readily released as needed [36].

LOHC technology can play a significant role in advancing a carbon-neutral hydrogen supply chain by addressing several key challenges associated with renewable energy. For hydrogen produced from intermittent renewable sources like wind and solar, which require large-scale, long-term storage to match variable supply with demand, LOHCs can offer a stable medium to store surplus renewable energy as hydrogen. This reduces reliance on high-voltage transmission across regions, allowing energy to be stored locally and used when needed. LOHCs can also facilitate efficient energy transport by enabling hydrogen generated in renewable-rich areas to be easily moved to regions with high demand, supporting a balanced and resilient energy distribution system. LOHCs can also offer a flexible and compatible solution for integrating both green and blue hydrogen into existing supply chains. Their liquid properties closely resemble those of fossil fuels, allowing them to be transported using existing petroleum infrastructure with minimal modifications. LOHCs can undergo hydrogenation to be transported safely in trucks, ships, and pipelines, where the hydrogen can then be dehydrogenated on-site for use in applications like fuel cells, industrial processes, or chemical synthesis [39]. To illustrate the trade-offs and system-level implications of different LOHC candidates, a comparison of major systems is presented in Table 5.

Table 5 summarizes the key performance parameters of representative LOHC candidates in terms of hydrogen storage capacity, dehydrogenation temperature, safety profile, economic feasibility, and commercial maturity. Among the widely studied systems, methylcyclohexane (MCH) offers relatively mild dehydrogenation conditions and

Table 5
Comparison of key performance metrics of representative LOHC systems

LOHC system	Hydrogen capacity (wt%)	Dehydrogenation temp. (°C)	Safety and toxicity	Raw material cost	Commercial status	Refs.
Perhydro-N-ethylcarbazole (H12-NEC)	5.8	~180	Medium; moderate toxicity; sealed	High	Pilot: Hydrogenious (Germany)	[81,190]
Methylcyclohexane (MCH)	6.2	~150–200	Moderate volatility; flammable	Medium	Demo: Chiyoda AHEAD (Japan)	[81,191]
Dibenzyltoluene (DBT)	6.3	~290–320	High stability; low vapor pressure	Medium-low	Multiple pilots in EU/China	[81,192]
Biphenyl-based LOHCs	> 7.0	~250–300	Chemically stable; dense	Medium	Lab-scale exploration	[82]
Ethanol/formic acid (bio-derived)	< 6.0	< 150	Nontoxic; abundant; biomass-based	Low	Emerging research	[193–195]

EU = European Union; LOHC = liquid organic hydrogen carrier.

moderate hydrogen content, making it suitable for applications prioritizing rapid release and integration with existing infrastructure. Perhydro-N-ethylcarbazole (H12-NEC), though possessing slightly lower hydrogen content, is often preferred for its compatibility with noble metal catalysts and manageable toxicity. DBT stands out for its exceptional thermal and chemical stability, albeit requiring significantly higher dehydrogenation temperatures, which can impact system energy efficiency. Notably, biphenyl-based compounds exhibit the highest theoretical hydrogen capacity (> 7 wt%) but face challenges related to handling and regeneration. On the other hand, bio-derived LOHCs, such as ethanol and formic acid, are gaining attention for their environmental compatibility and low cost, although their hydrogen density is currently inferior. These emerging candidates highlight a promising shift toward more sustainable LOHC systems and merit further investigation. The comparative analysis underscores that no single LOHC satisfies all performance criteria; rather, trade-offs exist between storage density, energy input, cost, and operational safety. Selection should therefore be scenario-dependent, considering specific technical and logistical constraints.

3. Applications of LOHCs in the hydrogen economy

The term "H₂ economy" refers to an economic system where the primary energy carrier is large-scale H₂ manufacturing using renewable power. Decarbonizing the energy industry and reducing reliance on fossil fuels are the goals of this system [196]. The key elements of the shift to this low-carbon economy are H₂ production, transportation, storage, and usage (HPTSU) as seen in Fig. 11 [197]. Hydrogen storage and transportation will be critical components in the whole deployment of the H₂ economy [12]. As a result, a significant amount of published research on LOHCs has concentrated on their capacity to securely and effectively manage H₂ in its chemical state for storage and transportation. However, scientists have lately begun to investigate the use of LOHCs in H₂ refining/purification as a phase in the H₂ generation process. This section will describe recent research advancements in broadening the uses of LOHCs in the H₂-based economy. Furthermore, instances of industrial uses and innovative demonstration initiatives now underway in various countries around the world are emphasized to show the LOHC technological advances' applicability and effect on the emerging H₂-based economy.

3.1. Large-scale, long-term hydrogen storage and transportation

It has been shown that long-term, large-scale H₂ storage and transportation are crucial for guaranteeing a consistent and dependable energy supply in the specifics of the future H₂-based economy [199]. The primary purposes of stationary H₂ storage facilities are stationary energy supply and on-site storage at the manufacturing or final consumer facility. Due to its commercial accessibility and maturation, compressed H₂ is currently the most prevalent use for these technological advances; however, large-scale stationary hydrogen storage employing LOHCs is emerging as an attractive option for security concerns and the benefit that LOHC technological advances can use existing liquid fossil fuel facilities [2]. Hydrogen must be transferred to the location of usage after it has been stored. Compared to alternative H₂ transportation techniques like compressed or liquefied H₂, long-range H₂ carriers (LOHCs) have shown to offer several advantages, including secure and efficient long-distance delivery. In addition, LOHCs can leverage the existing fossil fuel facilities such as trucks, trains, and tanker ships, which can lower transportation costs and streamline cross-regional and international H₂ logistics. Notably, certain LOHC structures have been identified as promising options for both stationary H₂ storage and transportation, such as dodecahydro-N-ethylcarbazole/N-ethylcarbazole, MCH/toluene, and perhydro-dibenzyltoluenes (H18-DBT)/dibenzyltoluenes (H0-DBT) [28]. For example, toluene and DBT are now employed for large-scale H₂ storage and transportation in

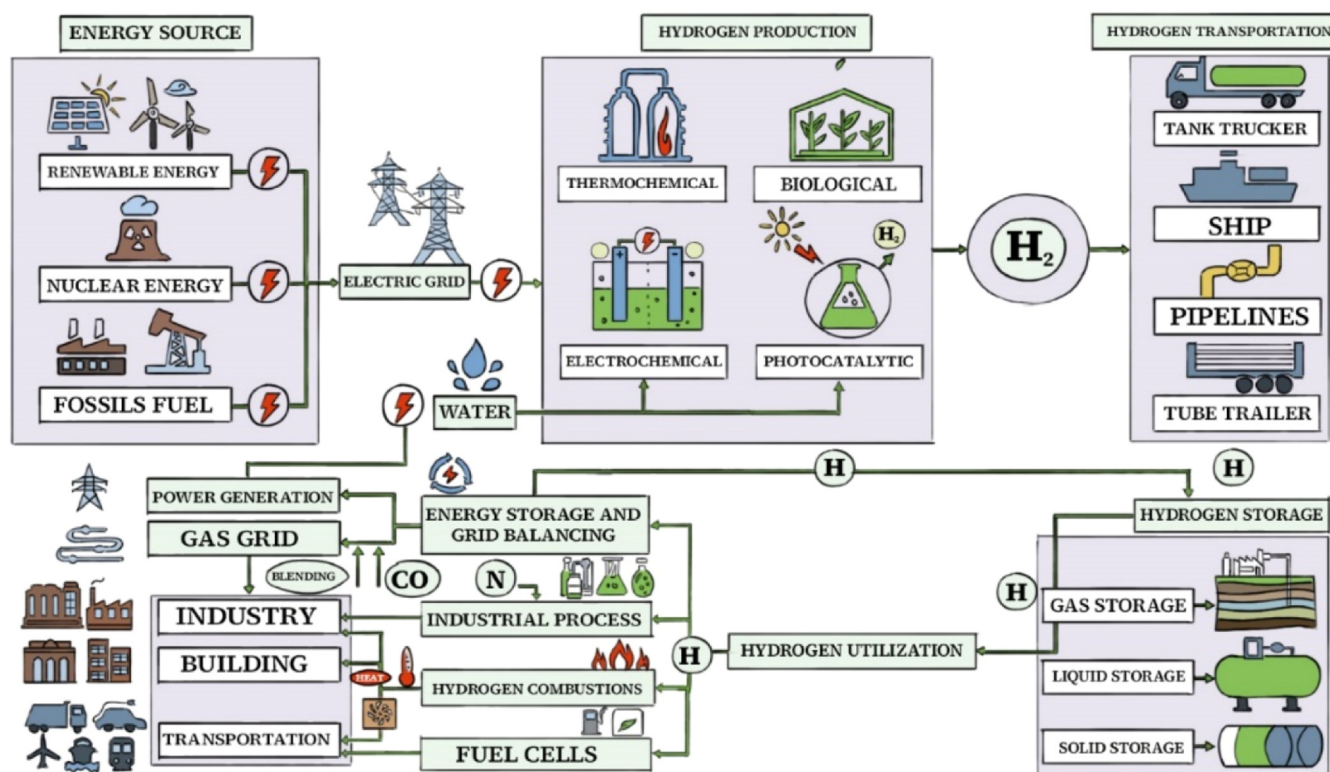


Fig. 11. A synopsis of technologies for HPTSU [198]. HPTSU = hydrogen production, transportation, storage, and usage.

Germany and Japan. Additionally, Hynertech Co. Ltd. in China has created an LOHC system based on carbazoles that is already in use for the commercial transportation of H_2 . Table 6 provides additional information about these commercial uses as well as related projects in different nations.

The AquaVentus initiative harnesses heat to support Helgolands heating requirements, boosting energy efficiency and aiming for H_2 power generation by 2035. Meanwhile, the SmartQuart project taps into heat from electrolysis for nearby industries and repurposes fossil fuel infrastructure for LOHC transport [36]. Kopernikus Power to X investigates methods for dehydrogenation heat, advancing LOHC technology. Similarly, GET H_2 , and TransHyDE/ H_2 Mare projects optimize existing systems to cut costs and enhance sustainability. Project HECTOR stands out by focusing on using surplus hydrogenation heat at a level unlike many other initiatives that only address aspects of LOHC technology without considering the complete supply chain [28].

The LOHC transport chain is classified into 2 types: uninterrupted (single link) and interrupted (multilink). Interrupted transportation consists of pre-carriage, main-carriage, and on-carriage phases. In the "Green H_2 @Blue Danube" initiative, LOHCs are mainly conveyed by ship along the Danube, with trucks covering lesser distances. This collaboration, which includes Hydrogenious LOHC Systems and VERBUND, brings together the whole LOHC supply chain. HySTOC in Finland supports H_2 refueling stations, whereas Green Crane in Spain concentrates on supplying LOHC to Germany [205]. The Orion Project, which runs until 2050, seeks to produce 32 TWh of H_2 , influencing future LOHC supply (see Table 7).

The Dii Desert Energy and Haru Oni projects, situated outside of Europe, are dedicated to H_2 research. Dii Desert Energy involves initiatives to improve energy supply stability through LOHC technology, but neither project considers the utilization of excess hydrogenation heat for energy efficiency. Haru Oni, located in Chile, plans to create e-fuels such as e-methanol, mostly for racing (see Table 8). However, these e-fuels are not categorized as LOHCs, which limits the system's sustainability advantages.

In Japan, the AHEAD (Advanced Hydrogen Energy Chain Association for Technology Development) project, directed by Chiyoda Corporation, uses a toluene/methylcyclohexane (TOL/MCH) system for H_2 storage, providing an alternative to the DBT [212]. The TOL/MCH system holds 6.2% H_2 by weight but has a lower volumetric storage density than DBT. Platinum-based catalysts, which are frequently coupled with metals such as tin, molybdenum, or copper, have been proven in studies to be quite effective in the hydrogenation process. Advances are also being made in catalyst support, with materials such as Al_2O_3 , USY zeolite, and TiO_2 demonstrating promise [213]. Chiyoda's AHEAD uses its LOHC system, SPERA, to transport H_2 from overseas to Japanese power plants, where it powers gas turbines. Because of the low boiling temperatures of toluene and MCH, the system needs good sealing and pressure management to reduce evaporation and environmental loss [214].

3.2. Mobility technologies

The Intergovernmental Panel on Climate Change reported that in 2019, the transportation sector was responsible for 23% of global CO_2 emissions from energy sources [216], with projections suggesting that this share may rise over time. However, although automakers globally are advancing the development of electric vehicles (EVs), these vehicles still face limitations, including restricted driving range and lengthy charging periods. From the perspective of low-emission energy systems, hydrogen is gaining attention as a promising fuel for mobile applications [217] because of its ability to produce clean energy, supporting the transport sector's decarbonization. Fuel-cell technology, which produces energy through a reaction between O_2 and H_2 , with just water vapor as an emission, is currently the most efficient method of using H_2 in mobile situations. Although fuel cell vehicles have advantages over battery-EVs, like longer driving range and quicker refueling periods, they also have infrastructure development issues.

Nonetheless, obstacles (such as the challenges) of storing sufficient hydrogen space and weight constraints, along with stringent safety

Table 6
Selected few identified LOHC projects in Germany

Projects	GET H ₂	AquaVentus	TransHyDE/H ₂ Mare	LOHC train
Description	Planning the implementation of facilities for the production, acquisition, transportation, and storage of green H ₂ in many projects.	Generate green H ₂ using electricity generated by offshore wind farms.	The H ₂ Mare-led initiative researches techniques to manufacture H ₂ and downstream products utilizing off-grid wind turbines directly on the sea.	Install LOHC systems in trains.
Year	2024–2030	2023–2035	2021–2025	2019–2023
Target	Develop a nationwide H ₂ network.	10 GW of green H ₂ (from offshore wind) will be transported by pipeline and stored as long-term organic H ₂ chains (LOHCs) by 2035.	Establish a LOHC supply chain using offshore wind energy and dehydrogenation at Hamburg's port.	Creation of a commuter train propelled by LOHC
Consortium	OGE, BP, Thyssengas, RWE, Evonik, Nowega.	Aktiengesellschaft EMS, Buckstay Group GmbH, Cuxport GmbH, DNV, E. ON SE, Hydrogenious LOHC Tech. GmbH.	Over 200 partners; additional information not provided	Research Center Jülich, FAU Erlangen/Nuremberg, Helmholtz Institute Erlangen/Nuremberg, Fraunhofer HHI, Hydrogenious LOHC Technologies
LOHC compound	Dibenzyltoluene	Benzyltoluene	Benzyltoluene	Benzyltoluene
Electrolysis	Utilizing surplus electrolyte heat, wind, and solar energy, and power-to-gas facilities	On-site electrolysis and offshore wind power.	-	-
Usage	Particular attention to large-scale industrial users and mobility.	Green H ₂ will be supplied to Helgoland by a 1 × 15 MW plant, which will reduce CO ₂ emissions and replace fossil fuels.	Integrating electrolysis offshore to lower transmission costs and help the industry become carbon neutral.	Transportation (commuter train)
Transport	Tank trucks, road	Tank ship, water	Water, road, and use of the current fossil fuel tanker fleet facilities	Rail
Supply chain	Hydrogenisation, electrolysis, transport	Hydrogenisation, electrolysis, transport	Hydrogenisation, electrolysis, dehydrogenization, transport	Dehydrogenization
Location	Lingen	Helgoland	Hamburg/Helgoland	Jülich
Financing	Applied for Important Projects of Common European Interest (IPCEI) funding	No information	Funded by BMBF (sum of 700,000,000 EUR; TransHyDE/H ₂ Mare component not disclosed)	The Bavarian State Ministry of Economic Affairs, Regional Development, and Energy allocated 28 million EUR.
Refs.	[49,200]	[201,202]	[203]	[204]

LOHC = liquid organic hydrogen carrier.

Table 7
Overview of key LOHC projects in Europe

Projects	Orion project	IPCEI "Green Hydrogen@Blue Danube"	Green Crane	HySTOC
Description	Transportation and H ₂ production.	To establish a sustainable green H ₂ supply chain throughout Europe, leveraging existing facilities for efficient LOHC transport and improving regional energy integration.	The Green Crane program intends to build green H ₂ flows from South to North across Europe while also increasing local demand in Spain and Italy. It also intends to establish cross-border connections with the Netherlands and France.	Research was conducted to build comprehensive H ₂ logistics for usage at refueling stations
Year Target	2026–2050 32 TWh of green/blue H ₂ produced year, with some exported to the UK and other nations	2022–2028 To establish a European supply chain for green H ₂ , with a specific focus on providing 1800 tons of green H ₂ via LOHCs in Bavaria.	2014–2035 Establishing a hydrogen transport network from Spain to Central Europe.	2018–2022 To develop advanced distribution and storage systems for H ₂ refueling stations, using LOHCs to improve supply and transportation efficiency.
Consortium	Highlands and Islands Enterprise, OGTC, and Shetland Islands Council.	VERBUND, Hydrogenious LOHC Technologies	Vopak, Enagás Renewable, Hydrogenious LOHC Technologies, Bosch, Terega, McPhy, Petronor, Arcelor-Mittal I + D, Gransolar, Vestas, Falck Renewables, IGNIS Energia	VTT Technical Research Center of Finland, Gasum, Neste, and Helsinki University of Technology
LOHC compound	Dibenzyltoluene	Dibenzyltoluene	Methylcyclohexane	Dibenzyltoluene
Electrolysis	offshore wind-powered electrolysis.	alkaline electrolysis, solar, and wind power.	Water electrolysis using both PEM and ALK electrolyzers	PEM and ALK electrolyzers
Usage	Green H ₂ will be exported to mainland Europe for energy needs	Transports LOHCs by ship along the Danube, integrating the full LOHC supply chain.	Enhancing mobility, energy, and industrial applications through its LOHC supply chain	Refueling station (mobility).
Transport	Pipelines and ships	Ship and truck	Pipeline - Compressed H ₂ , Ship - LOHC	Truck, road
Supply chain	Hydrogenisation, electrolysis, transport	Electrolysis, hydrogenisation, transport, dehydrogenization, usage	Electrolysis, hydrogenisation, transport, dehydrogenization, usage	Hydrogenisation, transport, usage
Location	Shetland	Austria and Germany to Romania and Bulgaria	Spain (La Robla)	Finland (Voikoski)
Financing	£5 million from the Shetland Islands Council	The 72.5 million Euro grant notification was personally delivered by the Regional Development and Energy department.	Partly EU-funded	€2499,921,25 (EU funded)
Refs.	[206]	[207,208]	[209]	[210,211]

ALK = Alkaline; EU = European Union; LOHC = liquid organic hydrogen carrier; PEM = proton exchange membrane.

Table 8
International LOHC projects: a selected few

Projects	AHEAD	Desertec Industrial Initiative (Dii) Desert Energy
Description	Focused on utilizing LOHC technology to transfer H ₂ from other countries to Japan, where the released gas fuels gas turbines in power facilities to provide a steady source of electricity.	Production of Green H ₂ and exportation.
Year	2017–2020	2009
Target	Demonstrating the potential for using LOHCs to create an H ₂ transport chain	Develop robust, emission-free energy distribution networks with the aid of LOHC technologies.
Consortium	Chiyoda Corporation, Mitsui & Co., Ltd., Nippon Yusen Kabushiki Kaisha (NYK Line), JERA Co., Inc.	ACWA Power, Masdar, TAQA, International Renewable Energy Agency (IRENA), Siemens, and various governmental and research organizations.
LOHC compound	Toluene and methylcyclohexane (MCH)	Dibenzyltoluene
Electrolysis	PEM and ALK	alkaline electrolysis, solar, and wind power.
Usage	Energy	Producing and exporting renewable energy, enhancing energy security, fostering economic development in desert regions, Ship
Transport	Ships, road	Electrolysis, hydrogenisation, transport
Supply chain	Hydrogenisation, transport, dehydrogenation, usage	North Africa and the Middle East
Location	4–6–2 Minatomirai, Nishi-ku, Yokohama	Funded by a consortium of European industrial companies and financial institutions
Financing	Funded by the New Energy and Industrial Technology Development Organization (NEDO)	
Refs.	[215]	[191]

ALK = Alkaline; LOHC = liquid organic hydrogen carrier; PEM = proton exchange membrane.

standards, continue to restrict the broader adoption of hydrogen fuel cell technology in transportation. However, these limitations are not insurmountable; this is particularly true because ongoing research may eventually address many of these issues. Although the potential benefits are substantial, the path forward remains fraught with difficulties [218].

The implementation of heat management presents increased challenges in mobility applications, primarily due to the necessity for lightweight and compact equipment, alongside more stringent safety regulations. As illustrated in Fig. 12, a typical on-board vehicle technology system would encompass a fuel cell, a dehydrogenation reactor, and a storage tank for LOHCs. To realize an autonomy of 500 km, one would need approximately 5 kg of H₂. This requirement translates to needing between 80 and 120 L of LOHC, which varies depending on the hydrogen content. Storing LOHC on board is facilitated by a dual-tank design, which accommodates both hydrogenated LOHC (Hx-LOHC) and dehydrogenated LOHC (H0-LOHC) [48]. During the refueling process, Hx-LOHC is pumped into one tank, while H0-LOHC is simultaneously extracted from the other. However, the complexity of managing these systems can be significant because of the interdependencies involved.

Although there are advantages to this approach, careful consideration must be given to safety and efficiency.

To use waste heat from the fuel cell to drive the dehydrogenation reaction, there must be an excess of thermal energy generated by the fuel cell; otherwise, additional heat from the battery or H₂ combustion will be required, reducing the fuel cell's energy self-sufficiency [36]. In general, several factors, including fuel cell performance and temperature management, determine the actual amount of H₂ consumed to generate heat. If all the required heat for dehydrogenation were sourced from H₂, about 20% of the stored H₂ could be expended. Given a fuel cell efficiency of 55%, this design lowers the dehydrogenation-fuel cell system's net efficiency to approximately 44%, which remains high compared to a gasoline combustion engine, typically around 20–40% [219]. Furthermore, with a dynamic discharge of H₂ from the LOHC, the storage facility must be prepared to respond quickly enough to fluctuations in power generation and demand from the fuel cell [220]. It is therefore difficult to provide the dehydrogenation reactor with heat quickly enough to produce H₂. Overall, the reaction temperature rises gradually over a brief period in response to variations in the H₂ demand. It has been conceptualized to operate a direct LOHC fuel cell on

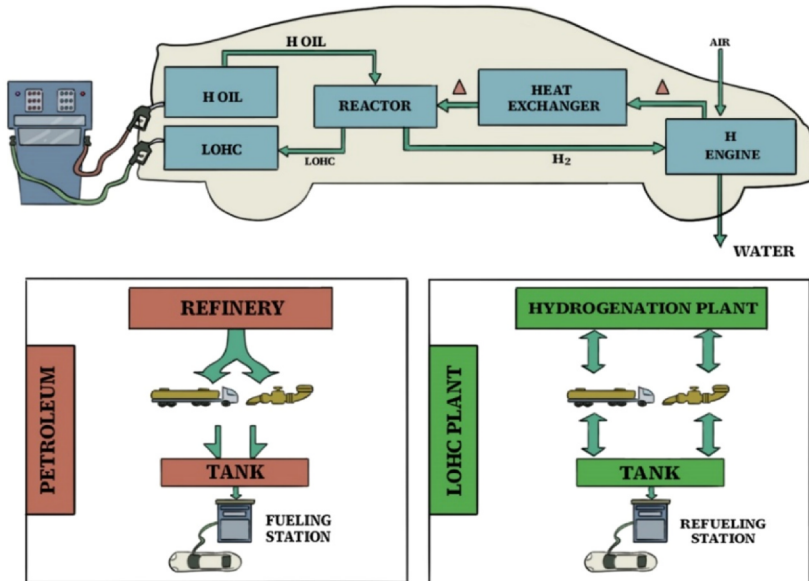


Fig. 12. Prototype of H₂ automobile using LOHC for H₂ supply. LOHC = liquid organic hydrogen carrier.

board without releasing basic H_2 during LOHC dehydrogenation or fuel cell operations [36].

One of the big challenges with the use of waste heat from fuel cells for LOHC dehydrogenation is the big temperature mismatch. Most types of fuel cells used for vehicle applications, like PEMFCs, operate below 100°C , whereas LOHC dehydrogenation typically requires temperatures higher than 200°C . This discrepancy thus calls for supplementary heat sources such as additional hydrogen combustion, high-efficiency heat exchangers, or battery-assisted heating to maintain dehydrogenation. Recent progress in catalyst development and reactor design, including MCHRs and autothermal dehydrogenation, will also have the effect of lowering the LOHC dehydrogenation temperature required, enhancing feasibility again in mobile applications. Optimization of these systems for onboard hydrogen release efficiently while retaining the whole-system efficiency at its optimum clearly requires further work.

On-board hydrogen generation using LOHCs is emerging as a promising option for large-scale mobile applications, such as ships and trains, where space constraints are less restrictive compared to smaller vehicles. In Germany, a significant development project is underway to evaluate the practicality and efficiency of this technology. The project's goal is to establish a clean and safe public railway system powered by hydrogen fuel and direct LOHC fuel cell technology [221]. To the best of our understanding, mobility implementations are still a long way from being technically deployed because of challenges related to on-board dehydrogenation and carrier safety considerations.

Other than the heating issues, a few key obstacles impede LOHC technology for practical implementation in mobile applications, including catalyst durability, energy demand, and system compactness. Long-term stability remains one of the most important challenges for dehydrogenation catalysts. Presently, most LOHC dehydrogenation catalysts lose their activity over time and are not able to provide high performance beyond 10,000 hours of operation. The only technology that has been commercially demonstrated so far is the MCH system by Chiyoda Corporation. Highly durable catalysts with very low deactivation are necessary to enable LOHC-based mobility solutions [144].

Conversely, dehydrogenation of LOHC is an endothermic reaction, thereby requiring high heat and enthalpy input. The reaction kinetics of hydrogen release are considerably slow, hence the system can't meet the quick startup requirements of automotive applications [222]. This operates at variance with the conventional compressed hydrogen storage, which guarantees nearly instant fuel availability; the LOHC systems need time to heat up and release hydrogen, a factor standing in the way of performance and practicality for every vehicle.

LOHC systems currently lack the compactness to be integrated on board passenger vehicles. The dual-tank requirement for storing hydrogenated and dehydrogenated LOHC, accompanied by heat exchangers and catalytic reactors, raises system weight and complexity [182]. In contrast to compressed or liquid hydrogen tanks, which have been designed for space-efficient integration in automotive applications, LOHC storage and processing equipment have yet to be miniaturized for feasibility. Meeting these challenges will require catalyst longevity, process intensification, and system miniaturization. While LOHC technology does offer enormous advantages concerning safety and storage aspects, these limitations need to be overcome before its widespread application in the field of mobility [223].

3.2.1. Integration of fuel cell waste heat in LOHC-based mobility systems

The challenge with LOHC in onboard hydrogen generation lies in its thermal requirements for dehydrogenation. Whilst PEMFCs, the commonly used fuel cells in mobility applications due to fast response and high efficiency, produce waste heat at around 80°C , LOHC dehydrogenation normally requires a temperature range of $200\text{--}300^\circ\text{C}$. This temperature disparity limits the direct utilization of PEMFC waste heat for the process.

Recent research in Germany is investigating the integration of high-temperature SOFCs with LOHC systems. Operating at $600\text{--}1000^\circ\text{C}$, the

SOFC produces a high amount of high-grade waste heat that can be used more effectively for LOHC dehydrogenation. In such a case, thermal coupling of SOFCs with LOHC dehydrogenation reactors could significantly raise the overall system efficiency and decrease the need for external heating. Furthermore, new catalytic materials and reactor designs like microchannel and autothermal dehydrogenation reactors are under investigation with respect to reducing the temperature required for hydrogen release even further and to further optimize the feasibility of LOHC-based solutions for mobility. Although PEMFCs remain the preferred technology for automotive applications, ongoing advancements in SOFC-LOHC integration could position it as a viable alternative for heavy-duty transport, maritime shipping, and stationary power generation. Nevertheless, further research and pilot projects are still necessary to find out how far this concept would be practical and scalable.

3.2.2. Critical evaluation of onboard LOHC dehydrogenation within the application to passenger vehicles

Innovative onboard dehydrogenation of LOHC is a very appealing approach for onboard hydrogen storage in mobility applications; up to now, great technical and financial challenges impede its use in road transport applications within passenger vehicles. Among the various challenges, temperature control remains important: dehydrogenation proceeds at elevated temperatures, usually from 200°C to 300°C —therefore, sufficient heat integration should be foreseen with the vehicle fuel cell. Unlike stationary applications, where external heat sources can be optimized, automotive systems face constraints in generating and managing the required thermal energy efficiently without compromising vehicle performance.

The second limitation involves an energy efficiency trade-off: The endothermic dehydrogenation process consumes part of the energy content of the stored hydrogen, reducing overall efficiency in the fuel cell-electric powertrain [224]. Compared with CH_2 or LH_2 storage, where hydrogen is already in a state that is immediately useful to the fuel cell, LOHC-based storage introduces additional conversion losses that could limit its competitiveness in lightweight, high-efficiency transportation applications.

Besides that, there is system complexity and infrastructural challenges: sophisticated catalyst systems, efficient reactor design, and durable heat exchangers increase weight and manufacturing cost for the vehicle onboard dehydrogenation. Besides, the introduction of a dual-tank system—one for hydrogenated LOHC and another for dehydrogenated LOHC—introduces further design constraints and logistical challenges at the refueling infrastructure. While gaseous hydrogen refueling stations are already being deployed in several regions, widespread LOHC infrastructure for automotive applications is still in its infancy.

Taking all these considerations into account, on-board LOHC dehydrogenation for now seems better applicable to heavy-duty transport missions, such as trains, ships, and long-haul trucking, since for these no severe weight limitation is at stake and surplus heat may be dealt with more conveniently [225]. Whereas several ways of improving the catalyst as well as process intensification methods are still objects of research to pave the way further for on-board dehydrogenation concerning passenger cars, significant system efficiencies, cost-cutting measures, and infrastructure installation would be necessary prior to industrial applications.

While the hydrogen supply to a fuel cell necessarily has to follow a dynamic operation, it by no means constrains the dynamics of hydrogen release from the LOHC system in any way. Intermediated storage of the released hydrogen is possible either via small buffer tanks or by exploiting the free volume of the release and purification unit to decouple immediate fuel cell demand from LOHC release dynamics [225]. This buffering capability allows for more stable and efficient fuel cell operation, especially under fluctuating power loads. This form of storage can make up for transient mismatches in

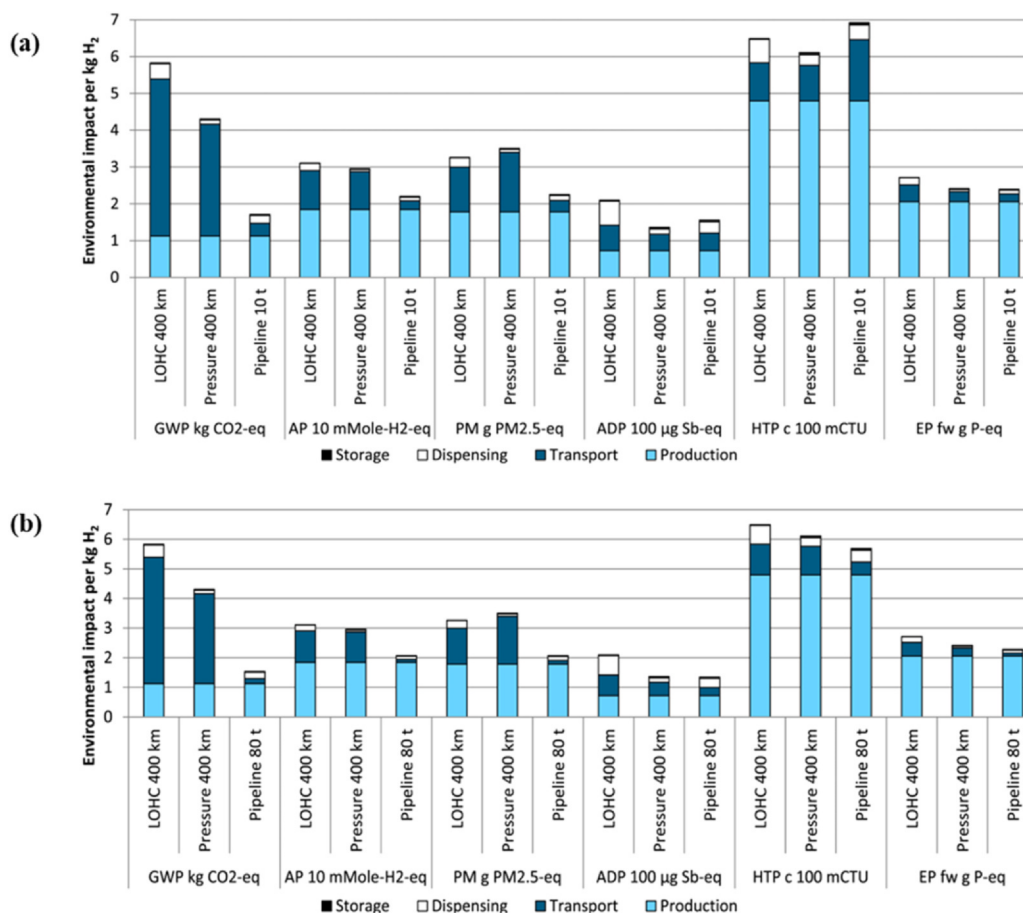


Fig. 13. Environmental impacts of different cases considered (a) Hydrogen demand at 10 t/d and transportation distance of 400 km (b) Hydrogen demand at 80 t/d and transportation distance of 400 km. [236].

hydrogen demand and release kinetics, hence improving overall performance and operational flexibility.

4. Environmental and economic considerations

While LOHCs offer promising solutions for hydrogen storage and transportation, a holistic evaluation of their environmental and economic implications is crucial. This section presents an extensive overview of the life cycle assessment (LCA) of LOHC systems, scrutinizing their carbon footprint and overall environmental impact. It also explores the economic feasibility and market potential of LOHCs by conducting an in-depth analysis of their current cost, emerging trends supporting their adoption, and future projections.

4.1. Life cycle assessment (LCA) of LOHC systems

LCA is an essential instrument for assessing the environmental impact and sustainability of LOHC systems. LCA studies can offer insights into the carbon footprint and overall environmental performance of LOHCs by examining their full life cycle, from manufacture to consumption and end-of-life. The systematic implementation of LCA enables comprehensive evaluation of environmental, social, and economic parameters, supporting evidence-based sustainable decision-making processes [226]. This standardized methodology, aligned with ISO 14040 and 14044 frameworks, progresses through 4 distinct stages: goal and scope definition, life cycle inventory analysis, impact assessment, and results interpretation [227,228]. The assessment encompasses the 16 life cycle impact categories recommended by the Environmental Footprint (EF) impact assessment method [229,230]. The EF is an LCA-based methodology developed to quantify the

environmental impacts associated with products (goods or services) and organizations, providing a comprehensive framework for evaluating their environmental footprint holistically [231,232]. The impact categories are normalized and weighted based on their relative importance.

The environmental impact of LOHC systems is influenced by several elements, including the LOHC synthesis process, hydrogen production techniques, transportation logistics, and the subsequent dehydrogenation and regeneration of the spent LOHC. LOHC storage cycle typically involves the catalytic hydrogenation of unsaturated organic molecules, such as naphthalene or DBT, utilizing hydrogen gas. The energy and material inputs necessary for this process, along with any related emissions or waste streams, contribute to the total environmental effect. The origin of hydrogen utilized in the hydrogenation process is pivotal in assessing the carbon footprint, as hydrogen generated from renewable sources (e.g., electrolysis powered by renewable electricity) exhibits a markedly lower impact than hydrogen obtained from fossil fuels [233,234]. In the use phase, the dehydrogenation of the charged LOHC liberates the stored hydrogen for many applications, including fuel cell cars and industrial processes. This phase entails energy inputs and possible emissions, contingent upon the dehydrogenation method utilized. Furthermore, the regeneration of the depleted LOHC for re-utilization necessitates energy-intensive procedures, which must be considered in the LCA. Extensive LCA studies have been performed to assess the environmental implications of LOHC systems across multiple impact categories and compare them with alternative hydrogen storage technologies. These investigations often encompass the cradle-to-gate approach, including LOHC synthesis, hydrogen production, transportation, dehydrogenation, and regeneration.

Wulf and Zapp [235] performed an LCA of a toluene-based LOHC system, integrated with hydrogen generation methods, such as SMR and

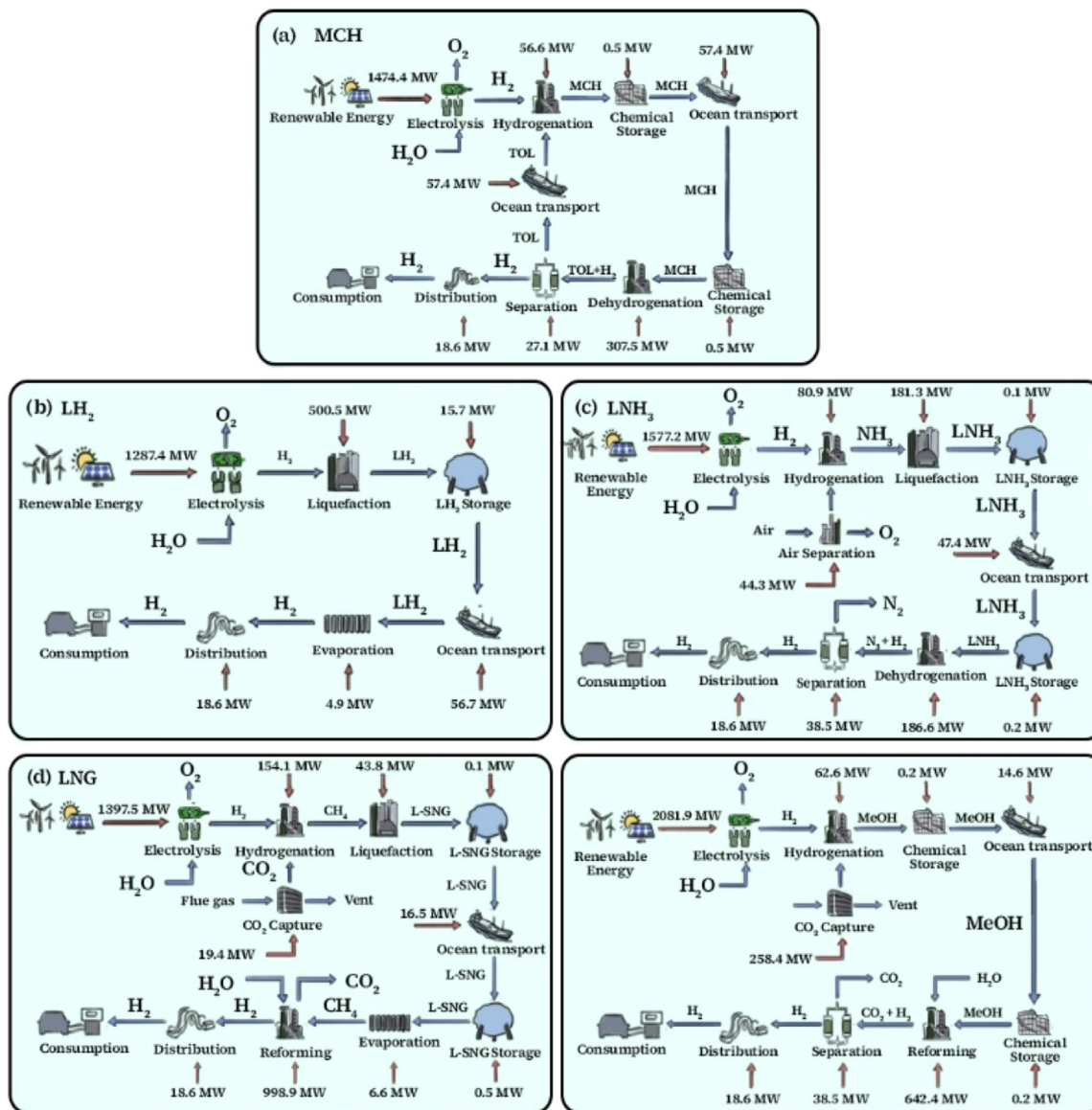


Fig. 14. Energy demand in various hydrogen carriers [245,246].

electrolysis, utilizing renewable energy sources. The findings indicated that the toluene-based LOHC system exhibited a reduced Global warming potential (GWP) in comparison to compressed hydrogen and liquid hydrogen systems when hydrogen was generated through electrolysis utilizing renewable energy. Nevertheless, when hydrogen was generated via SMR, the GWP of the LOHC system exceeded that of compressed hydrogen, while remaining lower than that of liquid hydrogen.

Similarly, Wulf et al. [236] critically analyzed the environmental impacts of different hydrogen transportation methods like pipelines, high-pressure trucks, and LOHCs across the entire lifecycle, from hydrogen production to its final delivery. In a typical analysis, the impact evaluation was on the changes in key parameters such as energy sources and transportation distance. Their result showed that pipeline transportation is the most environmentally favorable option due to its lower energy demand and emissions, while LOHC might be preferable for shorter distances economically, but not environmentally. The environmental impact assessment for hydrogen supply systems operating at 10 t/d and 80 t/d production rates over a 400 km transport distance is illustrated in Fig. 13. Analysis indicates that LOHC transport systems demonstrate the highest environmental burden in scenarios

characterized by low hydrogen demand and extended transport distances, with significant impacts observed across various categories: climate change, ozone depletion, acidification, resource depletion, and freshwater eutrophication. The study identifies heat production as the primary contributor to climate change impacts in LOHC systems. For acidification, the dehydrogenation process accounts for nearly half of the transport-related emissions, with road transport and electricity for compression also playing significant roles. The production of DBT, driven by additional electricity requirements, notably affects freshwater eutrophication. While these environmental burdens are present, they are generally small compared to the impacts associated with hydrogen production itself. The study notes that electricity consumption, particularly from photovoltaic (PV) sources, is a key factor in abiotic resource depletion due to the high resource demand of PV electricity generation.

Integrating LOHCs with renewable hydrogen production systems has been shown to reduce the carbon footprint of LOHC systems. Reuß et al. [237] explored the potential of coupling LOHC systems with wind-powered electrolysis for hydrogen production and found that this approach could result in a significant reduction in GWP (1.9 kg CO₂-eq per kg of hydrogen) compared to fossil-based hydrogen production.

Additionally, the type of LOHC material can influence the environmental impact associated with LOHC production and disposal [238]. A comparative LCA analysis of dibenzyltoluene-perhydro-dibenzyltoluene (DBT-PHDBT) and toluene-MCH system by Bontron et al. [239] revealed that the former has a lower environmental impact, emitting over twice the CO₂ equivalent per kilogram of hydrogen. Likewise, a comparative analysis of 2 LOHC compounds, DBT and toluene, reveals similar overall environmental profiles with subtle distinctions. DBT production generates higher emissions than toluene; however, toluene's advantage is offset by its greater heat and electricity demands during dehydrogenation [240].

The overall energy consumption of each adopted technology represents a critical parameter influencing both the economic viability and environmental impact of various hydrogen carrier pathways. The energy requirements for hydrogen recovery from LOHCs have been a subject of concern. Despite their economic advantages, LOHCs demonstrate the least favorable environmental profile, exhibiting the highest ecological impact among hydrogen storage and transport options [236,237]. This adverse environmental effect is primarily attributed to the high energy requirements of the dehydrogenation process, which can result in significant greenhouse gas (GHG) emissions, particularly when heat is supplied by natural gas combustion. However, utilizing produced hydrogen for the dehydrogenation reaction substantially mitigates the environmental impact of this step [241].

When compared to circular hydrogen carriers such as methane and methanol, which necessitate hydrogen recovery at temperatures exceeding 800 °C and 500 °C, respectively, LOHCs demonstrate a significant advantage. The dehydrogenation process for LOHCs typically occurs at approximately 250 °C, resulting in substantially lower energy demand for hydrogen recovery [242–244]. A comprehensive analysis conducted by Li et al. [245,246] provides valuable insights into the energy demand metrics associated with major chemical pathways for hydrogen delivery (see Fig. 14).

Among the studied scenarios, the methanol-based hydrogen delivery pathway exhibited notably lower hydrogen efficiency. This reduced efficiency is primarily attributed to the limited conversion rate in the methanol synthesis reaction, as reported by previous studies [247]. In the case of liquid hydrogen delivery, the liquefaction process emerged as a significant energy consumer [248]. This observation aligns with established knowledge regarding the substantial energy requirements for hydrogen liquefaction, which involves cooling hydrogen to extremely low temperatures. The energy intensity of this process highlights the need for continued research into more efficient liquefaction technologies or alternative storage methods for long-distance hydrogen transportation. For the substitute methane scenario, the steam reforming step required for hydrogen release proved to be particularly energy-intensive [249]. When comparing the energy demand specifically for the dehydrogenation process across different carriers, a clear hierarchy emerged. Ammonia demonstrated the lowest energy requirement for dehydrogenation, followed by MCH, methanol, and finally methane. Across all examined pathways, electrolysis consistently emerges as the most energy-intensive step, highlighting a universal challenge in hydrogen production regardless of the subsequent storage and delivery method employed. The observed variations in energy demand among different scenarios can be attributed to the varying efficiencies of hydrogenation processes and subsequent steps in each pathway. To facilitate a meaningful comparison, all scenarios were normalized to the final consumption stage, providing a standardized basis for evaluation.

4.2. Economic feasibility and market potential

Alongside environmental issues, the economic viability and market potential of LOHCs are essential for their extensive implementation in carbon-neutral hydrogen production and use. Although LOHCs provide benefits for safety, handling convenience, and storage capacity relative

to compressed or liquefied hydrogen, their economic feasibility continues to pose a significant barrier. The current costs associated with LOHCs are comparatively elevated in relation to alternative hydrogen storage methods, chiefly owing to the expenditures of carrier materials and the energy-demanding hydrogenation and dehydrogenation processes [27,39,48,250,251]. The research conducted by Le et al. [39] approximated the expenses associated with hydrogen storage and transportation utilizing dibenzyltoluene (H18-DBT) to be around \$5–7/kg H₂, markedly exceeding the costs of compressed or liquefied hydrogen storage. Future cost forecasts indicate that LOHC systems may attain more economic viability when technological advancements occur and economies of scale are realized. The research conducted by Ahn et al. [252] anticipated that the expenses associated with hydrogen storage and delivery via LOHCs may diminish to \$2–3/kg H₂ by 2030, propelled by advancements in carrier material synthesis, process optimization, and the establishment of larger-scale production facilities.

The economic feasibility of LOHCs is affected by the market dynamics and regulatory environment pertaining to hydrogen energy systems. The rising demand for clean hydrogen, propelled by decarbonization initiatives and the expansion of hydrogen-centric industries, such as fuel cell automobiles and green steel manufacturing, may provide a conducive market environment for the adoption of LOHC. Government policies and incentives that promote hydrogen infrastructure development, carbon pricing mechanisms, and investments in research and development can expedite the commercialization of LOHC technology [253–257]. Several companies and research initiatives are actively working on the development and commercialization of LOHC systems. For example, Hydrogenious LOHC Technologies, a spin-off from the Friedrich-Alexander University Erlangen-Nürnberg, has developed a commercially available LOHC system using dibenzyltoluene (H18-DBT) as the carrier material [258,259]. Additionally, the European Union's (EU's) Horizon 2020 research program has funded projects like HYDRA and SHEC, focusing on the development of sustainable and cost-effective LOHC systems for hydrogen storage and transportation [260,261].

Niermann et al. 2019 [22] conducted a techno-economic analysis of an LOHC system based on DBT, considering various hydrogen production pathways and transport distances. The study found that the levelized cost of hydrogen (LCOH) for the DBT-based LOHC system ranged from 4.18 to 5.46 €/kg H₂, depending on the hydrogen production method and transport distance. These costs were competitive with compressed hydrogen and liquid hydrogen systems for long-distance transport scenarios (> 500 km). The authors also projected that the LCOH of the LOHC system could decrease to 2.77–3.63 €/kg H₂ by 2030, driven by improvements in catalyst performance and process optimization. Reuß et al. [93] demonstrated that LOHC-based pathways offer significant economic advantages for hydrogen supply in the mobility sector, especially for smaller-scale demand scenarios. In these contexts, LOHC systems outperform both compressed and liquefied hydrogen storage methods. However, it is crucial to note that the use of LOHCs as storage or transport media corresponds to the highest energy demand among the compared options. This high energy requirement necessitates the implementation of effective heat integration strategies and the availability of low-cost renewable heat sources to address challenges related to low dehydrogenation pressure and potential high GHG footprint [262].

Given the substantial renewable electricity potential from wind and solar sources in many regions, LOHCs present significant opportunities for domestic hydrogen storage, transport, and utilization. This advantage extends to long-distance hydrogen export scenarios, positioning LOHC technology as a promising solution for the development of a global hydrogen economy. The economic advantages of LOHC systems, particularly in smaller-scale applications and when integrated with renewable energy sources, suggest that this technology could play a crucial role in the transition to a hydrogen-based energy infrastructure. However, the high energy demand associated with LOHC

Table 9
Summary of selected studies on GWP and economic analysis of LOHC carriers

LOHC system	Capacity	Cost and emission	Key findings	Refs.
MCH-based	50 tons H ₂ /day	1.88 €/kg H ₂ and 5.5–12.8 kg CO ₂ -eq/kg H ₂	The total cost for storage, conversion, and transportation is lower on LOHC systems	[237]
DBT-based	100 tons H ₂ /day	2.1–2.8 €/kg H ₂ and 10.2 kg CO ₂ -eq/kg H ₂	The dehydrogenation process accounts for about 46% global warming impact for 11,000 km transport	[263]
H18-DBT-based	10 tons H ₂ /day	3.0–4.0 €/kg H ₂ and 4.2 kg CO ₂ -eq/kg H ₂	The LOHC system accounts for 11.6% of the electricity cost, thereby exerting a negligible impact on overall cost-effectiveness.	[264]
MCH-based	40 tons H ₂ /day	3.5–4.5 €/kg H ₂ and 10.11 kg CO ₂ -eq/kg CO ₂	LOHC shows the highest GHG emissions in short transport distances	[265]
DBT-based	88 tons H ₂ /day	2.5–3.5 €/kg H ₂ and 3.58 kg CO ₂ -eq/kg H ₂	The huge energy requirement during the DBT post-treatment dehydrogenation process is the main input of the highest GHG emissions	[266]
Toluene-based	50 tons H ₂ /day	8.88 €/kg H ₂ and 5.84 kg CO ₂ -eq/kg H ₂	Transport using liquid organic hydrogen carriers is more economical than in a liquefied state, but not environmentally friendly	[235]

DBT = dibenzyltoluene; GWP = Global warming potential; GHG = greenhouse gas; LOHC = liquid organic hydrogen carrier; MCH = methylcyclohexane.

systems underscores the importance of continued research and development efforts focused on improving energy efficiency, particularly in the hydrogenation and dehydrogenation processes. Table 9 gives summaries of some selected studies on the economic analysis of the LCOH system. It shows the performance of LOHC systems compared to the available alternatives, such as Compressed H₂ and liquid nitrogen.

5. Technological advancements and innovations in LOHCS

In this section, we examine the latest technological breakthroughs in LOHC systems, focusing on innovations that address current challenges. Recent progress has focused on areas such as increasing the practical HSC and enhancing energy efficiency to reduce costs for large-scale applications. The section also explores emerging approaches to sustainable carriers and substrate sources, highlighting solutions that balance technical performance with environmental considerations. Particular attention is given to breakthroughs in LOHC compounds and advanced catalysis as they play a crucial role in improving the practical HSC of LOHCs. However, a detailed analysis of potential developments and their impact on the future of LOHC technology is also provided.

5.1. Advances in LOHC compounds

Conventional LOHCs, such as benzene-cyclohexane, toluene-MCH, and naphthalene-decahydronaphthalene, are well-studied but face limitations, including high dehydrogenation temperatures (200–350°C), energy inefficiency, and catalyst degradation. DBT, a safer and more stable option, also requires high dehydrogenation temperatures, which can impact its economic feasibility [183]. [267] tested DBT mixture solution containing dibenzylbenzene (DBB) and dibenzylethylbenzene (DBEB) as potential LOHC, finding that DBEB is viable, though with slower hydrogenation, while DBB has faster hydrogenation but lower thermal stability, limiting its use. Optimal hydrogenation occurred at 180 °C with Ru/Al₂O₃ catalyst, and high yields were achieved at 100 bar pressure, suggesting that lower pressures with continuous hydrogen supply could also be effective. A microkinetic model was developed, accurately predicting concentration trends and temperature effects based on the Langmuir-Hinshelwood mechanism. Highest yields were 100% for H18-DBB, 85.75% for H18-DBT, and 82.98% for H18-DBEB, aligning well with model predictions.

Innovations in LOHC chemistry have introduced heteroatom-enriched compounds, such as nitrogen-doped aromatics, that offer reduced dehydrogenation temperatures and enhanced stability. N-ethyl-carbazole (NEC) is highlighted for its efficient hydrogen release at lower temperatures and improved kinetics, though it faces economic and practical limitations due to its cost and physical state at room temperature. N-propylcarbazole offers similar efficiency and stability benefits, with slightly reduced HSC but improved suitability for long-term storage.

Biomass offers a renewable alternative for producing LOHCs, with its unique carbon-balance properties and the ability to produce aromatic and nitrogen-doped compounds. Biomass-derived LOHCs offer sustainability and economic viability through a pathway encompassing direct thermal cracking, catalytic conversion of lignocellulosic biomass, and alcohol production from biomass fermentations. Through thermal catalytic conversion and ammonification (TCC-A), nitrogen-rich biomass is converted into N-heterocyclic compounds like indoles, pyridines, and pyrroles. Phenazine-based systems show potential as they leverage renewable sources like lignin to reduce costs and environmental impact, although they require additional handling for solvent separation [67]. Alcohol-based LOHC systems are also gaining attention due to their low cost, broad availability, and lower dehydrogenation temperatures, which present potential energy savings. This ease of management further positions alcohol-based systems as a viable alternative within the LOHC landscape [22]. Catalysts and operating conditions have been

optimized to produce these compounds with high selectivity, providing a sustainable hydrogen storage option [41,268,269].

5.2. Catalyst improvement for hydrogenation and dehydrogenation processes

Innovations in catalyst design are advancing LOHCs towards broader commercial viability by improving the efficiency of hydrogenation and dehydrogenation processes. Obstacles, including high dehydrogenation temperatures, catalyst degradation, and energy demands, are being addressed in this regard.

Bi/Polymetallic catalysts have demonstrated enhanced performance in accelerating the kinetics and improving the efficiency of hydrogenation/dehydrogenation processes in LOHC systems. Polymetallic systems capitalize on the synergistic effects of combined metals to optimize C-H bond cleavage, reaction speed, and product selectivity. For instance, combining Ni, Cu, and Sn with noble metals improves both catalytic activity and economic feasibility. Such innovations make polymetallic catalysts a promising avenue for scaling LOHC technologies [144]. Recent improvements and innovations in catalysts for the tetrahydroquinoline/quinoline LOHC system have enhanced its efficiency and sustainability. In 2019, Beller's group introduced a nickel silicide catalyst on fumed silica that facilitated both hydrogenation and dehydrogenation, maintaining activity over multiple cycles. The same year, Co-Phen@C, a durable cobalt-based catalyst, was developed for the reversible hydrogenation of N-heterocycles. A bimetallic Pd-Ni catalyst in MIL-100(Fe) demonstrated remarkable activity for reversible reactions in water under mild conditions. Further, bimetallic Pd-Au catalysts on multiwall carbon nanotubes provided enhanced (de)hydrogenation activity. A novel light-driven dehydrogenation method, using an iridium(III) complex, also emerged as a promising low-energy alternative [41,270–272].

Other notable progress includes a Pd/Al₂O₃-YH₃ catalyst, which efficiently facilitated hydrogen release at lower temperatures, and a Pd-Rh nanoparticle catalyst that demonstrated high hydrogen uptake and release within hours, maintaining efficiency across multiple cycles [273,274]. Catalysts developed for the bicyclohexyl/biphenyl and perhydro-terphenyls/terphenyls LOHC systems have achieved significant improvements by targeting selectivity and minimizing side reactions like cracking and coking. The introduction of 1% Pt/Mg₂NiHx, along with terphenyl substrates, allowed nearly complete dehydrogenation selectivity, enhancing system longevity to 20 cycles. Additionally, high-purity hydrogen release has been achieved using Pd/C and Ru/Al₂O₃ catalysts in biphenyl/diphenylmethane systems, maintaining thermal stability and supporting PEM fuel cells with minimal material degradation over multiple cycles [41].

Conversely, photocatalysis, such as acceptorless dehydrogenation, offers a promising alternative by enabling hydrogen release at ambient temperatures through visible light activation. Materials such as modified TiO₂ and quantum dots (CdSe/CdS) have demonstrated potential in reducing energy costs while integrating solar energy [28,49]. Notable advances include the development of hybrid Pt@TiO₂-N catalysts, which facilitate both hydrogenation and photoinduced dehydrogenation at room temperature. These catalysts have achieved high conversion rates under visible light irradiation, demonstrating the feasibility of sustainable, light-driven LOHC systems [41,275,276]. Advancements in hydrogenation and dehydrogenation catalysts for the benzene/CHE LOHC system have introduced high-efficiency noble metal catalysts like Pt, Pd, and Ru in nanoparticle-supported catalysts. These catalysts, such as nano-Pd/CeO₂ offer high surface areas and efficiency in (de)hydrogenation cycles.

5.3. Innovations in energy-efficient strategies for LOHCs

With increased focus on energy-efficient dehydrogenation, an innovative autothermal dehydrogenation process for LOHC systems,

allowing hydrogen release without external heat or burning hydrogen, is mentioned by [277]. By integrating exothermic partial oxidation to provide necessary heat, it improves hydrogen usability by up to 30%. Selective catalysts enable efficient and stable partial oxidation, with a 93% selectivity achieved for diphenylmethane (H0-DPM) to benzophenone (H0-BP). The full cycle was successfully demonstrated, showing feasibility for real product mixtures, which supports potential commercial applications, particularly in heavy-duty transport where compact, heat-independent systems are essential. Advances in molecular engineering, such as introducing alkali metals in hydrogen bonds, reduce dehydrogenation enthalpy and improve storage capacity. Examples include sodium phenoxide-cyclohexanolate pairs that lower dehydrogenation temperatures, making hydrogen release possible below 150 °C, thus enhancing LOHC practicality for industrial use. Effective thermal management techniques, including waste heat recovery and the use of external energy fields, have further enhanced LOHC efficiency by addressing high energy demand in dehydrogenation.

5.4. Integration of LOHC systems with renewable energy technologies

Combining renewable energy technologies with LOHC systems is essential to addressing the intermittent problems associated with renewable energy sources such as solar and wind power [278]. Excess energy can be converted into hydrogen using LOHC systems, which are designed to make it easier to store the energy produced by these renewable sources. This hydrogen can then be extracted and utilized during periods characterized by low generation, thereby augmenting energy security and reliability [49]. One particularly promising strategy entails the coupling of LOHC systems with electrolysis, which is powered by renewable energy sources. In this configuration, excess electricity derived from solar or wind sources is employed for hydrogen production via electrolysis; this hydrogen is subsequently stored within LOHC systems. During intervals marked by heightened energy demand or diminished renewable generation, the stored hydrogen can be released and converted back into electricity through fuel cells or combustion processes. [279] underscored the potential of such closed-loop systems to realize round-trip efficiencies that range from 40% to 50%. This efficacy is largely influenced by the incorporation of heat recovery mechanisms and advancements in electrolysis technology. However, challenges remain in optimizing these systems for broader implementation, because the interplay between storage, generation, and demand necessitates intricate engineering solutions. Although the pathway is fraught with complexity, the potential rewards justify the exploration of LOHC systems in renewable energy applications.

The direct coupling of LOHC systems to renewable power plants significantly augments integration. For example, wind farms can directly charge an LOHC during periods of peak generation, thus effectively serving as a buffer to absorb surplus energy and subsequently release it during intervals of low generation. This methodology is particularly beneficial for offshore wind farms, where the transportation of energy over extensive distances can pose logistical challenges. Storing energy in a chemical form facilitates a more efficient delivery and utilization of renewable energy [22]. LOHC systems can be seamlessly integrated into hybrid renewable energy systems that employ multiple energy sources (such as solar and wind). Although such configurations optimize energy input utilization and mitigate supply fluctuations, they also ensure a stable energy output. However, the complexity of these systems necessitates careful consideration. Because the integration of diverse energy sources introduces variability, this approach demands meticulous management to prevent inefficiencies [225].

The decentralized deployment of LOHC systems, especially when implemented alongside local renewable energy sources, offers significant opportunities for enhanced energy resilience and diminished transmission losses. Such systems empower communities to autonomously manage their energy requirements, thereby fostering a state of energy self-sufficiency. However, remote regions abundant in

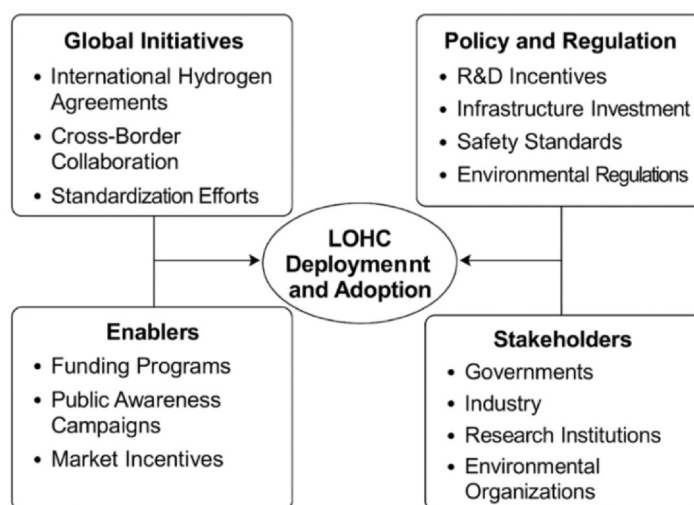


Fig. 15. Policy and stakeholder ecosystem for LOHC adoption: a schematic representation of the key governmental, industrial, financial, and societal actors involved in LOHC implementation, along with their interdependencies and enabling policy instruments. LOHC = liquid organic hydrogen carrier.

renewable resources can derive pronounced advantages from these frameworks, as they mitigate dependence on fossil fuels or external power grids. The amalgamation of LOHC systems with renewable energy technologies represents a fundamental advancement in energy storage solutions [280].

5.5. Role of AI and automation in improving LOHC efficiency

The role of artificial intelligence (AI) and automation in enhancing LOHC efficiency cannot be overstated. Such technological integrations promise to optimize processes and improve overall performance metrics. This synergy highlights the potential of AI to optimize processes in LOHC technology [281–284]. To elaborate, consider the following elements:

- a) *Data-driven optimization:* AI algorithms possess the capacity to scrutinize extensive datasets derived from experimental inquiries and operational workflows. These analyses reveal intricate patterns and relationships that are instrumental in enhancing LOHC performance. Machine learning methodologies, which include both supervised and unsupervised learning frameworks, can be employed to discern optimal conditions for hydrogenation and dehydrogenation reactions, predict catalyst efficacy, or even propose innovative materials predicated on pre-existing datasets [285].
- b) *Accelerated Material Discovery:* AI-driven high-throughput screening greatly accelerates the discovery of novel LOHCs. By automating synthesis and characterization and applying machine learning models to predict material performance, researchers can rapidly identify compounds with optimal hydrogen storage and release properties [286,287].
- c) *Lifecycle Assessment:* Moreover, AI can substantially enhance LCA methodologies pertinent to LOHC systems. This enhancement allows researchers to systematically evaluate the environmental and economic ramifications of various LOHC formulations and processing technologies. By meticulously analyzing historical data and modeling prospective scenarios, AI can offer invaluable insights aimed at optimizing resource utilization and minimizing the carbon footprint associated with LOHC technologies [288].
- d) *Decision Support Systems:* Furthermore, the integration of AI with decision support systems serves to empower stakeholders in navigating the intricate logistics associated with LOHC transportation and storage. By utilizing AI models for the analysis and forecasting of market trends, resource availability, and regulatory fluctuations, companies are positioned to make informed and strategic decisions

about LOHC deployment and investment. However, it is essential to recognize that the successful implementation of these AI-driven methodologies requires careful consideration of the complexities involved.

The prospective landscape of LOHC technologies seems quite auspicious, primarily propelled by ground-breaking research and the revolutionary integration of AI and automation. However, sustained investment is essential; collaborative efforts will play a pivotal role in this endeavor.

6. Policy and regulatory landscape for LOHC adoption in the hydrogen economy

With the growing instability in the global oil market, the decline of conventional oil reserves, and the increasing demand for clean energy, the transition from fossil fuels to renewable and more stable, clean energy sources has become imperative [289]. The EU is actively pursuing carbon neutrality and pollution reduction goals in the energy sector by 2050, as outlined in the 2020 European Green Deal. Hydrogen, a carbon-free energy carrier, is viewed as the key driver of this transition. It is projected to account for 60% of the GHG emission reductions in the final phase of the energy transition [290]. The Energy Transition Commission emphasizes hydrogen's crucial role in decarbonizing industries where direct electrification is either impractical or too costly, such as steel production, long-distance shipping, and aviation. Consequently, hydrogen demand is expected to grow significantly in many energy markets [291]. The overall policy landscape and the interaction between global stakeholders, regulatory initiatives, and funding mechanisms supporting LOHC technology adoption are summarized in Fig. 15.

With the current global policy frameworks, governments worldwide have significantly increased research and funding for the hydrogen economy (see Table 10). Recent studies highlight substantial advancements in hydrogen production, storage, and transportation technologies, leading to cleaner and more efficient methods for hydrogen production, storage, and transportation [291].

Global support has accelerated the development of zero- and low-carbon techniques, such as electrolysis and natural gas reforming, enabling the production of various hydrogen types, including green, blue, and pink hydrogen. Projections suggest that by 2030, annual hydrogen production could surpass 20 million tons, marking a 30% increase compared to 2022 levels. Approximately 50% of the planned hydrogen projects are still in the feasibility stage, over 45% are in early

Table 10
Summary of global LOHC-related policies

Region	Key policies	Funding support	National roadmap inclusion	Notes	Refs.
EU	Fit-for-55, IPCEI Hydrogen	€5B +	Yes	Strong R&D backing	[292]
Japan	Basic hydrogen strategy	¥2T +	Yes	Pioneer in LOHC R&D	[293]
USA	Hydrogen shot initiative	\$8B	Yes	Focused on carriers & storage	[294,295]

LOHC = liquid organic hydrogen carrier.

Table 11
Hydrogen policy adoption levels by some countries (2020–2021)
Modified from [291].

Country	Policy level	Focus area
China	R&D Program	A combination of green and fossil-based hydrogen
Portugal	Vision Document	Green hydrogen
Morocco	Roadmap	Green hydrogen
Finland	Strategy	Green hydrogen
Denmark	Strategy	Green hydrogen
Uruguay	Strategy	Green hydrogen
Netherlands	Strategy	Combination (green + fossil-based)
Paraguay	Strategy	Green hydrogen
Australia	Strategy	Green hydrogen
Spain	Strategy	Green hydrogen
France	Strategy	Green hydrogen
Germany	Strategy	Green hydrogen
Norway	Strategy	Combination (green + fossil-based)
European Union	Strategy	Green hydrogen
UK	Strategy	Combination (green + fossil-based)
South Korea	Strategy	Green hydrogen
Chile	Strategy	Green hydrogen
Japan	Strategy	Green hydrogen
Russia	Strategy	Fossil-based hydrogen
Italy	Strategy	Green hydrogen

development, and roughly 4% have either begun construction or secured necessary funding [2,22,296,297]. Table 11 illustrates the global landscape of countries actively pursuing hydrogen policies at varying levels, including R&D programs, vision statements, roadmaps, and strategic frameworks. Notably, in the recent period (2020–2021), several nations have advanced their strategies, though the focus and scale of these plans vary. Some countries emphasize green hydrogen, others on fossil-based hydrogen, or a combination of both. Additionally, the scope ranges from having no defined targets to setting highly ambitious, quantifiable goals for hydrogen production and electrolyzer capacity.

Notably, in the recent period (2020–2021), several nations have advanced their strategies, though the focus and scale of these plans vary. Some countries emphasize green hydrogen, others on fossil-based hydrogen, or a combination of both. Additionally, the scope ranges from having no defined targets to setting highly ambitious, quantifiable goals for hydrogen production and electrolyzer capacity. Fig. 16 provides a clear overview of regional trends and the varying levels of commitment to hydrogen as a clean energy carrier. The map illustrates the global adoption of hydrogen policies, highlighting key countries and their respective focus areas in the hydrogen economy as of 2020–2021. Countries are color-coded based on their primary hydrogen focus: green for those prioritizing green hydrogen, blue for fossil-based hydrogen, and yellow for countries pursuing a combination of green and fossil-based hydrogen.

Historically, one of the primary challenges for the hydrogen economy has been the storage and transportation of hydrogen. Hydrogen's low energy density necessitates larger storage tanks compared to conventional fuels, while the energy demands of high-pressure compression and liquefaction reduce supply chain efficiency. Moreover,

hydrogen's reactivity can cause material embrittlement, increasing the risk of leaks and cracks, while its highly flammable nature poses safety hazards during storage and transport. To address these obstacles, researchers have explored hydrogen carriers as a potential solution. One promising approach is the use of LOHCs, which are organic compounds that chemically bond with hydrogen, allowing it to be stored and transported in liquid form under ambient conditions. LOHCs address many of the challenges associated with compressed or liquefied hydrogen, providing a safer, scalable solution that can utilize existing infrastructure. LOHCs have also been found to be the most cost-effective option for long-distance transport (greater than 200 km) and competitive over shorter distances, especially in terms of transport costs [46]. This positions LOHC technology as a crucial component in overcoming hydrogen storage and transport challenges, with the potential to play a pivotal role in the emerging hydrogen economy [36]. In this section, we explore how the global policy and regulatory landscape is evolving to address these challenges in hydrogen storage and transportation.

6.1. Global and regional initiatives for LOHC technology

Herein, we examine the global and regional policies on the safe storage and transport of hydrogen through LOHCs. By promoting and funding LOHC technology, governments are helping to address major challenges associated with hydrogen transportation. For instance, Hydrogenious LOHC Technologies, a German company specializing in hydrogen storage and transport solutions, utilizes LOHC technology to bond hydrogen to benzyltoluene through hydrogenation, which is subsequently released from the oil by dehydrogenation [2]. Recently, the German and Bavarian governments awarded a grant of €72.5 million to Hydrogenious LOHC Infra Bavaria for the Green Hydrogen@Blue Danube project. This initiative aims to establish a large-scale hydrogen supply chain using LOHC technology to safely transport green hydrogen from Eastern Europe to Southern Germany. Recognized by the European Commission in February 2024 as an Important Project of Common European Interest under the hydrogen "Hy2Infra" initiative, this project is pivotal to Europe's efforts in building a robust hydrogen economy. It also supports a sustainable and reliable hydrogen supply for industries in Central Europe, advancing the region's sustainability objectives.

The UK and Scottish governments, in partnership with international organizations such as Axens, Chiyoda, EnQuest, ERM, Koole Terminals, Port of Rotterdam, Shetland Islands Council, Storegga, and the Net Zero Technology Center, have launched the Hydrogen Highway project. This initiative aims to enable hydrogen transport from Scotland to Rotterdam utilizing the LOHC technology. These efforts are in line with broader European and global objectives to boost hydrogen production and importation, including the European Commission's target of importing 10 million tons of hydrogen by 2030.

A key player in researching LOHC technology is the Helmholtz Institute Erlangen-Nuremberg for Renewable Energy (HI ERN). Established in 2013, HI ERN is an independent institution that receives substantial funding and support from governmental agencies, which greatly contributes to the advancement of LOHC technology. In 2021, Siemens Mobility and HI ERN formalized their collaboration through an agreement focused on joint research into the use of LOHC technology in

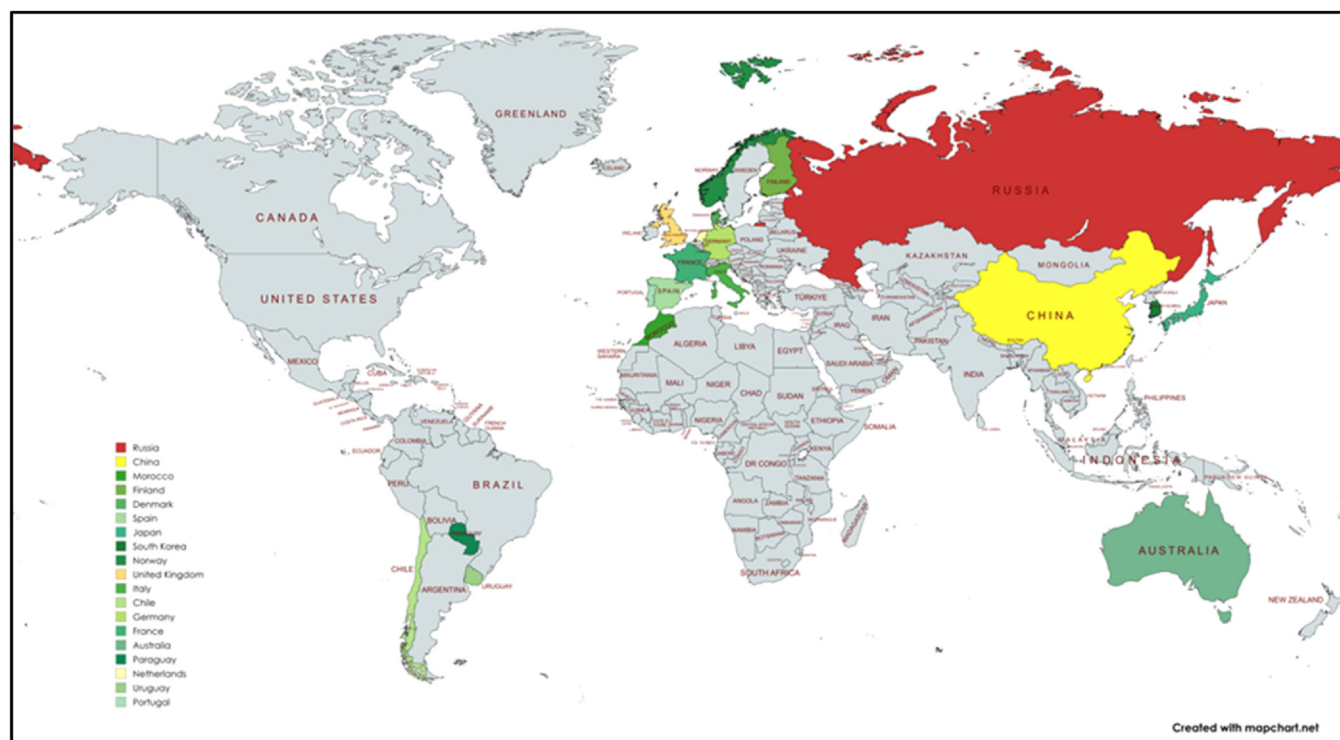


Fig. 16. Global map highlighting key countries and their hydrogen policy focus areas (2020–2021). Green shade colorations = green hydrogen; red coloration = fossil-based hydrogen; yellow = combination of green and fossil-based hydrogen.

rail transport. This partnership aims to develop innovative hydrogen applications for rail traction, allowing hydrogen to be safely stored in an organic carrier liquid that releases it as needed, thereby improving the safety and efficiency of hydrogen storage and transport within rail systems.

In 2023, CWP Global, a prominent renewable energy development firm, and Hydrogenious LOHC Technologies, a German leader in LOHC technology, entered a Memorandum of Understanding to conduct a joint feasibility study. This study focuses on establishing a hydrogen transport chain capable of delivering 500 tons of green hydrogen daily from Morocco to Europe using Hydrogenious' LOHC technology. The study's initial efforts will be centered on the 15 GW AMUN project near Tan-Tan, Morocco, which is aimed at producing green hydrogen for local ammonia production, contributing to the decarbonization of sectors like fertilizer manufacturing and maritime shipping. In pursuit of greater safety, flexibility, and cost-efficiency, the partners are also exploring the option of directly transporting green hydrogen to Europe via LOHC technology, leveraging existing infrastructure for more efficient long-distance transport.

Recently, the UK Government's Department for Energy Security and Net Zero initiated a funding initiative on Low Hydrogen Supply 2 (HYS2) Competition. The competition seeks to accelerate the commercialization of low-carbon hydrogen solutions by funding projects that address key challenges in hydrogen production, storage, and transportation. It is part of the UK's broader goal to support the hydrogen economy as a means to achieve net-zero carbon emissions by 2050. One significant project funded under the HYS2 competition focuses on bulk hydrogen storage through the use of LOHC, alongside the development of innovative electrolyzer technologies and hydrogen transport systems. These initiatives often involve partnerships between academic institutions, industry leaders, and government bodies to leverage expertise and drive innovation in hydrogen supply solutions. These partnerships and projects highlight the growing global support and favorable policies driving the hydrogen economy's rapid expansion, as governments and companies invest in sustainable energy solutions.

6.2. Standards and safety regulations for LOHC use

Whilst the use of organic carriers in hydrogen storage and transportation poses less risk of explosive decompression compared to compressed or liquefied hydrogen, they still require careful management of chemical hazards specific to each carrier to mitigate the risks associated with (de)hydrogenation processes. For instance, Spatolisano et al. [240] pointed out that the hydrogenation process, which typically involves gaseous hydrogen, can generate significant pressure within the system. Without proper pressure relief mechanisms, this could lead to leaks or catastrophic failures, posing significant safety hazards. Additionally, the highly endothermic nature of the hydrogenation reaction presents challenges. The presence of hydrogen, combined with the heat needed for the process, introduces flammability risks. Without adequate temperature control, unsafe conditions could arise, leading to system malfunctions or hazardous reactions. High temperatures may also heighten the risk of thermal runaway if not managed properly. Markiewicz et al. [241] also noted that many organic compounds used as hydrogen carriers are volatile, flammable, and lipophilic, requiring careful consideration to ensure that the carriers chosen are not only effective but also environmentally friendly and nontoxic. These risks highlight the need for robust safety protocols and comprehensive risk assessments when utilizing LOHC technology for hydrogen storage and transport. Proper design, safety protocols, and training are essential to minimizing these risks.

LOHC technologies, like other hydrogen-related systems, require compliance with a wide range of international and national safety standards. These include ISO regulations for hydrogen quality, transport safety rules such as agreement concerning the international carriage of dangerous goods by road and regulations concerning the international transport of dangerous goods by rail, chemical safety measures under Registration, Evaluation, Authorization, and Restriction of Chemicals (REACH) and Globally Harmonized System of Classification and Labeling of Chemicals, and specific fire protection and storage guidelines like National Fire Protection Association 2

Hydrogen Technologies Code and American Society of Mechanical Engineers standards. LOHC safety protocols ensure hydrogen remains stable and secure during long-term storage, and that proper risk management practices are in place for transportation and operational environments. Several safety regulations currently exist to address the risks of LOHC usage. In the U.S., OSHA provides guidelines for handling hazardous materials, which apply to LOHCs and the hydrogenation process. These regulations emphasize safe working conditions and aim to minimize chemical handling risks. The European Union's REACH regulation provides a framework for chemical safety assessments. ISO standards, such as ISO 14687, ISO/TS 19880-1, and others, focus on hydrogen fuel quality and indirectly affect LOHC applications by ensuring the quality of hydrogen used in the process. LOHC manufacturers must provide Safety Data Sheets, detailing the hazards, precautions, and safety measures required for their products [298,299]. Furthermore, the European Committee for Standardization has also contributed to establishing standards for hydrogen technologies, including LOHCs, focusing on risk assessments, safety management, and operational practices. These standards and regulations are essential for ensuring the safe implementation of LOHC systems throughout the hydrogen supply chain.

7. Challenges and prospects of LOHCs

7.1. Technical challenges

The efficiency limitations and energy penalties intrinsic to LOHC systems pose considerable technical challenges: one of the primary issues stems from the endothermic and exothermic characteristics of their hydrogenation and dehydrogenation processes, which demand substantial energy input for hydrogen release, while concurrently producing heat during hydrogen absorption. As a result, the energy efficiency of LOHC systems is compromised because external energy sources must be employed, often leading to cumulative energy losses. This energy intensity influences the overall feasibility and competitive position of LOHC technologies, especially when contrasted with alternative hydrogen storage methods such as compressed gas or cryogenic liquid hydrogen [36]. However, although optimizing reaction conditions and innovating new catalysts are essential for mitigating these energy penalties, such initiatives also raise concerns about catalyst degradation and recycling difficulties.

A significant technical challenge pertaining to LOHCs involves the degradation of catalysts and their recyclability. The processes of hydrogenation and dehydrogenation that are intrinsic to LOHC systems necessitate highly specialized catalysts often derived from precious metals to enable reactions under manageable temperatures and pressures [144]. However, extended use of these catalysts results in degradation due to various phenomena, including sintering, poisoning, and fouling. This degradation not only diminishes efficiency; it also complicates the overall sustainability of the process. Although the initial choice of catalysts is of paramount importance, their longevity remains a critical determinant of the viability of LOHC technology, because without it, the potential benefits may be undermined. This degradation subsequently diminishes reaction efficiency, compelling the need for costly catalyst replacement or regeneration [142]. Moreover, catalyst recycling and degradation represent substantial concerns,

primarily because of their implications for both operational expenses and environmental sustainability. Addressing these challenges requires innovations in catalyst material science (see Table 12). Although the current focus is on developing more durable, abundant, and cost-effective catalytic materials, these must endure repeated hydrogenation-dehydrogenation cycles with minimal performance loss [142].

7.2. Economic and market barriers

High initial costs and market adoption hurdles present a significant challenge for the economic viability of LOHCs. The substantial investment required, encompassing catalyst development, specialized reactors, and the necessary handling and storage infrastructure, creates a formidable barrier to entry. This is particularly true for emerging economies or companies that lack significant capital resources. However, the market adoption of LOHCs also contends with competition from other hydrogen storage and delivery technologies, which often possess more established infrastructures and greater public awareness. Although there exists a comparative lack of familiarity with LOHC technologies among potential investors and industry stakeholders, this knowledge gap further impedes market penetration. Strategies aimed at reducing costs through economies of scale could be effective, but they should also include efforts to enhance stakeholder education and awareness [46]. This dual approach could facilitate broader acceptance of LOHC technologies within the global market. The need for infrastructure development and standardization remains crucial in this context.

For LOHC technologies to be integrated seamlessly into the hydrogen economy, there exists a critical need for infrastructure development and the establishment of standardized protocols. Existing infrastructure for hydrogen storage and transportation is typically tailored to gaseous or liquid hydrogen; however, LOHCs necessitate specialized containment systems and processing units that are compatible with organic carriers. Although standardized regulations and guidelines must be developed, this is essential to ensure the safe, efficient, and environmentally friendly use of LOHCs across diverse applications and regions [301]. Because there is a lack of such infrastructure and standardization (Fig. 17), LOHC technologies encounter significant barriers to scalability and interoperability with other hydrogen-based solutions. Addressing these issues requires concerted efforts from industry, regulatory bodies, and governments to facilitate infrastructure investments and regulatory frameworks.

7.3. Future prospects for LOHCs

Despite myriad technical and economic challenges, LOHCs exhibit considerable long-term potential within the global hydrogen economy [36]. Their distinctive capacity to safely store and transport hydrogen in a nonpressurized liquid state renders them especially attractive for sectors requiring prolonged hydrogen storage and extensive transportation distances.

Furthermore, LOHCs facilitate integration of renewable energy sources; hydrogen produced from solar, wind, or other renewable modalities can be effectively captured and stored within LOHC systems. This, consequently, aids in stabilizing the renewable energy supply chain. As hydrogen's prominence in the energy landscape continues to

Table 12
Categorized challenges and recommendations [21,300]

Category	Challenge	Proposed solution
Techno-Economic	High dehydrogenation cost	Catalyst innovation, integration with waste heat
Regulatory	Transport classification ambiguity	Harmonized hydrogen safety codes
Infrastructure	Lack of carrier-specific logistics	Dedicated LOHC pipelines, modular stations
Safety	Toxicity of carriers	Safer LOHC material development

LOHC = liquid organic hydrogen carrier.

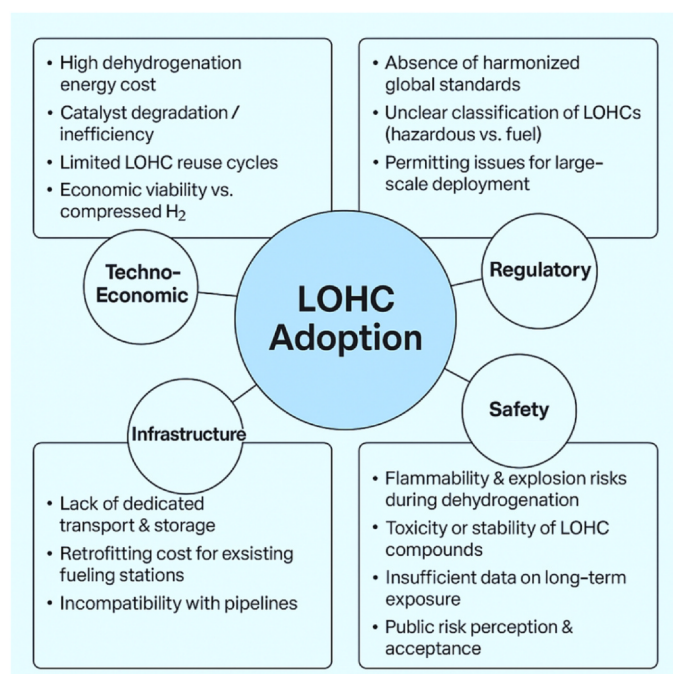


Fig. 17. Conceptual map of key technological and market challenges in the adoption of liquid organic hydrogen carriers (LOHCs), categorized into 4 domains: techno-economic, regulatory, safety, and infrastructure. Each domain encompasses specific barriers such as high dehydrogenation energy costs, lack of regulatory frameworks, safety risks during handling, and insufficient transport infrastructure.

grow, LOHCs are poised to be pivotal enabling technologies [302]. However, regions characterized by underdeveloped hydrogen transport infrastructure may discover that LOHCs are particularly advantageous because they present a feasible alternative to conventional high-pressure hydrogen transport solutions. Strategic pathways for scaling up of LOHC technologies are thus essential to consider.

To fully realize the potential inherent in LOHCs, a complete strategy aimed at scaling this technology is imperative. This undertaking necessitates not only the advancement of research and development to overcome technical obstacles but also the cultivation of collaborations among myriad stakeholders, which include government agencies, private enterprises, and academic institutions. Investment in pilot projects and demonstration facilities can provide crucial insights into operational hurdles and economic feasibility; however, policy support and incentives are vital because they can stimulate early adoption and foster development of requisite infrastructure [252]. Moreover, international collaboration in the establishment of technical standards and best practices can expedite the evolution of a globally interoperable LOHC framework, thereby facilitating the transition toward a hydrogen-centric energy economy.

8. Conclusion

This work reveals that LOHCs play an important role in allowing carbon-neutral hydrogen production, storage, and usage by conducting a complete investigation of LOHCs' recent developments and the economic viability of large-scale implementation. LOHC technology is distinguished by its high energy density, chemical stability, and compatibility with existing fossil fuel infrastructures. Such characteristics make it suitable for use in a variety of sectors and applications. Furthermore, LOHCs provide a viable avenue for incorporating renewable energy sources, which is consistent with global climate goals and promotes a resilient, low-carbon energy future. However, to fully exploit the potential of LOHCs,

improvements in catalyst performance, life cycle evaluation of each LOHC material, and supportive policies that stimulate innovation and adoption are required.

Despite persisting challenges, the possibilities for LOHCs are quite promising as they represent a significant advancement in the construction of a more sustainable energy storage system. Future research should concentrate on optimizing these systems and examining their long-term environmental implications. This is critical to fully realizing the benefit of LOHCs as a hydrogen storage and transport option in the global energy transition. Additionally, issues like scalability and market dynamics need to be addressed.

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CRediT authorship contribution statement

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

Declaration of Generative AI and AI-assisted technologies in the writing process

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