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Original Research Article

Efficient hydrogen transportation via liquid organic hydrogen carriers: Optimizing hydrogenation and dehydrogenation processes

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ABSTRACT
Hydrogen has attracted widespread attention as a carbon-neutral energy source, but developing efficient and safe hydrogen storage technologies remains a huge challenge. Traditional hydrogen transport methods, such as high-pressure gas cylinders and cryogenic liquid hydrogen storage, face safety concerns, high energy consumption, and economic limitations. This research explores the use of liquid organic hydrogen carriers (LOHCs) as a promising alternative for hydrogen transportation. LOHCs enable hydrogen storage through hydrogenation, where hydrogen is chemically bonded to a carrier molecule, and its subsequent release through dehydrogenation. This study focuses on optimizing the hydrogenation and dehydrogenation processes using catalysts such as nickel-cobalt to enhance reaction efficiency and reduce energy consumption. The research evaluates different LOHCs, including N-ethylcarbazole, based on parameters like hydrogen storage capacity, stability, reversibility, and cost-effectiveness. A techno-economic analysis is conducted to assess the feasibility of this method compared to conventional hydrogen transport methods, considering factors such as infrastructure requirements, energy efficiency, and environmental impact. By improving the efficiency of LOHC-based hydrogen transportation, this research aims to contribute to the development of a scalable and economically viable hydrogen supply chain, facilitating the transition to a clean energy economy.

1. Introduction

The depletion of fossil fuels has forced humans to search for alternative energy sources like renewable energy systems derived from the wind, sun, oceans, etc [1]. However, the energy obtained from renewable sources is not consistent since these sources fluctuate under seasonal and meteorological conditions. These situations may lead to a shortage of power in unfavourable weather conditions and produce additional power in favourable weather, which hinders the production of a stable energy system. The electrolysis of water is another technique of saving electricity through the generation of hydrogen [2]. The gravimetric energy density of hydrogen storage is 120 MJKg^{-1}, which can be converted into electrical energy without producing any toxic emissions [3]. Thus, hydrogen has been depicted as a clean energy resource to replace fossil fuels in the coming decades.

The growing concerns over climate change, air pollution, and the depletion of fossil fuel reserves have prompted a global search for cleaner and more sustainable energy solutions. Hydrogen has emerged as a promising alternative fuel due to its abundance, high energy content, and environmentally friendly combustion properties [4]. When used in internal combustion engines (ICEs), hydrogen combusts primarily to form water vapor, offering a significant reduction in greenhouse gas emissions compared to conventional fossil fuels. Despite its benefits, the use of hydrogen in ICEs presents several technical and logistical challenges. These include hydrogen storage and distribution, ensuring safe handling, and addressing combustion characteristics such as high flame speed, wide flammability range, and susceptibility to pre-ignition and knocking. Furthermore, the efficiency and emissions profile of hydrogen ICEs must be optimized to compete with advanced conventional and alternative fuel technologies [5, 6].

Liquid organic hydrogen carriers (LOHCs) have sparked immense interest in both industrial as well as academic fields, based on their potential for hydrogen storage and transportation, safety, capability to undergo reversible dehydrogenation and hydrogenation, cheap and facile thermal management, and strong compatibility with the present infrastructure of gasoline. The H lean form specifically signifies the LOHC and constitutes mainly polycyclic organic aromatic compounds [7-9]. The release and storage of hydrogen gas within organic compounds are very slow processes, and hence, are accelerated in the presence of catalysts. The transportation of hydrogenated LOHCsis feasible at atmospheric pressure, utilizing the existing transportation facilities [10, 11]. The volumetric density is 50 gL^{-1}, while the gravimetric density is over 5 wt% for hydrogen stored using the LOHC storage techniques. The dehydrogenation of the LOHCs releases hydrogen which is free of unrequired gases like CO_2, CO, and their by-products.

2. Brief history of LOHCs

Research studies toward hydrogen storage in LOHCs via hydrogenation/dehydrogenation processes first took place in the early 1980s[12]. Based on the (de)hydrogenation processes, the most predominant task was pointed out as toluene/methylcyclohexane (MCH) system [13]. Following the MCH system research, numerous LOHC concepts have been assessed based on hydrogenation and dehydrogenation criteria for hydrogen storage. In the early 2000s [16], the basic concept of a cyclohexane/benzene LOHC system was investigated by Japanese researchers and they examined similar systems in more detail. Notably, NEC was proposed as a LOHC candidate by Pez et al., and then in-depth research of this carrier material has been continued by various groups. The latter, different research groups have also shown significant interest in alternative azaborine carrier materials. In 2008 [19], Crabtree suggested that N-containing heterocyclics are more advantageous LOHC materials, in terms of ease of H_2 release, safe storage, low vapor pressure, better biodegradability, and simple heat management. Muller et al. proposed that nitrogen-containing aromatic compounds are well-suited for better hydrogen storage based on thermodynamic evaluation, emphasized by their enthalpy changes during hydrogenation. Based on the LOHC concept, a mile stone was reached by the Chiyoda Corporation (Japan; Scheme 2) [21-24]. This company completed a pilot-plant facility for large-scale hydrogenation and dehydrogenation of LOHC materials in 2018. Additionally, numerous potential LOHC candidates were proposed and exploited in practical applications such as decentralized energy storage network, combined heat and power (CHP) systems. Eypasch et al. demonstrated theoretical assumption of energy supply based on LOHC for industrial production plants. Very recently, Niermann et al. indicated that LOHCs are technologically efficient and economically promising safe transportation and storage materials [25].

3. Critical issues in developing LOHC media

According to reported literature, we briefly discuss a few important LOHC characteristic properties and key aspects of LOHC catalytic systems as well as numerous other factors more generally for practical implementation of LOHCs.

3.1 Hydrogenation/dehydrogenation

Usually, reversible hydrogenation and dehydrogenation at ambient temperature conditions is the primary requirement for hydrogen storage LOHC candidates. Thermodynamically, the feasibility of these reactions is strongly influenced by the thermodynamic reaction enthalpies. In particular, aromatic hydrogenation reactions are highly exothermic and thermodynamically favorable (e.g., aromatic benzene ring enthalpy is $\Delta^R h = -68.73$ kJ/mol H_2, but the released energy not typically used. On the other hand, dehydrogenation is endothermic and requires high heat demand which is in the range of 64–69 kJ/mol H_2, and as a result, this reaction is unfavorable both kinetically and thermodynamically (e.g., cyclohexane/benzene and MCH/toluene pairs) [22-28]. In contrast, liquid hydrogen carriers require low heat management though it needs an active catalyst in the dehydrogenation process. Considering these thermodynamic difficulties, Pez and coworkers for the first time suggested in their patent, the use of N-heterocyclics (e.g., NEC) decreases endothermicity and enhance hydrogen release as compared with alicyclics at relatively lower temperatures. Based on Muller et al.'s contribution, the facile reaction enthalpy for an ideal LOHC candidate is about 40 kJ/mol H_2, and they suggested nitrogen-substituted aromatic compounds could reach this requirement easily, despite non-aromatic compounds being barely suitable [27]. Hence, these reports encourage the nitrogen-containing compounds are the focus of interest in developing novel LOHC systems.

3.2 Reaction catalysts for LOHC

Another crucial aspect in LOHC development is selection of the proper catalytic system for hydrogenation and dehydrogenation reactions. As discussed, chemical storage of hydrogen in liquid carriers is attained exothermically during hydrogenation process whereas endothermic liberation of hydrogen is observed during dehydrogenation process. Moreover, this endothermicity is a major drawback due to the requirement of high heat demand. Though thermodynamic evaluation is the one of the concerns in designing the LOHCs, an active catalytic system with a high to moderate loading of precious metal catalysts (e.g., Pd, Pt, Rh, and Ru) can actually achieve acceptable dehydrogenation kinetics at low temperatures (150°C). In particular, numerous homogenous catalysts have been developed for this purpose; however, the achieved stability, recycling, and practical advantages in large-scale applications have more precisely promoted heterogeneous catalyst systems. Various commercial heterogeneous catalysts with different supporting substances (e.g., Pd/C, Pt/Al_2O_3, and $Pd\text{-}Pt/Al_2O_3$) have attracted much attention for LOHC technology. However, high catalyst efficiency is the major criteria in which metal loading, appropriate support selection, and structural properties (particle size, porosity, active surface area, etc.) are all important characteristics. To this end, development and optimization of efficient catalyst systems obviously requires both experimental and theoretical approaches. Theoretical studies using DFT calculations can provide fundamental insight into catalytic activity and selectivity over different metal surfaces and experimental methods comprised of various spectroscopic techniques (typically X-ray photoelectron spectroscopy (XPS), temperature-programmed desorption (TPD), and infrared reflection absorption spectroscopy (IRAS), etc.) further support the field of LOHC dehydrogenation catalysis. Combining these complementary approaches, the development of efficient catalytic systems is promising [28].

4. Physical and chemical method of hydrogen storage

There is no linear connection of the volumetric density of hydrogen and pressure. The energy needed for hydrogen gas compression, coupled with its low density, poses challenges for efficient hydrogen transport. The liquefaction of hydrogen can enhance the density. The density of liquid hydrogen is 70.8 kg/m^3 at 21 K. There is a loss of hydrogen due to the transfer of the heat through containers as the critical temperature of hydrogen is 33 K. Additionally, the cooling system adds extra weight and also utilizes energy. Thus, the system should be equipped with strong thermal insulators to decrease the loss of hydrogen through vaporization. The theoretical density of hydrogen to be stored should be high for efficient storage capacity. The liquefaction of hydrogen gas is not feasible as it uses one-third of the total amount of energy stored in hydrogen. Another route, for the physical storage of hydrogen, is by adsorption into porous materials with high surface areas like activated carbons, a covalent organic framework, or a metal–organic framework. However, the capacity of hydrogen storage is less with these materials [27, 29].

To overcome the limitation of the physical storage of hydrogen, the chemical storage of hydrogen has evolved to make the transportation of stored hydrogen economically feasible on a large scale. The densities of hydrogen in chemical storage are enhanced to 170 kg H_2/m^3, as hydrogen forms covalent bonds which are shorter. The release and storage of hydrogen from the organic liquids are carried out through the dehydrogenation and hydrogenation process. The liquefaction and compression processes are eliminated, since hydrogen molecules are stored by the formation of covalent bonds. Hydrogenated LOHC is dehydrogenated catalytically on-board, on the vehicles for releasing the stored hydrogen [29]. The used (dehydrogenated) fuel is preserved in a storage tank which is later re-generated off-board by hydrogenation. The main criteria of LOHC are that it must possess a low melting point and a high boiling point, such that it retains its liquid phase at a low temperature. Additionally, LOHC compounds should not decompose and should undergo dehydrogenation and hydrogenation selectively. However, there are challenges to meet the requirements of the storage capacity, as the temperature required for the dehydrogenation reaction is very high and inconvenient during transportation. This step has been improved by the introduction of a heteroatom into the aromatic ring. The presence of nitrogen in five- or six-membered heterocycles reduces the heat of hydrogenation, enhancing this crucial step [16] [27-29].

4.1 Thermodynamics of hydrogen storage

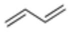
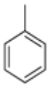
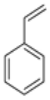
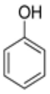
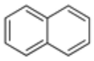
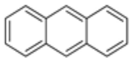
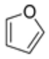
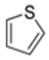
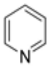
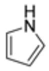
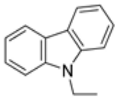
The probability of hydrogenation and dehydrogenation of an organic molecule to be used as a hydrogen carrier is dictated by the thermodynamics of the reaction. Notably, the hydrogenation process is thermodynamically favourable, while the process of dehydrogenation is difficult. This is because both the processes possess the same free energy of reaction with different algebraic signs. The free energy of the reaction is given by the following Equation (1).

$$\Delta G = \Delta H + T\Delta S \quad (1)$$

where, ΔG is free energy of the reaction, ΔH is enthalpy of the reaction, ΔS is entropy of the reaction, and T represents the temperature.

Although the hydrogenation reaction is not favoured by entropy, it is driven by the equilibrium of the reaction. The free energy of the reaction [36 KJ (mol H_2)$^{-1}$] exceeds that of the enthalpy of the reaction. The hydrogenation of unsaturated compounds is an exothermic reaction and hence is the thermodynamically propelled at room temperature, although not feasible due to entropy. Elevating the temperature shifts the equilibrium toward dehydrogenation. Thus, the organic compounds having an enthalpy of reaction lower or equal to −40 KJ (mol H_2)$^{-1}$ should be selected for LOHC, as this will make the process of dehydrogenation feasible. A higher enthalpy of reaction requires the temperature to be lower than 20^{o}C to pass the thermodynamic barrier. Thus, the kinetics of the reaction will be sluggish. Conversely, the temperature required for the dehydrogenation reaction will be very high, if the enthalpy of the reaction is lower than the desired value of −40 KJ (mol H_2)$^{-1}$.The thermodynamic properties of some organic compounds as potential LOHCs are shown in Table 1.

Table1: Thermodynamic properties of LOHCs.

LOHC	Free Energy of Reaction	Enthalpy of Reaction
	−100.4	−136.3
	−83.3	−118.4
	−32.6	−68.7
	−31.6	−68.3
	−43.6	−79.7
	−25.6	−63.3
	−20.5	−66.3
	−19.6	−66.9
	−40.3	−74.7
	−40.4	−74.6
	−29.5	−63.1
	−22.8	−55.9
	−18.2	−53.2

Organic compounds with isolated double bonds can be easily hydrogenated, while their dehydrogenation is very tough [26, 30]. The hydrogenation of ethene is thermodynamically feasible to form ethane, while its dehydrogenation is not viable. Compounds containing conjugated double bonds are a better choice as LOHC, but with a feeble force for initiating the dehydrogenation reaction. The dehydrogenation process is viable only to form aromatic compounds since they are highly stable. The dehydrogenation to form double bonds outside the aromatic ring, although conjugated, requires high energy. The thermodynamic properties of a ring are altered by the introduction of a heteroatom. Nitrogen positively enhances dehydrogenation, while sulphur and oxygen have negative impacts. The presence of boron along with snitrogen propels the dehydrogenation reaction further. The temperature can be further decreased by diminishing the partial pressure of the system or by the addition of inert gas. Nevertheless, the quality of hydrogen is affected by these methods [31].

5. Reactors used for dehydrogenation

The dehydrogenation process of hydrogenated LOHCs is complex, as the reaction heat should be effectively transported to the activation site of the catalysts. Additionally, a significant amount of hydrogen gas is evolved within the reactor. A reactor was fabricated, using selective electron beam melting (SEBM) instead of laser beams, in which a metal powder in the bed was melted locally using an electron beam for dehydrogenating LOHCs [32]. The reactor was layered with the simultaneous fabrication of the inner 3D structure and its walls. The electron beam was based on electromagnetic fields and a high speed was obtained, appropriate for Ti-6Al-4V powders.

The rate of the reaction was high at an elevated temperature which is attributed to a high rate constant of the reaction under high-temperature conditions, and a fasterpropelling force to carry out the endothermic dehydrogenation reversible reaction. The initial dehydrogenation from H12-NEC to H8-NEC is the most feasible step, while further dehydrogenation steps are slothful. An enhancement in the rate of flow of LOHC decreases the generation of hydrogen as the residence time declines. The stability of the system is also very high.

6 Catalysts used for dehydrogenation

The dehydrogenation of 12H-NEC is not feasible at atmospheric pressure, which has led to the development of different catalysts. Homogeneous catalysts, prepared from PCP pincer iridium complexes, failed to produce complete dehydrogenated products even after a prolonged reaction time [33]. A comparative study revealed that Pd supported on alumina was found to be effective as compared to Pt, Rh, and Ru [17]. The hydrogenation and dehydrogenation process are shown in Figure 1.

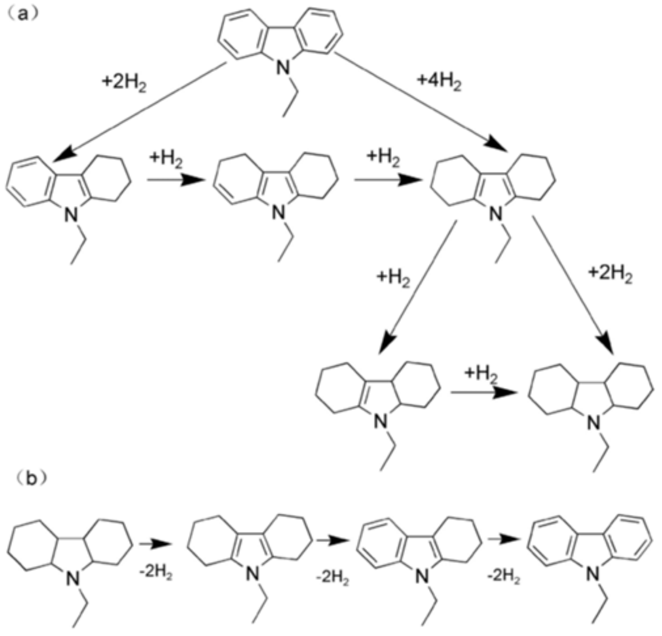


Figure 1: The hydrogenation and dehydrogenation process.

It was found from a density-functional theory (DFT) analysis that the catalytic activity of Pd (111) for adsorption and dissociation is better as compared to Pd (100) and Pd (110) due to greater facet proportion [33]. The dehydrogenation process followed three stages as shown in Equations (1)–(3). The initial step is the removal of 2 moles of hydrogen from the five-membered ring of 12H-NEC to form 8H-NEC, followed by the removal of another 2 moles from one of the six-membered ring producing 4H-NEC which is a stable intermediate. The last stage is the elimination of hydrogen sluggishly to form NEC.

$$12H\text{−}NEC \rightarrow 8H\text{−}NEC + 2H_2 \quad (1)$$

$$8H\text{−}NEC \rightarrow 4H\text{−}NEC + 2H_2 \quad (2)$$

$$4H\text{−}NEC \rightarrow NEC + 2H_2 \quad (3)$$

In onereport, hydrogen (600 mL) was obtained through the complete dehydrogenation of liquid H12-NEC (1 mL) and NEC (0.85 mL) upon reaching the ambient temperature. The high gas flow rate influences the mass transfer technique in catalyst pores. Continuous reactors are preferred to study and estimate the kinetics of a reaction, after reaching a steady-state condition, after a constant time-period of reaction. A study of the kinetics was conducted by substituting powder catalysts with catalyst pellets to prevent the fluctuations, the formation of bubbles, and pulsation associated with powder catalysts. The rate of diffusion depends on the particle size of catalysts which can be made constant by applying catalyst coating layers with different thicknesses. Coating methods include slurry coating, sol-gel coating, and a hybrid combination of both techniques. Different coating methods are: brush coating and spin coating for flat substrates; drop coating and dip coating for microreactors; as well as spray and electrostatic sol spray coating on various structures. In another study, the dehydrogenation of LOHCs was studied with Pd in combination with different metals (Ru, Pt, Cr, W, and Ge) supported on TiO_2, of which W, Cr, and Ge show no activity [33]. Pd-Ru/TiO_2 and Pd-Pt/TiO_2 were effective at 195ºC after 6 h. The dehydrogenation was also carried out under microwave at 10 W with complete conversion in 100 min. Intermediates formed under microwave were 10H-NEC, 6H-NEC, and 2H-NEC. Additionally, the formation of side products like methane and ethane was suppressed. Microwaves are absorbed by the metal centres instead of the matrix composed of TiO_2. A polarized monoatomic layer is created by the dissociation of hydrogen at the surface of metal nanoparticles. Thus, the selectivity of this hydrogenation process is improved because the metal centres are solely activated. The dehydrogenation of 12H-NEC was studied by using a 0.5 weight % of Pd on mesoporous MoO_3, where the dispersed Pd particles offer active sites for the dissociation of H-H bonds and the movement of protons on the mesoporous surface [34]. The intermediate strength of binding between the molecules and the active centres on the surface of catalysts dictates the activity of the catalysts. A weak strength indicated that the molecules were less activated and the reaction was slower. In contrast, a strong intermediate strength indicated that the products failed to be desorbed from the catalytic active sites, thus blocking the requisite sites for the reaction. This intermediate strength was found to be optimal for Pt and Pd.

A dehydrogenation catalyst was synthesized by the homogeneous dispersion of nanoparticles of Pd-Cu on the surface of reduced graphene oxide, where Pd1.2Cu/reduced graphene oxide was found to be effective for complete conversion to NEC within a reaction time of 7 h at 453 K . The observed reaction rate was similar to that of Pd/reduced graphene oxide. However, as the amount of Pd was reduced in this study, the cost of the catalyst was substantially decreased.

The activity of the catalysts was higher with a smaller size. An enhancement in the amount of Cu to 50% does not affect the binding energy of Pd-3d. However, a higher amount of Cu (more than 50%) initiates the transfer of electronsfrom Pd to Cu, resulting in a decreased dehydrogenation rate. The electron distribution in the outer shell of Pd is also affected by the ratio of the Pd and Cu.

8. Conclusions

As the demand for clean and sustainable energy grows, hydrogen has proven to be a promising alternative. However, its storage and transportation remain significant challenges. This research highlights Liquid Organic Hydrogen Carriers (LOHCs) as a potential solution, offering a safer and more practical way to store and transport hydrogen.

Our study explored various LOHC candidates, their performance, and the role of catalysts in improving hydrogen release efficiency. While molecules like dibenzyl toluene and toluene show great promise, further advancements are needed to optimize reaction conditions, reduce energy costs, and ensure environmental safety. Future research should focus on developing better catalysts, improving the stability of LOHCs, and conducting life cycle and economic analyses to make this technology more viable for large-scale implementation.

The LOHC molecules have been discussed based on their varied characteristics, such as stability, storage capability, catalysts used, and reusability. The technology associated with LOHCs demands huge development. The stability of LOHCs can be enhanced by lowering the enthalpy of dehydrogenation of these molecules. Hence, studies encompassing thermodynamic tuning of both the hydrogenation and dehydrogenation processes are required. The use of catalysts lowers the activation energy of dehydrogenation of LOHC molecules. This results in the requirement of a low dehydrogenation temperature. Future research should be directed toward investigating efficient catalysts for the dehydrogenation of LOHC molecules at low temperatures and make the system economically viable.

The asset of LOHCs is that they resemble crude oil; hence, the existing systems can be used for the generation of energy. Therefore, dibenzyl toluene and toluene as LOHC candi-dates exhibit high prospects for mass commercial production. The application of LOHC molecules as energy sources should be further explored. The reactors for dehydrogenation can be improved for making the reaction conditions more feasible.

The toxicity of LOHC molecules should be minimal to conform with environmental concerns. Since these LOHC molecules are well-studied, it is easier to handle the risks of these molecules as compared to crude oil. LOHC should always be computationally designed considering the eco-toxicological profiles like the mutagenic, carcinogenic, and toxic profiles. However, these parameters are challenging to predict by the quantitative structure-activity relationship (QSAR). There has been a dearth of computer modelling studies, including biodegradability, efficacy, and toxicity, for the search of apposite catalysts for the dehydrogenation of LOHC molecules. Additionally, life cycle analysis and economic analysis of the LOHC technology should be studied to make the system sustainable. LOHC systems require transportation, hydrogenation, and dehydrogenation.

Authors' contributions

The author read and approved the final manuscript.

Conflicts of interest

The author declares no conflict of interest.

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Data availability

No new data were created.

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