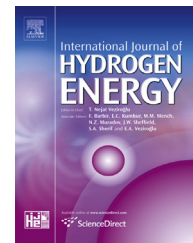




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# Efficiency analysis of novel Liquid Organic Hydrogen Carrier technology and comparison with high pressure storage pathway

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## ABSTRACT

Liquid Organic Hydrogen Carrier (LOHC) is under fundamental research in recent years as a potential hydrogen storage pathway for fuel cell vehicles. This report focuses on the energy efficiency of the LOHC supply system from centralized off-site hydrogenation to vehicle on-board dehydrogenation. In this paper, a system model using LOHC-based hydrogen storage/transport/supply is established, the energy flow is analyzed by calculating the energy consumptions in the steps of hydrogenation process, LOHC transport and dehydrogenation process. Based on physical properties and some experimental data, the energy efficiency and its variations along with the decrease of dehydrogenation capability and energy recycling are also simulated and compared with other commercialized hydrogen storage methods (high pressure or liquefied). It is found that the energy efficiency of the LOHC has a highest number of 69.17% and 88.74% with and without energy recycling respectively, and will reduce with the dehydrogenation capability decrease. The results also show that, the energy efficiency of LOHC technology is somewhat lower than those of 70 MPa and 35 MPa high-pressure storage methods. Compared with liquefied pathways, the LOHC efficiency is higher with energy recycling and a little lower without energy recycling.

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## Introduction

Fuel cell electric vehicles, using pollution-free hydrogen as its fuel load a promising future. A series study results show that using hydrogen results in an obvious decrease in carbon monoxide, nitrogen oxides and sulfur dioxide emissions [1–8]. The entire process of hydrogen utilization in on-board fuel cells includes hydrogen production and purification, hydrogen transportation and distribution, hydrogen fueling and finally the on-board redox reaction that provides energy to power the vehicles.

However, there are rather substantial obstacles holding back the promotion of fuel cell electric vehicles (FCEV). The

first is the life span and cost of the fuel cells. Finding proper materials for the core components such as the Proton Exchange Membrane (PEM), bipolar electrode and catalyst is the key to removing the commercialization barriers to fuel cells [9–17]. Moreover, there is another obstacle to the application of FCEVs, as well as the use of hydrogen in various fields, and that is hydrogen storage. Currently, only a few ways dominate hydrogen storage, namely high-pressure gaseous hydrogen storage and liquefied hydrogen storage. Several approaches are still under development. These approaches include H<sub>2</sub> storage in Liquid Organic Hydrogen Carriers (LOHC), H<sub>2</sub> storage in metal hydrides, H<sub>2</sub> storage in sorbents, chemical hydrogen storage, etc.

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Comprehensive studies of these different approaches have been published. Ahluwalia, for instance, evaluated several on-board H<sub>2</sub> storage options for their potential to meet the near-term, intermediate and ultimate targets for light-duty vehicles [18]. In 2014, Zhou and others reviewed the research progress of the three main chemical hydrogen storage technologies: metal hydrides, complex hydrides and organic liquid hydrides [19].

Among the storage technologies mentioned above, which are mainly in the experimental stage, hydrogen storage based on Liquid Organic Hydrogen Carriers has gained worldwide attention in recent years [20–32]. Previous research has contributed greatly to this field of study, most of which laid the emphasis on the reaction mechanism between hydrogen and the carriers, the catalyst and the reaction conditions. For instance, researchers have examined the quality of different catalysts and the corresponding reaction conditions. In terms of the reaction mechanism, Sotoodeh notes the different levels of difficulty for hydrogenation and dehydrogenation [33]. However, Manas and his co-researchers report that two types of catalysts are suitable for the hydrogenation and dehydrogenation processes and that they do not require high temperatures, high pressures of H<sub>2</sub> or a strong base for the hydrogenation step [34]. Choi has analyzed thermodynamic efficiencies of hydrogenation and dehydrogenation reactions assuming an idealized process which also developed a framework useful for screening candidate LOHC molecules [20].

In a paper published in 2011, Alhumaidan provided a review of the effort devoted to the development of LOHC use for hydrogen storage over the preceding few decades and indicated that the main technical limitations are related to the amount of energy required to extract hydrogen from the liquid organic hydride and the insufficient stability of the dehydrogenation catalyst [35]. Similarly in 2012, Jiang described the advantages and disadvantages of different hydrogen storage methods and highlighted the current research on the LOHC process [36]. Additionally, from a more systematic view, Markiewicz gave an environmental and health-impact evaluation of LOHC and offered recommendations for future assessments to improve both academic accuracy and public acceptance of LOHC [25]. Teichmann published a paper assessing the feasibility of LOHC for the transport and storage of renewable energy from the perspective of costs and energy efficiency [37]. Interestingly, the same writer also discussed the application of LOHC to store energy in residential and commercial buildings [38].

However, as mentioned above, most of the previous research focused on the mechanism of the reaction between hydrogen and the carriers, the catalyst and the reaction conditions. That is to say, they are mainly laboratory-based experiments, rather than an examination of the total LOHC on-board application. Differing from the previous theoretical research, this paper takes a systematical approach to analyze the energy efficiency of hydrogen storage by LOHC, starting with the hydrogenation, and ending with the hydrogen going into the on-board fuel cells. Next, this new hydrogen storage technology is compared to the commercialized high-pressure storage methods, focusing on aspects of energy efficiency.

## LOHC process: hydrogenation and dehydrogenation

Hydrogen storage using LOHC creates a certain environment with a catalyst causing gaseous hydrogen to react with the LOHC, generating a type of liquid organic hydride. The stability of the hydride at room temperature allows its direct transportation and distribution by trucks. Dehydrogenation reverses the hydrogenation reaction, in a process that is endothermic, under slightly different conditions. Theoretically, all of the hydrogen reacting with the carriers at the former stage would be released, and the carriers would be recycled for reuse. Then, the hydrogen could be utilized in different ways such as powering on-board fuel cells.

### Basic features of LOHC

Currently, there are several carriers that show promise for future use. Attention has been given to benzyltoluene and dibenzyltoluene [39], amine borane-based compounds [40], methylcyclohexane [35] and others. Most recently, Li reports a new promising carrier-2-methylindole which has a hydrogen storage capacity of 5.76 wt% and relatively moderate reaction circumstances [41]. The basic physical properties of some of these carriers are shown in Table 1.

Following the research conducted by Pez and others on ethylcarbazole as a possible hydrogen carrier, published in 2006, this specific carrier has gained increased attention [42,43]. Ethylcarbazole is an intermediate in the synthesis reactions of Sulphur Vat Blue GNX and Permanent Violet RL. Its melting and boiling points are 341.15 K and 463.15 K, respectively [36]. Fig. 1 shows the ethylcarbazole hydrogenation and dehydrogenation processes. Intermediates with different degrees of hydrogenation are formed during these reactions. The percentage of the different intermediates during the (de)hydrogenation process depends on both the degree of reaction and the properties of the catalyst [44].

In terms of its life cycle, the current research focuses on the mechanism of the dehydrogenation process showing that the decay of the carrying capacity, leading to a short life span of ethylcarbazole and a reduction of hydrogen generated in the dehydrogenation reaction. In the research conducted by four scholars in Zhejiang University, they reported that the catalytic activity of the dehydrogenation reaction would decrease

**Table 1 – Physical properties of some Liquid Organic Hydrogen Carriers [37].**

Carrier	Melting point, K	Boiling point, K	Mass density, %
Cyclohexane	279.65	353.85	7.19
Methylcyclohexane	146.55	374.15	6.18
Tetralin	237.35	480.15	3.0
Cis-Decaline	230.15	466.15	7.29
Trans-Decaline	242.75	458.15	7.29
Phenylcyclohexane	278.15	510.15	3.8
4-Aminopiperidine	433.15	338.15	5.9
Carbazole	517.95	628.15	6.7
Ethylcarbazole	341.15	463.15	5.8

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