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Assessment of system variations for hydrogen transport by liquid organic hydrogen carriers

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ABSTRACT

One option to transport hydrogen over longer distances in the future is via Liquid Organic Hydrogen Carriers (LOHC). They can store 6.2 wt% hydrogen by hydrogenation. The most promising LOHCs are toluene and dibenzyltoluene. However, for the dehydrogenation of the LOHCs – to release the hydrogen again – temperatures above 300 °C are needed, leading to a high energy demand. Therefore, a Life Cycle Assessment (LCA) and Life Cycle Costing are conducted. Both assessments concentrate on the whole life cycle rather than just direct emissions and investments. In total five different systems are analysed with the major comparison between conventional transport of hydrogen in a liquefied state of matter and LOHCs. Variations include electricity supply for liquefaction, heat supply for dehydrogenation and the actual LOHC compound. The results show that from an economic point of view transport via LOHCs is favourable while from an environmental point of view transport of liquid hydrogen is favourable.

1. Introduction

One option to tackle problems in the mobility sector, such as greenhouse gas emissions, local exhaust gas emissions or oil dependency [1–3] is the usage of hydrogen in fuel cell electric vehicles. No exhaust gas emissions occur, depending on the hydrogen source the greenhouse gas emissions can be decreased significantly if renewable energy sources are used [4] and sources of primary energy can be diversified. A comprehensive implementation of hydrogen as a fuel, however, demands a new transmission and distribution infrastructure for hydrogen. For short distances, in this study around 100 km, transport of pressurised hydrogen is from an environmental as well as economic point of view reasonable [5]. For longer transport distances (300 km and longer) the construction of a new hydrogen pipeline system is the most economical solution [6] in the long run. The initial investment, however, is very high [7] and probably only an option for strategic long term decisions. At the moment liquefaction of hydrogen is common for long distance transport of hydrogen. It requires, though, a great amount of electricity for liquefaction, roughly 24–36 % of its energy content [8]. A new option to transport hydrogen over longer distances are liquid organic hydrogen carriers (LOHCs) [9]. They can store 6.2 wt% hydrogen by hydrogenation [10]. The most promising LOHCs are toluene and dibenzyltoluene [11]. However, for the dehydrogenation of the LOHCs – to release the hydrogen again – temperatures above 300 °C are needed, leading to a high energy demand.

1.1. Literature review

Already in 1999 Scherer, Newson [12] analysed economically LOHCs as a means of electricity storage for fluctuating renewable energies. They come to the conclusion that for storing the electricity in the form of hydrogen efficiencies between 40 and

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25 % can be achieved depending on the technology of re-electrification. Compared to fossil fuels, however, the costs are ten times higher. A similar assessment performed Eypasch, Schimpe [13] more recently, where a cost optimization of an industrial production facility supplied with renewable energy sources is performed. Again LOHC and hydrogen are used as a storage medium for electricity. An electricity system that is 60 % self-sufficient (cost optimum) is found to have 33 % lower cost than supply with electricity exclusively from the grid, while a 100 % self-sufficient system has 47 % higher cost. Several papers also discussed LOHCs not only as a storage medium but also as a transport medium for hydrogen. In one study the long distance transport, e.g. from Iceland or Northern Africa, of hydrogen is analysed. It is concluded that transport in LOHCs is cheaper than transport as liquid hydrogen and on the same cost level as domestic hydrogen supply with renewable power methane [14]. Reuβ, Grube [6] compared the transport of hydrogen via LOHCs with other options like liquefied hydrogen transport, pipeline transport and transport of pressurised hydrogen in trucks. They also come to the conclusion that transport as liquefied hydrogen is more expensive than LOCHs. However, depending on the transport distance and the daily hydrogen demand hydrogen transport as compressed gas in trucks or pipelines is more cost efficient. Next to these techno-economic assessments Markiewicz, Zhang [15] performed an assessment of environmental and health impacts on the LOHC compounds by a hazard assessment. After the assessment they stress out that also many mineral oil based compounds are toxic and that even diesel a mass product used every day has a long list of safety issues. As LOHC compounds are very diverse a general statement regarding environmental and health impacts is not yet possible. Further testing should be used to optimize selected LOHC compounds.

Life Cycle Assessment (LCA) is an established tool for assessing hydrogen systems cf. [4,16–18]. Even review articles regarding LCA of hydrogen production have been published [19,20]. A comprehensive LCA for hydrogen transport in LOHCs, which not only considers the direct emissions of a process but also pre-processes, has not been performed yet. Therefore, in this paper an LCA for the supply of hydrogen by transport in LOHC is conducted flanked by the calculation of life cycle cost.

1.2. Analysed options

How future long distance transport of hydrogen will develop is not clear yet. Therefore, different system variations are analysed. A new technology of transporting hydrogen in LOHCs is compared to conventional transport of liquid hydrogen in this paper. In both supply chains some hydrogen is lost, e.g. through boil-off, although in different amounts. Therefore, it is important to include the hydrogen production in this assessment. Alkaline water electrolysis powered by wind electricity is assumed here. The final usage of hydrogen in fuel cell electric vehicles is not part of the assessment, because it is a very complex topic on its own. In Fig. 1 the different supply chains considered are visualised.

The first system variation concerns the chemical compound used as LOHC. Different compounds have been identified to be suitable for hydrogen transport [11,21]. In Germany dibenzyltoluene is favoured due to its low toxicity and its high boiling point [10]. In Japan, on the other hand, toluene is favoured because of its lower price. In this paper both compounds are assessed. The dehydrogenation of LOHCs, toluene as well as dibenzyltoluene, requires temperatures above 300 °C. The conventional technology to provide this heat is the burning of natural gas as it is widely available and cheap. Another possibility to provide this heat is by burning part of the transported hydrogen. This option is analysed here for dibenzyltoluene as a system variation. For the liquefaction one of the important assumptions is the choice of the electricity mix. Conventionally the grid mix for the reference year and country is chosen. As the liquefaction is probably situated next to the electrolyser, which is run by wind power, it is justified to assume wind power as electricity source for the liquefaction also. Summarizing these variations in total five different supply chains are assessed:

1. Transport of liquid hydrogen, liquefaction with electricity grid mix (LH2 grid),
2. Transport of liquid hydrogen, liquefaction with electricity from wind power (LH2 wind),
3. Transport of hydrogen in dibenzyltoluene as LOHC, heat production with natural gas (LOHC DBT),
4. Transport of hydrogen in toluene as LOHC, heat production with natural gas (LOHC Tol),
5. Transport of hydrogen in dibenzyltoluene as LOHC, heat production using hydrogen (LOHC own).

1.3. Technological background

Here a short introduction into the technologies of hydrogen transport in a liquefied form and by LOHCs is given.

1.3.1. Liquid organic hydrogen carrier

LOHCs are aromatic carbohydrates that bind hydrogen in a catalytic reaction. Already investigated pairs of the hydrogen lean (pure LOHC) and hydrogen rich (LOHC + H_2) form are N-ethylcarbazole/perhydro *N*-ethylcarbazole, toluene/methylcylohexane and dibenzyltoluene/perhydro dibenzyltoluene. The hydrogenation is an exothermic reaction where heat at a temperature level of 150 °C is released. Correspondingly the dehydrogenation is endothermic and temperatures between 270 and 350 °C are required depending on the used chemical compound [11,21]. The dehydrogenation is depicted in equation (1) on the example of toluene/methylcyclohexane.

$$C_7H_{14} \xrightarrow{\text{Heat}} C_7H_8 + 3H_2 \quad (1)$$

For hydrogenation as well as dehydrogenation catalysts, e.g. Pt/C, are required. Depending on the applied LOHC compound the storage capacities vary between 5.8 and 7.3 wt% [11,22]. During transport and distribution the hydrogen rich and lean LOHC can be handled like diesel under ambient conditions. That makes the handling much simpler than for cryogenic or high compressed hydrogen [14].

1.3.2. Liquefaction of hydrogen

The liquefaction is a standard pretreatment for transporting hydrogen. It has the advantage that liquefied hydrogen at

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