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Techno-economic feasibility of road transport of hydrogen using liquid organic hydrogen carriers

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HIGHLIGHTS

- Techno-economic model for point-to-point large scale road transport of hydrogen.
- The LOHC concept can decrease long-distance delivery costs significantly.
- Heat supply method for dehydrogenation and heat integration are key.
- Utilization of waste heat can reduce costs by 40%.

GRAPHICAL ABSTRACT

ABSTRACT

The cost of storing and transporting hydrogen have been one of the main challenges for the realization of the hydrogen economy. Liquid organic hydrogen carriers (LOHC) are a promising novel solution to tackle these challenges. In this paper we compare the LOHC concept to compressed gas truck delivery and on-site production of hydrogen via water electrolysis. As a case study we consider transportation of by-product hydrogen from chlor-alkali and chlorate plants to a single industrial customer, which was considered to have the greatest potential for the LOHC technology to enter the markets. The results show that the LOHC delivery chain could significantly improve the economics of long distance road transport. For economic feasibility, the most critical parameters identified are the heat supply method for releasing hydrogen at the end-user site and the investment costs for LOHC reactors.

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Introduction

Preventing disastrous climate change requires transformation from the current fossil carbon dependent system to a renewable based one. Hydrogen will play a significant role in the transformation as decarbonisation of several industrial sectors – such as chemical, steel and transportation sectors – will depend on the availability renewable and low-carbon hydrogen. Currently around 70 million tonnes [1] of dedicated hydrogen is produced annually and the hydrogen demand could increase almost tenfold by 2050 [2].

Due to the higher cost of renewable production options, hydrogen is today supplied almost entirely from fossil feedstocks causing annual CO₂ emissions of 830 Mt [1]. However, a significant amount of hydrogen is also formed as an unavoidable by-product of industrial processes, which could be an alternative source for low-carbon hydrogen. A notable portion of this by-product hydrogen is currently just vented into atmosphere or used in relative low value on-site applications, such as heat production, mainly due to the lack of cost-efficient hydrogen logistic methods.

A particularly interesting source of by-product hydrogen are the electrolytic processes in which a high concentration stream of relatively high purity hydrogen is formed, namely chlor-alkali and sodium chloride processes. In chlor-alkali plants, caustic soda (sodium hydroxide, NaOH) and chlorine (Cl₂) are produced via electrolysis of aqueous solution of sodium chloride (NaCl) according to

\[
2 \text{NaCl (aq)} + 2 \text{H}_2\text{O (l)} \rightarrow 2 \text{NaOH (aq)} + \text{Cl}_2 (g) + \text{H}_2 (g) \quad (1)
\]

Theoretically, for each tonne of caustic soda produced, 25 kg of hydrogen forms as a by-product. Hydrogen can be used to produce hydrochloric acid (HCl) or used as fuel to provide the needed energy for the process. The annual production of caustic soda was ~72 Mt in 2016 [3]. This corresponds to ~1.8 Mt of hydrogen and it has been estimated [4] that around 15% of this is currently vented.

Sodium chlorate (NaClO₃) is also produced via sodium chloride electrolysis but the reaction proceeds according to

\[
\text{NaCl (aq)} + 3\text{H}_2\text{O (l)} \rightarrow \text{NaClO}_3 (aq) + 3\text{H}_2 (g) \quad (2)
\]

Theoretically, 56 kg of hydrogen is then generated for each tonne of sodium chlorate. Sodium chlorate is mostly used for making bleaching chemicals in the pulp and paper industry. The sodium chlorate market was 4.2 Mt/a in 2019 [5], which corresponds to 0.24 Mt/a of by-product hydrogen. Despite the one magnitude smaller global market, in some countries such as Finland, chlorate production is the dominant source of electrolytic by-product hydrogen because of the extensive pulp industry.

The total amount of by-product hydrogen from chlorate and chlor-alkali industries is thus ~2 Mt/a, which corresponds to 67 TWh/a (240 PJ/a) based on the lower heating value (LHV). This is around 3% of the global dedicated hydrogen production. In addition to effective utilization of by-product hydrogen the need to transport renewable hydrogen, for example to hydrogen refuelling stations, will only increase.

Hydrogen supply chains (HSC) have been modelled in numerous studies and reviews of them are available [6–8].

In practice, the main current logistic option for by-product hydrogen has been truck delivery in the form of compressed gas. Transporting compressed hydrogen by truck is considered the most suitable for delivering relatively small amounts of hydrogen over short or moderate distances (<200–300 km) [8]. Hydrogen has traditionally been transported in 200 bar steel bottle containers, which have limited hydrogen payloads (200–300 kg) due to high weight of the bottles and the low maximum pressure. More advanced options for compressed gas delivery are also available today. Glassfibre composite cylinders (up to 350 bar) and carbon fibre cylinders up to 500 bar are already commercially available, and these have been used in the latest analyses of hydrogen transportation cost [5,9–11]. Hydrogen payloads even exceeding 1000 kg are achievable with carbon fibre cylinders [9], even with 44-tonne trucks, but have the drawback of higher investment costs. For vehicle on-board applications, 700 bar has been considered the most feasible pressure [12].

Hydrogen could also be transported via pipelines, or it could be liquefied and delivered by cryogenic tanker trucks [6,13]. However, pipelines are cost effective only for large volumes or short distances, making them rarely an option for maximizing the value of by-product hydrogen. Liquefaction would enable hydrogen to be trucked more efficiently over long distances due to significantly higher payloads (4000–4500 kg) [6]. However, the hydrogen liquefaction process is very capital intensive and has high energy requirement [6,14]. In general, liquefaction is cost- and energy efficient only for very large-scale plants (>100 MWₜₜ.). Furthermore, transportation and storage of liquid hydrogen leads to boil-off losses.

The liquid organic hydrogen carrier (LOHC) concept is a promising novel solution for more efficient and safe storage and transportation of hydrogen. LOHCs are liquids that can be reversibly hydrogenated and dehydrogenated. Hydrogen storage densities of different LOHCs are typically in the range of 5–7 wt% [15] and 40 tonne tanker trucks could then carry around 1500–2000 kg of hydrogen [6,13].

Techno-economic studies for using LOHC for transporting hydrogen have already been performed. Several of these papers have considered LOHC as one possible energy carrier for the intercontinental energy trade [13,16–18]. Teichmann et al. [13] studied options for transporting renewable energy from Africa or Iceland to Europe and found that the LOHC concept based on dibenzyl toluene (DBT) could be a feasible option. Analysis by Niemann et al. [16] showed that methanol was the most cost effective option followed by dibenzyl toluene and toluene. Wijayanta et al. [17] compared ammonia, liquid hydrogen and LOHC for energy import from Australia to Japan and found that liquid hydrogen would be the most feasible option in 2050 when pure hydrogen is required. For the LOHC concept the main identified drawback was the high energy demand for dehydrogination. Hank et al. [18] found that liquid hydrogen would be the most energy efficient option to transport energy from Africa to Germany. Also, the costs where roughly on par with ammonia, which was the lowest cost option. The delivered costs of energy carriers ranged from 124 to 156 €/MWh with LOHC (DBT) having the highest cost. Reuß
et al. [6] did not consider intercontinental transportation but developed a flexible country-wide hydrogen supply model for mobility which extended the previous models with seasonal storage and LOHCs as a novel hydrogen carrier. The LOHC technology was found to be very promising for future hydrogen supply chains. Pradhan et al. concluded in their study [19] that LOHC technology has immense techno-economic potential in transporting hydrogen for mobility applications in India. The applicability of the LOHC concept for other purposes such as seasonal large-scale energy storage [20], energy storage in residential and commercial buildings [21] or industrial plant [22] or use as a transportation fuel [23] have also been considered. Also, coupling of endothermic dehydrogenation with industrial waste heat from a cement plant has been studied [24].

In addition to the techno-economic considerations, also the environmental aspects of hydrogen transport using the LOHC technology have been studied [25,26]. The results from Ref. [25] show that, for the European conditions, the use of LOHC technology is environmentally superior to gaseous compressed hydrogen for distances above 365 km. However, in Ref. [26] compressed gas was found to be more feasible in most of the environmental impact categories even with a 400 km transport distance. The assumed compressed gas tube trailer capacities differed markedly between these studies (300 kg in the former and 1100 kg in latter), which might be the main factor for the different outcomes. Also, the considered LOHC compounds were different as well as the required end-use pressure and the CO₂ intensity of the electricity. Both studies assumed that dehydrogenation is carried out using natural gas as the heat source.

The previous HSC analyses have mostly focused on the role of the LOHC concept as a part of the mid-term or long-term future energy systems that will have a high amount of intermittent electricity production or import of renewable hydrogen on a large-scale. However, for rapid commercialization of the LOHC technology the most attractive near-term markets should be analysed in detail, such as large-scale road transport of hydrogen between a specific industrial producer and a consumer.

In this study we analyze the competitiveness of the LOHC supply chain in road transport of by-product hydrogen from chlor-alkali/chlorate plants to a single industrial user in today’s market. This case was considered the most promising option for the LOHC technology to enter the markets due to the current low-value use of by-product hydrogen, suitable scale and heat integration benefits. This case is also close to a real-life case in Finland.

The LOHC chain is compared to delivery as compressed gas as it is the only near-term alternative in the considered scale. The analysis is limited to road transport, but extending to rail transport would be relatively straightforward. In the feasibility analysis the total costs of the delivery chains are calculated. Delivery costs are also compared to on-site production of hydrogen via water electrolysis, which is the alternative way to supply hydrogen for the industrial hydrogen consumer. The cost of on-site production sets the upper limit for the delivery costs.

Characteristics of the LOHC concept

Liquid organic hydrogen carriers (LOHCs) are liquids that can be reversibly hydrogenated and dehydrogenated. During dehydrogenation hydrogen is released as the sole product and the carrier liquid will be returned to its original state ready to be hydrogenated again (Fig. 1). Hydrogenation step is exothermic and it is typically carried out at temperatures between 100 and 250 °C and pressures 10–50 bar in the presence of catalysts [15]. Endothermic catalytic dehydrogenation takes place at elevated temperature (150–400 °C) and at low pressure [15]. The temperature and pressure levels are highly dependent on the chosen carrier molecule.

There are several LOHC compounds under research. For example, aromatic hydrocarbons or heterocyclic compounds such as carbazoles, pyridines or pyroles could be used as LOHCs. These have been recently reviewed by Aakko-Saksa et al. [15]. The ideal LOHC would have high hydrogen storage density, low reaction enthalpy, a low degradation rate and it would be non-toxic, low-cost and have a high enough melting point to stay in liquid form even in cold conditions. Furthermore, the conversion reactions would take place at mild conditions with low-cost catalysts. Lately most attention has been paid to dibenzyl toluene—perhydro-dibenzyltoluene (H0-DBT—H18-DBT), toluene—methylcyclohexane (TOL—MCH) and N-ethyl-carbazole—dodecahydro-N-ethylcarbazole (NEC—H12-NEC) systems, whose main properties are compared in Table 1.

The advantages of LOHCs compared to elemental hydrogen are multifold. LOHCs are considered safe, compatible with the existing fuel infrastructure and they enable higher hydrogen payloads for transportation and cost-efficient storages. In
addition there is no hydrogen loss even in the long-term storage or when transported overseas at normal conditions.

There are also some drawbacks that might limit the economic feasibility in some cases. One distinct feature is the high reaction enthalpies, meaning that a significant amount of heat is required to release the hydrogen. Considering the inevitable heat transfer losses, -25–30% of the released hydrogen would have to be burned if the heat is provided by hydrogen. Furthermore, as the required temperature level is quite high, it is not possible to use low-value waste-heat sources in most cases. However, the same amount of heat is released during hydrogenation; although at a lower temperature level. If this heat can be utilized, the energy penalty for dehydrogenation can be at least partially compensated. Therefore, the utilization of the hydrogenation heat and cost of dehydrogenation heat are important parameters for the analysis.

One drawback is also that dehydrogenation must be carried out at close to atmospheric pressure, while hydrogenation in most cases requires some additional pressurizing. In cases where high-pressure hydrogen is required by the user – such as bottle filling stations or mobility applications – this leads to a high energy demand for compression. Compression of hydrogen for hydrogenation requires energy and adds capital costs.

Furthermore, the depleted LOHC must be delivered back to the hydrogenation source. This complicates delivery chains if the same truck needs to deliver hydrogen to multiple locations in one trip. Lastly, the LOHC concept requires hydrogenation and dehydrogenation reactors, which increase hydrogen delivery costs.

### Methodology and theory

#### Case description

In this study we analyze the competitiveness of the LOHC supply chain in transporting by-product hydrogen from a chlor-alkali or chlorate plant to a single industrial customer (point-to-point delivery) by trucks. Dibenzyltoluene (DBT) was selected as the LOHC for this study. DBT offers high storage density, has a low melting point and is widely available at reasonable prices (commercial heat transfer fluid marketed under the trademark Marlotherm) and there is potential for even lower costs [30].

Two hydrogen demands and three transport distances are considered (Table 2). The selected hydrogen demands correspond to amounts of by-product hydrogen that could be available from typical sodium chlorate or chlor-alkali plants. The LOHC concept is compared to compressed gas delivery because it is the main alternative at the moment. Both steel bottle containers (200 bar) – representing the current logistic method – and the more advanced glassfibre composite cylinders (350 bar) are considered. For LOHC, two different investment costs were used due to the high uncertainty of the actual costs as will be discussed below. The delivery costs are also compared to costs of on-site production of hydrogen via water electrolysis, which is the alternative method for the hydrogen consumers to obtain low-carbon hydrogen. The difference between the delivery costs of by-product hydrogen and the on-site production costs of hydrogen must also cover the cost of by-product hydrogen (if it has any value for the producer) and possible additional purification.

By-product hydrogen is considered to be at atmospheric pressure after the conventional purification and is then compressed to the required pressure for each delivery option. There are differences in the purities of the hydrogen from chlor-alkali or chlorate plants. Hydrogen from the chlor-alkali plant is typically relatively pure and for industrial use it typically requires only some additional drying. Hydrogen from the chlorate plant, on the other hand, can contain around 2 vol-% of oxygen and trace amounts chlorine and carbon dioxide after the washing and drying steps [31]. Thus, some additional purification would be required, at least a de-oxo unit followed by a dryer. Possible additional purification is not, however, considered because the main aim of this study is to compare the competitiveness of the LOHC concept against compressed gas delivery and because hydrogen purity requirements will vary case by case. Similar purification would likely be required for each delivery method, when hydrogen is used in an industrial process.

In this study, hydrogenation heat is assumed to substitute for heat that would otherwise be generated by burning

### Table 1 — Comparison of the main properties of the most developed LOHCs [15,27–29].

<table>
<thead>
<tr>
<th></th>
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<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrogen storage capacity</td>
<td>6.2 wt%</td>
<td>5.8 wt%</td>
<td>6.2 wt%</td>
</tr>
<tr>
<td>Melting point/boiling</td>
<td>-39°C/390°C</td>
<td>69°C/378°C</td>
<td>-95°C/111°C</td>
</tr>
<tr>
<td>Enthalpy of reaction</td>
<td>65.4 kJ/mol H2 (27% of H2 LHV)</td>
<td>53.2 kJ/mol H2 (22% of H2 LHV)</td>
<td>68.3 kJ/mol H2 (28% of H2 LHV)</td>
</tr>
<tr>
<td>Temperature</td>
<td>50 bar</td>
<td>70 bar</td>
<td>20–40 bar</td>
</tr>
<tr>
<td>Dehydrogenation</td>
<td>150°C–300°C</td>
<td>170°C</td>
<td>95–125°C</td>
</tr>
<tr>
<td>Temperature</td>
<td>270–310°C</td>
<td>Close to ambient</td>
<td>3 bar</td>
</tr>
<tr>
<td>Price</td>
<td>4 €/kg</td>
<td>Close to ambient</td>
<td>250–450 °C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.3 €/kg</td>
</tr>
</tbody>
</table>
hydrogen at the by-product hydrogen production site. Both chlor-alkali and chlorate plants have several processes requiring steam which is produced by burning part of the by-product hydrogen. In chlor-alkali plants the main steam consumer is the concentration of the NaOH solution by water evaporation [32]. In a chlorate plant steam is used in drying, dissolving and precipitation of chlorate. Majority of the steam is typically used at around 3 bar (130 °C) although the process steam network can be at higher pressure. Hydrogenation of DBT is typically carried out at 150–180 °C making the reaction heat suitable for generating LP process steam. Furthermore, it has been shown [28,29] that it is possible to carry out hydrogenation in temperatures up to 300 °C using the same Pt–Al catalyst as for dehydrogenation. This means that the hydrogen that was previously burned in chlor-alkali or chlorate plant for steam production can be delivered to the merchant for free when corresponding amount of steam is received from the merchant.

The purity of hydrogen from dehydrogenation after simple condensation is assumed to be sufficient without any additional purification for the hydrogen consumer. There is a limited amount of public data on the purity of released hydrogen [33,34] and also the purity requirements are case specific. It is, however, important to control the humidity of DBT to minimize degradation during dehydrogenation. Bulpinar et al. [35] have shown that degradation during dehydrogenation can be reduced if water content in DBT is minimized. Hydrogen pressure from dehydrogenation (<5 bar) is assumed to be sufficient without additional compression. It is assumed that there is no waste-heat available for the dehydrogenation. Hydrogen release using fossil sources (e.g. natural gas or propane) was not considered as a sustainable option and it was assumed that there is no access to renewable alternatives such as biogas. This leaves two main options to consider:

1) Part of the released hydrogen is burned to provide the required heat. In this case, the total amount of hydrogen delivered would be 1.4 times the amount delivered by other methods (~30% of hydrogen needs to be burned).
2) Hydrogen is released using electrical heating.

As chlor-alkali and chlorate plants typically currently vent part of the hydrogen, the first option was seen as the most appropriate initial choice. Furthermore, as the utilization of steam from the hydrogenation reaction will free up more hydrogen, even more hydrogen might have to be vented. The effect of the heat source will be studied in the sensitivity analysis section. The considered case is illustrated in Fig. 2.

### Economic evaluation method

The delivery options are compared in terms of the total specific delivery costs of hydrogen (SC\textsubscript{del}), \(\text{€ kg}^{-1}\), which are divided into hydrogen processing costs and trucking costs (Eq. 3). Hydrogen processing includes compressors (SC\textsubscript{comp}), hydrogenation (SC\textsubscript{hyd}) and dehydrogenation (SC\textsubscript{dehyd}) reactors, stationary LOHC storage tanks (SC\textsubscript{storages}), and other site costs (SC\textsubscript{site}). Equations used to calculate each cost component will be presented in the following chapters.

\[
SC_{\text{total}} (\text{€ kg}^{-1}) = SC_{\text{processing}} + SC_{\text{trucking}} = SC_{\text{comp}} + SC_{\text{dehyd}} + SC_{\text{hyd}} + SC_{\text{storages}} + SC_{\text{site}} + SC_{\text{trucking}}
\]

(3)

Investment costs (IC) were annualized using the Capital Recovery Factor (CRF) method using an interest rate (i) of 8% and process specific lifetimes (n, years) (Eq. 4). Annualized investment costs (IC\textsubscript{ann}) are obtained by multiplying investment costs (IC) with CRF (Eq. 5).

\[
CRF = \frac{i \times (1 + i)^n}{(1 + i)^n - 1}
\]

(4)

\[
IC_{\text{ann}} = CRF \times IC
\]

(5)

The specific delivery costs are determined by first calculating the annual costs and dividing the annual costs by the delivered amount of hydrogen. For the LOHC cases only the share of the hydrogen that remains after part of the hydrogen has been burned to release the hydrogen, is considered when calculating the specific costs. Term “useable hydrogen” is used to describe this share of hydrogen later in the paper.
As chlorate and chlor-alkali plants are typically operated throughout the year at close to the maximum capacity, the full load hours (FLH) for the hydrogen production and consumption processes were assumed to be 8500 h/a. The electricity price considered in the analysis was 50 €/MWh, which includes grid fees and taxes.

Hydrogen conversion/processing

Hydrogenation and dehydrogenation (LOHC)

Assumptions for the DBT-based LOHC system are given in Table 3. Dehydrogenation pressure was not considered in the analysis. The typical pressure range of <5 bar was considered sufficient for the end-user without additional compression.

There is a high degree of uncertainty for the investment costs for dehydrogenation and hydrogenation reactors due to immaturity of the technology. Teichmann et al. [13] used the costs 260 and 40 €/kW_H2,LHV for "large-scale" hydrogenation and dehydrogenation reactors, respectively. Reub et al. [6] used values of 96 and 72 €/kW_H2,LHV for 300 t/d (417 MW_H2,LHV) units and suggested using a scale factor of 0.6. Eypasch et al. [22] estimated costs for small-scale LOHC systems. For 1 MW_H2,LHV system, specific costs were 252 and 368 €/kW_H2,LHV for hydrogenation and dehydrogenation reactors, respectively. Thus, there is a wide range in cost estimations.

There is also significant inconsistency regarding the respective costs of hydrogenation and dehydrogenation reactors. For example Teichmann estimated hydrogenation reactor to be significantly more expensive than dehydrogenation reactor while Eypasch and Reub estimated the reactor costs to be a lot closer to each other. Eypasch estimated that dehydrogenation reactor is more expensive and Reub had it the other way around.

In order to tackle the uncertainty regarding investment costs two different capital cost estimates are used in calculations. The values from Ref. [6] are considered as upper limit values (high CAPEX) and values from Refs. [22] represent lower limit values (low CAPEX). The specific delivery costs from the LOHC reactors were calculated using Eqs (6) and (7):

\[
SC_{dehyd} = \frac{IC_{dehyd} \times (CRF_{dehyd} + FC_{dehyd})}{Annual \ delivered \ useable \ hydrogen}
\]

\[
SC_{hyd} = \frac{IC_{hyd} \times (CRF_{hyd} + FC_{hyd})}{Annual \ delivered \ useable \ hydrogen}
\]

In addition to the LOHC reactors, two stationary storage tanks – one for the hydrogen rich and one for hydrogen lean LOHC – are required for both the hydrogen source and utilization sites. The total specific storage cost (SC_storage) is a sum of the specific costs of the steel tanks (SC_tanks), DBT (SC_DBT) and degradation of the DBT (DBT_degradation). The storage tank costs.
investment cost used in the analysis was 5 €/kgHz [6] and the required amount of DBT (DBTstorage) was considered to be three times the combined capacity of the tanker trucks. Cost of DBT (DBTprice) was assumed to be 4 €/kg [6]. Degradation rate (DBTdegradation) of 0.1% per cycle was used in the analysis. There is limited amount of information regarding degradation of DBT [33,34] so the same value that was used in Ref. [13] for N-ethylcarbazole was adopted. The total specific delivery costs from storages were calculated using Eq. (8).

\[ SC_{storages} = SC_{tanks} + SC_{DBT} + DBT_{degradation} \]

\[ IC_{comp} = \frac{IC_{tanks} \times CRF_{tanks} + SC_{DBT} \times DBT_{price} \times CRF_{DBT}}{p_{max} - p_{min} \times Useable\ storage\ density} \]

For the LOHC cases, the discharge pressure (p_d) equals the pressure of hydrogenation (50 bar). For filling tube trailers with compressed hydrogen, the logarithmic mean of the maximum pressure of hydrogenation (50 bar). For filling tube trailers with hydrogenation/dehydrogenation reactors represent the main equipment cost, there will be additional costs e.g. from piping, buildings and engineering ICsite. For GH2 cases these are estimated to be 500 and 1000 € for 2.5 and 10 MW cases respectively. For the LOHC cases slightly higher values of 750 and 1500 € were used due to the higher hydrogen flows and the added complexity caused by the utilization of steam. To calculate the specific cost, the investments were annualized using 15 years time span while no O&M costs were assumed for these investments (Eq. (14)).

\[ SC_{site} = \frac{IC_{site} \times (CRF_{site})}{Annual\ delivered\ useable\ hydrogen} \]

**Table 4 – Assumptions for calculating the specific works of compression.**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Description</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Z</td>
<td>hydrogen compressibility factor</td>
<td>Case dependent</td>
</tr>
<tr>
<td>R</td>
<td>universal gas constant</td>
<td>8.3145 J/(mole K)</td>
</tr>
<tr>
<td>T</td>
<td>suction (and intercooling)</td>
<td>313.15 K</td>
</tr>
<tr>
<td>M</td>
<td>molar mass of hydrogen</td>
<td>2.016 g/mol</td>
</tr>
<tr>
<td>P1</td>
<td>suction pressure</td>
<td>1 atm</td>
</tr>
<tr>
<td>P2</td>
<td>discharge pressure</td>
<td>Case dependent</td>
</tr>
<tr>
<td>N</td>
<td>number of compressor stages</td>
<td>Calculated</td>
</tr>
<tr>
<td>η</td>
<td>compressor isentropic efficiency</td>
<td>75%</td>
</tr>
<tr>
<td>γ</td>
<td>hydrogen specific heat ratio (\left(\frac{C_p}{C_v}\right))</td>
<td>1.41</td>
</tr>
</tbody>
</table>

Annual electricity costs of compression (ECcomp) were calculated from the specific work of compressor, annual hydrogen production and electricity price (Eq. (11))

\[ EC_{comp} = W_{comp} \times Annual\ hydrogen\ production \times Electricity \ price \]

The investment costs of compressors (ICcomp) were determined using the methodology created in a study for the Fuel Cells and Hydrogen Joint Undertaking [38]. The equation (Eq. (12)) takes into account the site capacity (Q, kg/h), total pressure ratio \(\left(\frac{p_2}{p_1}\right)\) and final pressure (Pout). The constants used were the same as in the study (A = 100, B = 300, Qref = 50, rref = 200/300, pref = 200, a = b = 0.66 and c = d = 0.25).

\[ IC_{comp} = A \left(\frac{Q}{Q_{ref}}\right)^a + B \left(\frac{Q}{Q_{ref}}\right)^a \left(\frac{p_2}{P_{ref}}\right)^c \left(\frac{p_1}{P_{ref}}\right)^d \]

Compressors were considered to have a lifetime of 15 years. Fixed O&M costs (FCcomp) were assumed to be 4% of the investment costs. Hydrogen losses were not considered as they are minor [6] and similar for each option. The specific cost of compression (SCcomp) in €/kg of useable hydrogen can then be written as

\[ SC_{comp} = \frac{IC_{comp} \times (CRF_{comp} + FC_{comp}) + EC_{comp}}{Annual\ delivered\ useable\ hydrogen} \]

**Transport/trucking**

The transport units consist of a truck and a trailer with a combined weight of ~53 t. The truck is assumed to be identical for each delivery method, but three different trailers are considered:

1) A trailer carrying two 200 bar steel bottle containers.
2) A trailer carrying a 350 bar glass fibre composite cylinder container.
3) A LOHC tanker trailer.

The first case acts as a reference case corresponding to the current logistic method, for example, in Finland. A 350 bar ISO40 (ADR 22) container is considered for the advanced compressed gas delivery option because it leads to a similar total mass as the steel container option. A tanker trailer with the capacity of 36 000 L was considered for the LOHC concept. It can carry around 2000 kg of hydrogen in the form of LOHC.
However, as hydrogen is burned to release the hydrogen, the useful capacity is only 1400 kg. The truck and trailer related assumptions are listed in Table 5.

The number of required deliveries per day will depend on the hydrogen demand and net hydrogen payloads. Theoretical maximum number of trips for each truck per day will depend on unloading/loading (drop-off/pickup) times, transport distance and average speed. Deliveries are assumed to take place 24/7 if necessary, which is also the case currently. For gaseous hydrogen delivery chains, it is assumed that the trailer (or container) full of hydrogen will be dropped off at the site and the empty one will be picked up.

The required number of trucks was calculated using the required number of deliveries and theoretical maximum number of trips each truck can make in one day and then rounding up to nearest larger integer. After rounding up, the lowest number of trips per day that meets the hydrogen demand is used in the analysis allowing non-integer numbers as well. For example 0.5 trips per day could mean delivery every other day.

The number of trailers needed for each truck is not the same for different hydrogen delivery options. For GH2 delivery options it is three times the number of trucks: one trailer is being transported, one trailer is being filled up at the hydrogen source and one trailer is being emptied at the hydrogen consumer site. The trailers act as storage and thus no additional storage is needed. In case of LOHC transport, the trucks will wait while the tanker trailer is first unloaded and then loaded and only one trailer per truck is needed. However, storage tanks are required for LOHC both at hydrogenation and dehydrogenation sites. These storage tanks were considered as part of the hydrogen processing costs.

The specific delivery costs from trucking consist of investment costs for trucks and trailers, operation and maintenance costs, fuel and personnel costs (Eq. (15)). The equations used to calculate the number of trucks and trailers required and trucking costs are given in the Supplementary material.

\[
SC_{\text{trucking}} \left( \frac{\text{€ kg}^{-1}}{} \right) = \frac{IC_{\text{trucking}} \times CRF_{\text{trucking}}}{\text{Delivered useable hydrogen per year}} + SC_{\text{trucking, O&M}} + SC_{\text{trucking, Fuel}} + SC_{\text{trucking, personnel}}
\]

(15)

### On-site production of hydrogen

For on-site production of hydrogen alkaline water electrolyzers with an efficiency of 62% ($\eta_{\text{electrolyser}}$) based on the lower heating value (LHV) of hydrogen are considered. The total investment costs ($IC_{\text{electrolysis}}$) for the 2.5 and 10 MW hydrogen demands were estimated to be 5 and 15 M€, respectively. The investment costs include also cost for installation, building, piping and grid connection. The lifetime of the electrolizers was 15 years and fixed operation and maintenance costs ($FC_{\text{electrolysis}}$) were taken as 5% of investment costs and they also include the stack replacement costs. It was assumed that neither oxygen nor low-temperature heat from the water electrolysis has any additional value. The same full load hours (FLH) as for hydrogen delivery cases was considered. The specific cost of on-site hydrogen was calculated using Eq. (16).

### Table 5 – Truck and trailer related assumptions.

<table>
<thead>
<tr>
<th>Truck</th>
<th>LOHC tanker trailer (36 000 l)</th>
<th>GH2 trailer (2 × 200 bar steel bottle ISO20 containers)</th>
<th>Advanced GH2 trailer (ISO40 HC 350 bar composite)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Investment cost</td>
<td>180 k€</td>
<td>140 k€ $^a$</td>
<td>530 k€ $^a$</td>
</tr>
<tr>
<td>Fixed O&amp;M</td>
<td>1.5 million km [39] or 8 years</td>
<td>15 years</td>
<td>15 years</td>
</tr>
<tr>
<td>Variable O&amp;M</td>
<td>4% of CAPEX</td>
<td>2% of CAPEX</td>
<td>2% of CAPEX</td>
</tr>
<tr>
<td>Net H₂ payload</td>
<td>0.1 €/km [39]</td>
<td>2000 kg (1400 kg useable$^b$)</td>
<td>400 kg</td>
</tr>
<tr>
<td>Unloading &amp; loading time (LOHC)</td>
<td>1 h + 1 h [13]</td>
<td></td>
<td>900 kg</td>
</tr>
<tr>
<td>Drop-off &amp; pick-up time (GH₂)</td>
<td></td>
<td>1 h + 1 h [13]</td>
<td>1 h + 1 h [13]</td>
</tr>
<tr>
<td>Fuel consumption</td>
<td>45 l/100 km</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fuel price (VAT0%)</td>
<td>1.05 €/l</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average speed (excl. unloading &amp; loading)</td>
<td>65–72–77 km/h (50–150–300 km)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Labour cost</td>
<td>26.5 €/h</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Truck availability</td>
<td>80% [9]</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^a$ Price indications from suppliers (rolling platforms for GH2 options was assumed to add 70 k€ to the indicated cost of the containers).

$^b$ In case hydrogen is released by burning hydrogen.
Results

Delivery costs

The delivery costs of hydrogen (bars) are compared with on-site production costs via water electrolysis with different electricity prices (horizontal lines) in Fig. 3. The difference between these costs is the maximum value for the by-product hydrogen. Thus, if hydrogen has any value at the chlorate/chlor-alkali plants or if additional purification is needed, these costs must be added to the delivery costs when comparing to on-site hydrogen production costs. These costs are based on useable hydrogen. In other words, the amount of hydrogen that is left after part of the hydrogen is burned in LOHC cases, as discussed above.

The calculated total delivery costs for 2.5 MW (1800 kg/day) and 10 MW (7200 kg/day) cases are 1.0–3.1 €/kg and 0.7–2.8 €/kg, respectively. With the low investment cost estimation for dehydrogenation and dehydrogenation reactors, LOHC and composite GH2 are almost equally competitive for 50–150 km transport distances while 300 km favors LOHC. Delivery costs using LOHC do not increase markedly with transport distance. However, if the investment costs for the LOHC reactors are in the upper range of literature estimates, composite GH2 is the most feasible option in every case. Delivery using 200 bar steel bottle containers is not the least-cost option in any of the cases and the costs increase steeply with transport distance.

LOHC and composite GH2 options scale more favorably from 2.5 to 10 MW. Cost reductions range from 23 to 37% for LOHC, 21–32% for composite cylinders and only 11–17% for the steel bottle containers. Thus, the higher the hydrogen demand, the more it favors the LOHC concept. However, the difference to composite GH2 is not so marked.

On-site hydrogen production costs are higher than the delivery costs of by-product hydrogen, which leaves some margin for the hydrogen raw material and purification costs. With the electricity price of 50 €/MWh, the margins for 2.5 MW and 10 MW cases were -2.2–3.0 and -2.1–2.8 €/kg respectively when the lowest cost delivery option is chosen for each transport distance. The production costs of electrolytic hydrogen are highly dependent on the electricity price. An increase of 10 €/MWh in electricity price will increase the cost (and thus the margin) by -0.5 €/kg. In case the hydrogen consumer would already have invested in an electrolyser, the margins would drop to -1.2–2.0 and -1.3–2.0 €/kg due to the fact that by-product hydrogen would then compete against the variable costs of electrolysis only. However, in this case by-product hydrogen could allow avoiding the use of an electrolyser when electricity prices are high. Thus, margins could be even higher than in the reference case, but the delivered amounts of hydrogen would be lower.

Cost breakdowns

The detailed costs breakdowns for 2.5 MW and 10 MW cases are shown in Fig. 4 and Fig. 5, respectively. For the LOHC delivery chains, the main costs are related to hydrogen processing, while for the GH2 delivery chains the costs are governed by trucking costs especially in the case of steel bottle containers and longer transport distances. This is the reason for the differences in the effects of transport distance on the delivery costs between the delivery methods. Additionally, electricity costs for compression make up only a minor share of total delivery costs, and there is not much difference between different options. Even though the hydrogenation pressure (50 bar) is lot lower than the pressure in GH2 options (200/350 bar), the compressor energy cost is
actually slightly higher in the LOHC cases. This is due to the fact that 1) when filling a bottle or a cylinder not all of the hydrogen has to be compressed to the final pressure and 2) more hydrogen needs to be delivered in the LOHC cases to meet the same demand as part of the hydrogen was assumed to be burned.

**Delivery fleet**

Table 6 shows the required number of trucks and trailers and the related CAPEX for each case. With the lower hydrogen demand, one LOHC tanker trailer can deliver the needed hydrogen even when the transport distance is 300 km. The higher hydrogen demand would increase the required number of tanker trailers to three. The corresponding case using steel bottle containers would require a massive fleet of 10 trucks and 30 trailers and even with the composite cylinders 5 trucks and 15 trailers would be needed.

The fleet costs vary from 0.3 to 1.0 M€ for the LOHC delivery, 1.8–7.8 M€ for the steel bottle containers and 1.4–7.2 M€ for the composite cylinders. The corresponding shares of fleet related CAPEX (of total CAPEX) vary from 2 to 8% for the LOHC, 46–77% for the steel bottle containers and 37–67% for the composite cylinders.

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**Fig. 4** – Detailed cost breakdowns for 2.5 MW hydrogen demand cases (50, 150 and 300 km). H₂ processing includes compression, hydrogenation, dehydrogenation and related site costs (piping, buildings, engineering) but not purification.

**Fig. 5** – Detailed cost breakdowns for 10 MW hydrogen demand cases (50, 150 and 300 km). H₂ processing includes compression, hydrogenation, dehydrogenation and related site costs (piping, buildings, engineering) but not purification.
Generalized results

The results are generalized in Fig. 6, which depicts the most feasible transport method and the corresponding delivery cost for different hydrogen demands and transport distances. For the LOHC concept, the low CAPEX case was considered. In most cases the LOHC concept becomes the lowest cost option when the delivery distance increases to 100 km. The irregularities are due to the requirement that the number of trucks and trailers need to be a natural number (non-ideal scaling). It should be noted that the model used did not optimize the delivery fleet for each case. For example, the capacities of trailers were fixed.

Sensitivity analysis

General sensitivity analysis

The sensitivity of the results with respect to the selected parameters (WACC, electricity price, diesel price, LOHC price, LOHC reactor costs, degradation of LOHC) were studied by comparing the delivery costs via LOHC (assuming low CAPEX) and composite GH2 when transport distance is 150 km and hydrogen demand 2.5 MW (1800 kg/day) (Fig. 7).

The most important factor is the capital costs of the hydrogen conversion reactors for the LOHC concept. As discussed earlier the capital costs involve a very high uncertainty due to immaturity of the technology.

The WACC, electricity price and diesel price were found to have only a minor effect on the respective competitiveness of the two delivery options:

- Electricity demands and total capital costs were in the same range: the higher hydrogen processing CAPEX for LOHC chain are compensated by the lower trucking related CAPEX.
- Fuel costs on the other hand represented only a small share of the total costs and thus lower fuel costs for LOHC did not reduce total costs markedly.
- The price of the LOHC compound (dibenzyl toluene, DBT) had only a relatively small effect on the results despite the high sensitivity price range (2–6 €/kg) because hydrogen is

<table>
<thead>
<tr>
<th>Hydrogen demand (kg/day)</th>
<th>GH2 (composite) most feasible</th>
<th>One-way transport distance</th>
<th>LOHC (low CAPEX) most feasible</th>
</tr>
</thead>
<tbody>
<tr>
<td>25 km</td>
<td>100 km</td>
<td>200 km</td>
<td>300 km</td>
</tr>
<tr>
<td>1800 2.5</td>
<td>0.98 1.03 1.09</td>
<td>1.14 1.24 1.30</td>
<td>1.34 1.37 1.44</td>
</tr>
<tr>
<td>3600 5</td>
<td>0.72 0.78 0.83</td>
<td>0.89 0.94 0.98</td>
<td>1.01 1.05 1.12</td>
</tr>
<tr>
<td>5400 7.5</td>
<td>0.62 0.78 0.83</td>
<td>0.77 0.85 0.92</td>
<td>0.89 0.97 1.04</td>
</tr>
<tr>
<td>7200 10</td>
<td>0.65 0.70 0.75</td>
<td>0.84 0.91 0.95</td>
<td>0.98 1.04 1.07</td>
</tr>
<tr>
<td>9000 12.5</td>
<td>0.60 0.65 0.76</td>
<td>0.72 0.81 0.89</td>
<td>0.82 0.91 1.01</td>
</tr>
<tr>
<td>10800 15</td>
<td>0.56 0.66 0.71</td>
<td>0.68 0.77 0.85</td>
<td>0.79 0.89 0.99</td>
</tr>
<tr>
<td>12600 17.5</td>
<td>0.58 0.63 0.71</td>
<td>0.60 0.72 0.81</td>
<td>0.72 0.86 1.01</td>
</tr>
<tr>
<td>14400 20</td>
<td>0.55 0.60 0.69</td>
<td>0.55 0.67 0.77</td>
<td>0.60 0.74 1.03</td>
</tr>
<tr>
<td>16200 22.5</td>
<td>0.53 0.61 0.67</td>
<td>0.50 0.67 0.74</td>
<td>0.59 0.72 1.05</td>
</tr>
<tr>
<td>18000 25</td>
<td>0.51 0.59 0.67</td>
<td>0.48 0.66 0.74</td>
<td>0.57 0.71 1.07</td>
</tr>
<tr>
<td>19800 27.5</td>
<td>0.52 0.60 0.65</td>
<td>0.46 0.63 0.72</td>
<td>0.55 0.69 1.09</td>
</tr>
<tr>
<td>21600 30</td>
<td>0.51 0.59 0.66</td>
<td>0.44 0.60 0.69</td>
<td>0.52 0.67 1.06</td>
</tr>
</tbody>
</table>

Fig. 6 – Lowest delivery cost of hydrogen at different hydrogen demands and one-way delivery distances. Orange background refers to cases when composite cylinders are the lowest cost option while turquoise background depicts cases where LOHC concept (assuming low investment costs) leads to the lowest costs.
not stored for extended periods of time and because of the low assumed degradation rate.

Degradation on the other hand could have a significant effect due to continuous hydrogenation/dehydrogenation cycles if the actual degradation is significantly higher than assumed here (0–0.2% per cycle).

**The effect of the heat supply method**

In the reference cases, it was assumed that part of the delivered hydrogen will be burned to provide the heat required to release hydrogen from the LOHC. In the considered case of the first application of the LOHC concept to chlorate/chlor-alkali plants this choice was justified. However, in future when the demand for hydrogen increases, it may no longer be viable to burn valuable hydrogen. Also, in the cases where hydrogen is purposely produced via water electrolysis, electrical heating for dehydrogenation would be more feasible due to the losses occurring during water electrolysis and due to the need to over dimension the hydrogen supply chain. Thus, the utilization of other external heat sources for dehydrogenation was also studied for the 2.5 MW and 150 km case with low CAPEX for the LOHC.

When dehydrogenation was considered to be carried out by burning part of the delivered hydrogen, the value of hydrogenation heat (or hydrogen) was not required to calculate the delivery costs because hydrogenation heat was assumed to be exchanged with this “additional fuel hydrogen” 1:1. However, when considering external heat sources, the value of heat released in hydrogenation must also be considered, in addition to the cost of heat for dehydrogenation.

**Fig. 8** shows the delivery costs with respect to the cost of dehydrogenation heat and the value of hydrogenation heat. Even with equally valued hydrogenation and dehydrogenation heats (diagonal of the matrix) the delivery costs would be lower than in the reference case (1.27 vs 1.03 €/kg of useable H₂) because less hydrogen needs to be transported when external heat is utilized. In the reference case the total amount of hydrogen delivered as LOHC was ~2600 kg/day, while for external heat utilization 1800 kg/day is sufficient.

<table>
<thead>
<tr>
<th>Cost of heat for dehydrogenation</th>
<th>Value of by-product heat from hydrogenation</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 €/MWh</td>
<td>0.03</td>
</tr>
<tr>
<td>10 €/MWh</td>
<td>0.94</td>
</tr>
<tr>
<td>20 €/MWh</td>
<td>0.85</td>
</tr>
<tr>
<td>30 €/MWh</td>
<td>0.76</td>
</tr>
<tr>
<td>40 €/MWh</td>
<td>0.67</td>
</tr>
<tr>
<td>50 €/MWh</td>
<td>0.58</td>
</tr>
<tr>
<td>60 €/MWh</td>
<td>0.49</td>
</tr>
<tr>
<td>0 €/MWh</td>
<td>1.03</td>
</tr>
<tr>
<td>10 €/MWh</td>
<td>1.12</td>
</tr>
<tr>
<td>20 €/MWh</td>
<td>1.21</td>
</tr>
<tr>
<td>30 €/MWh</td>
<td>1.30</td>
</tr>
<tr>
<td>40 €/MWh</td>
<td>1.48</td>
</tr>
<tr>
<td>50 €/MWh</td>
<td>1.57</td>
</tr>
</tbody>
</table>

**Fig. 7** – Hydrogen delivery cost sensitivity analysis for the 2.5 MW & 150 km case.
The first column in Fig. 8 corresponds to a situation where there is no value for the heat released during hydrogenation. This could be the case when the utilization of heat from hydrogenation would just increase the amount of hydrogen that will be vented at the chlorate/chlor-alkali plant. In this case the use of external heat would be more beneficial than using excess by-product hydrogen if heat can be obtained for less than ~26 €/MWh. Renewable or low-carbon heat at sufficient temperature is not typically available at such low prices. However, if hydrogenation heat is valued at 30 €/MWh, the maximum price for external heat would be ~56 €/MWh which is close to current electricity prices for industry. This makes direct electrical heating an economically competitive option as well.

In addition to economic aspects, a simple analysis was carried out to compare the CO2 emissions from using by-product hydrogen and electricity as heat sources for dehydrogenation in today’s situation in Finland. In the analysis it was assumed that by-hydrogen is CO2-free as it is a by-product that would be vented otherwise. Electrical heating was considered to have an ideal efficiency of 100% and the average CO2 intensity of Finnish grid electricity (114 gCO2/MWh in 2018 [40]) was used. Diesel in Finland contains on average 13.2 vol-% of biodiesel which corresponds to an average CO2 intensity of Finnish grid electricity (114 gCO2/MWh in 2018 [40]) was used. Diesel in Finland contains on average 13.2 vol-% of biodiesel which corresponds to an emission factor of 2.2 gCO2/km [41]. The analysis showed that in today’s situation burning by-product hydrogen leads to a significantly lower CO2 emissions (0.56 vs. 1.42 kgCO2/kgH2 for the 150 km cases). Thus, the utilization of grid electricity for dehydrogenation emits a lot more CO2 than are saved via lowered fuel consumption of the trucks and reduced compression demand. To achieve lower emissions using electrical heating would require electricity having CO2 intensity less than ~8 gCO2/MWh. Even with 300 km transport distance the maximum CO2 intensity would still be only ~15 gCO2/MWh. However, a more detailed analysis should be carried out.

The first row in Fig. 8 represents a situation where there is free waste-heat at sufficiently high temperatures available at the hydrogen consumption site. In this special case, the costs could decrease by ~40% compared to the reference case if the value of hydrogenation heat is 30 €/MWh. This indicates that LOHC-based deliveries would be a very attractive solution for the sites where waste- or low-cost heat is available. For example, an industrial site where hydrogen is produced by steam methane reforming (SMR) and which would require additional hydrogen production capacity, could be a favourable case for LOHC-based hydrogen delivery. If the by-product steam from reforming is not needed or is converted to electricity at low efficiency, it could be used for dehydrogenation of LOHC instead and the investment to a new hydrogen production unit could be avoided.

Validation of the results

In real world cases the delivery costs would be slightly higher for every option than reported here, as not all cost parameters were included. The uninterrupted supply of hydrogen would require investment into backup units and some safety margin, which were not considered in this study. It was also assumed that the purity of the released hydrogen is sufficient without complex purification steps. The validity of this assumption should be considered case by case as purity requirements will vary depending on the end-use. The cost increase for the possible additional purification must also be considered case by case and it may be significant, for example, for fuel cell applications.

On the other hand, the LOHC concept would have other advantages that are not captured by the calculated hydrogen delivery costs. The LOHC concept would enable significantly larger hydrogen storage at reasonable costs, which could make the delivery chain much less vulnerable compared to GH2 delivery. With minor over dimensioning of the system, the delivery schedule could be more flexible and it would be possible to prepare for the maintenance and unscheduled breaks at the hydrogen source plant.

Conclusions

A techno-economic model comparing hydrogen road delivery costs via the LOHC concept and compressed gas was created. Higher hydrogen demands and especially longer transport distances were shown to favour the LOHC concept over compressed gas, as the costs for the LOHC-based delivery are mainly governed by hydrogen processing costs while for compressed gas delivery the trucking costs are more significant.

Heat integration was found to be key for the LOHC concept. If hydrogen is burned to provide the heat for dehydrogenation, LOHC was found competitive against composite cylinders for over 100 km transport distances if the capital costs for the LOHC reactors are in the lower end of the literature estimates. Burning of hydrogen to provide heat is justified in the early applications for LOHC in chlorate/chlor-alkali plants but in the future external heat supply will become a more feasible option. In the most favourable cases the heat from dehydrogenation would be utilized in the hydrogen source site and dehydrogenation would be carried out with external low cost heat sources at the hydrogen consuming site. In these kind of cases, LOHC supply method was found to be the least-cost method. Also, the utilization of direct electrical heating was found competitive as long as low-cost electricity is available.

Sensitivity analysis shows that the highest uncertainty is in the capital cost for the LOHC reactors followed by the degradation rate of the LOHC compound.

Declaration of competing interest

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ijhydene.2020.08.186.

REFERENCES


