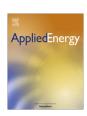


Contents lists available at ScienceDirect

Applied Energy

journal homepage: www.elsevier.com/locate/apenergy



Seasonal storage and alternative carriers: A flexible hydrogen supply chain model



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HIGHLIGHTS

- Techno-economic model of future hydrogen supply chains.
- Implementation of liquid organic hydrogen carriers into a hydrogen mobility analysis.
- Consideration of large-scale seasonal storage for fluctuating renewable hydrogen production.
- Implementation of different technologies for hydrogen storage and transportation.

ARTICLE INFO

Article history: Received 21 December 2016 Received in revised form 4 April 2017 Accepted 4 May 2017 Available online 18 May 2017

Keywords: Hydrogen Hydrogen infrastructure Seasonal storage Renewable energies LOHC FCEV

ABSTRACT

A viable hydrogen infrastructure is one of the main challenges for fuel cells in mobile applications. Several studies have investigated the most cost-efficient hydrogen supply chain structure, with a focus on hydrogen transportation. However, supply chain models based on hydrogen produced by electrolysis require additional seasonal hydrogen storage capacity to close the gap between fluctuation in renewable generation from surplus electricity and fuelling station demand. To address this issue, we developed a model that draws on and extends approaches in the literature with respect to long-term storage. Thus, we analyse Liquid Organic Hydrogen Carriers (LOHC) and show their potential impact on future hydrogen mobility. We demonstrate that LOHC-based pathways are highly promising especially for smaller-scale hydrogen demand and if storage in salt caverns remains uncompetitive, but emit more greenhouse gases (GHG) than other gaseous or hydrogen ones. Liquid hydrogen as a seasonal storage medium offers no advantage compared to LOHC or cavern storage since lower electricity prices for flexible operation cannot balance the investment costs of liquefaction plants. A well-to-wheel analysis indicates that all investigated pathways have less than 30% GHG-emissions compared to conventional fossil fuel pathways within a European framework.

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1. Introduction

The transition to renewable energy is a centrepiece of global environmental policies. The Paris Agreement from November 2015 is intended to reduce net green-house gas (GHG) emissions to zero by the second half of this century [1,2]. The targets set out by the German government foresee a reduction of GHGs in the energy system of 80% by 2050 against 1990 levels [3]. These targets fundamentally depend on the penetration of renewable energy technologies like wind and solar power across all energy sectors. For example, in 2015 the German electricity sector already

produced 32.6% renewably [4]. However, with increasing renewable power, the necessity of storage options to counter the effects of the fluctuating nature of wind and solar power correspondingly rises. In 2014, 1581 GWh of renewable power was curtailed due to grid congestion, resulting in compensation payments by the EEG levy¹ of 82 million Euro [4]. To avoid such financial and efficiency-

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¹ The German EEG levy was introduced in the year 2000 with the German Renewable Energy Act (EEG). It supports the penetration of renewable energy in the power sector by setting fixed feed-in remunerations for renewably-generated energy. Furthermore, renewable energy gain unlimited priority feed-in. In case of curtailment of renewable power generation due to grid congestion, the renewable energy plant gets compensation payments for the loss of revenues. The EEG represents the gap between revenues and expenses caused by renewables and is paid by the consumers [61].

usurious costs, with the share of renewables projected to be 80-100% by 2050, Germany is in need of storage options at the TWh-scale, which can be achieved with Power-to-Gas via water electrolysis [5,6]. Meanwhile, the mobility sector accounts for around 17.7% of total GHG emissions in Germany [4]. In 2015 however, renewables met only 5.3% of the total energy consumption of this sector [7]. Furthermore, the latest figures on air quality in German cities indicate major problems in fulfilling European regulations [8]. Zero emission vehicles like battery electric vehicles (BEV) and fuel cell electric vehicles (FCEV) fuelled by renewably-produced hydrogen have the potential to reduce both CO2 emissions and locally active pollutants at the same time [3,9]. Moreover, hydrogen facilitates the coupling of electricity with the mobility sector: producing hydrogen via electrolysis during periods when high renewable power generation exceeds grid load would offer an emissionfree fuel for FCEVs [10-13].

Establishing hydrogen as a fuel for transportation requires a detailed analysis of the entire supply chain. This includes how hydrogen is to be produced, its large-scale storage that takes the seasonal intermittency of renewable power generation² into account, its transportation and distribution from a central production plant to fuelling stations as well as the fuelling stations themselves. The optimal tank storage system for on-board storage in FCEVs is generally considered to be a 350 or 700 bar compressed gas vessel [14]. However, the supply chain up until the onboard storage is still a focus of investigation. Numerous studies [14–19] investigate the most cost-efficient supply structure between production and transportation. The "state of the art" hydrogen supply chain thereby mostly relies on pure hydrogen provided as compressed gas or cryogenic liquid [20,21]. Yang and Ogden [22] investigate a method for comparing the different transport possibilities of tube or liquid trailer truck vs. pipeline delivery. They show that each technology has a maximally cost-efficient niche and there is no single perfect solution for the entire system. Elgowainy and Reddi [18] develop an Excel tool for calculating the cost of hydrogen supply while varying different input parameters like FCEV market penetration, refuelling station capacity, transmission mode or production volume for different delivery scenarios. Like Yang and Ogden, they focus on the three main delivery pathways of tube trailer, liquid trailer and pipeline. Although, hydrogen production is not calculated inside either model and is instead assumed to be an input. As such, the influence of hydrogen production on storage demand was not investigated and, considering hydrogen mobility as part of a future renewable energy system and the utilization of electrolysis systems from renewable sources, this influence should not be overlooked. Seasonal storage is identified as a key factor in several studies [10,18], although only consideration of subterranean options, such as salt caverns or depleted oilfields, has been found.

High-pressure storage tanks remain cost-intensive (800 \$/kg hydrogen [23]) while the liquefaction of hydrogen is energy-intensive (30% of the LHV of hydrogen [24]). Aside from the compressed and liquid applications, many different storage solutions for hydrogen are possible. Chemisorption – like metal hydrides, chemical hydrides or liquid organic hydrogen carrier (LOHC) – along with physisorption – via carbon nanotubes or metal organic frameworks (MOF) – are the two basic mechanisms for storing hydrogen other than conventional compressed and liquid storage [25]. The 2010 Nexant Report [15] included alternative carrier systems like LOHCs and metal hydrides in its calculations. Thereby, it was determined that "using alternative carriers in a pathway that discharges hydrogen at the fuelling station and supplies compressed hydrogen to vehicles will offer little or no benefit for fuelling station costs" [15]. In contrast, Teichmann [2,26,27] shows

that the main benefit of an LOHC system lies in the ease and low cost of storage and transportation.

According to Dagdougui [28], most hydrogen supply chain models focus on mathematical optimization methods to minimize the cost of an explicit case, like Samsatli [29], who models a hydrogen infrastructure for supplying Great Britain's transport sector with hydrogen. Nevertheless, the literature is lacking a modelling approach that enables the easy investigations of upcoming new technologies for hydrogen infrastructure like LOHC or additional chain parts like seasonal storage. Implementing them directly into an optimization approach without checking their potential applicability will diminish the model performance. Therefore, this work investigates the application area of different hydrogen supply chain architectures through a point-to-point analysis based on the methodology of Yang and Ogden drawing on current data and extending the considered technologies. Therefore, it considers the full supply chain from hydrogen production by electrolysis. large-scale storage for the temporal gap between demand and supply, the transportation and the fuelling station facilities necessary to fill a 700 bar compressed gas tank. Furthermore, LOHCs are discussed as an alternative carrier system to investigate their impact on hydrogen mobility. All results shown with this model have a European scope. Applying this model to other regions of the world could significantly change the results. Nevertheless, the elaborated sensitivity analysis of this paper shows the most sensitive input parameters.

2. Hydrogen storage and delivery

2.1. Storage methods

A key challenge for hydrogen mobility is its extremely low density (0.09 kg/m³), in accordance with its being the lightest element [30,31]. Even with a high specific energy of 33 kWh/kg, energy density remains low at ambient conditions (0.003 kWh/l) compared to conventional fuels such as gasoline (10 kWh/l). Depending on the storage and transportation technology, higher energy densities lead to lower specific costs due to limited volume and weight. Therefore, the energy density of hydrogen requires further adjustments.

Fig. 1 displays the volumetric and gravimetric density ranges of different technologies for hydrogen storage found in literature. Compressed and liquid hydrogen are the current state of the art in hydrogen storage. All alternative hydrogen carriers substances like MOFs, metal hydrides, chemical hydrides or LOHCs were initially investigated for on-board hydrogen storage in a fuel cell vehicle. However, the on-board hydrogen storage system seems to be fixed for the near future at 700 bar [14]. This study focuses on infrastructure for the storage and transportation of hydrogen. While storage systems for mobile storage allow higher capital investments, the use of carrier systems for the supply infrastructure requires a cheap carrier compound with easy handling and a high hydrogen share. Solid carriers like MOFs or metal alloys do not fulfil these requirements. Chemical and metal hydrides still lack regenerable carriers, which are not dismantled during unloading, with scalable loading and unloading reactions, and offering easy transportation of the loaded and unloaded carrier compound. LOHCs are thereby promising candidates for hydrogen infrastructure, since the loaded as well as unloaded carrier exist naturally in a liquid state [2,26,33-35]. From here, each of the storage methodologies implemented in this work are explained in finer detail.

2.1.1. Compressed hydrogen

The most common way to achieve higher hydrogen storage densities is via compression in gaseous form (GH₂). Stationary tube

² In case of Germany, wind power plants produce more energy during the winter than the summer contrary to photovoltaic, which produces more in the summer.

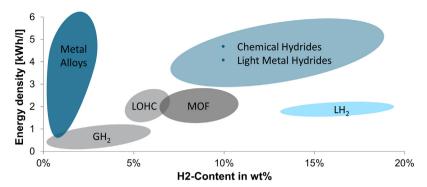


Fig. 1. Comparison of different hydrogen storage technologies [2,32]; LOHC – Liquid Organic Hydrogen Carriers; MOF – Metal Organic Frameworks; GH₂ – Gaseous Hydrogen; LH₂ – Liquid Hydrogen.

systems normally have pressures of between 200 and 350 bar [30]. GH_2 at 700 bar is generally regarded as the most viable storage system for on-board hydrogen storage in automotive applications [14]. However, even at this high pressure the density of hydrogen remains low (700 bar: 40 g/l = 1.3 kWh/l). Furthermore, high-pressure gas vessels have high investment costs and special requirements for the vessel material. Even low pressure applications have capital costs of about 850\$ per kg of storable hydrogen [21].

2.1.2. Liquid hydrogen

Liquid hydrogen (LH₂) offers the possibility of increasing the density up to 71 kg/m³ (2.4 kWh/l) by cooling the hydrogen below 21 K. While the minimum theoretical energy demand for the lique-faction process is 3.9 kWh/kg (with conversion to para-LH₂), actual liquefaction plants typically need between 12 and 15 kWh/kg in the form of electricity [36]. This amounts to 36–45% of the overall hydrogen energy content. LH₂ can be stored in cryogenic tanks with a robust insulation at low pressure (<10 bar), which allows the use of large bulk storage systems with high energy densities. Since it is not possible to prevent all heat from flowing into the tank, hydrogen boil-off must be taken into account.

2.1.3. Liquid organic hydrogen carrier (LOHC)

LOHC systems are composed of pairs of hydrogen-lean and hydrogen-rich, liquid organic compounds that store hydrogen by means of repeated, catalytic hydrogenation and dehydrogenation cycles. While catalytic hydrogenation for hydrogen storage is exothermal, the corresponding hydrogen release by catalytic dehydrogenation is endothermal (Fig. 2). Theoretically, hydrogen storage and release is a reversible reaction, but reusing the heat arising from hydrogenation during the dehydrogenation process represents a challenge.

The main advantage of the LOHC technology is that it enables hydrogen storage in chemically bound form under ambient conditions. Thus, no high-pressure or super insulated tank is required. Furthermore, the technology can build on the existing infrastructure for fossil fuel, e.g. making use of tanker ships, rail trucks, road tankers and tank farms [26].

In the past, different chemicals have been investigated in the past for their potential as LOHC compounds. Brückner et al. show that the LOHC system dibenzyltoluene (H0-DBT)/perhydro dibenzyltoluene (H18-DBT) is highly promising for several reasons. While its reaction enthalpy (65 kJ/mol = 8.9 kWh/kg_{H2}) and the dehydrogenation temperature of H18-DBT are relatively high (>260 °C), its storage density of up to 6.2°weight-% of hydrogen compared to the total weight of the carrier, its ease of handling (no "dangerous goods" complications), its thermal robustness and its high cycle stability offer significant benefits. Furthermore,

HO-DBT is available in large quantities, as it is used as an industrial heat transfer fluid on a large scale (e.g. under the tradename Marlotherm SH) [37]. Considering these advantages, this study assumes an LOHC system based on the HO-DBT/H18-DBT pair.

2.1.4. Seasonal storage

The storage of hydrogen for on-board applications has been widely investigated [25,38,39]. Such applications undergo moderate numbers of charge cycles during a year and have small capacities compared to the overall throughput. While German wind power peak production takes place in winter, solar power production in the northern hemisphere reaches maximum power output during the summer. Seasonal storage will close the gap between the seasonal fluctuations of renewable power generation from winter to summer. Therefore, seasonal storage has high storage capacities with a low number of charge cycles during the year. From an economic point of view, a seasonal storage system requires small expenses regarding the capacity and small losses over long residence times [40].

2.2. Delivery

Today, hydrogen is transported either by trucks as LH₂ or GH₂ in trailers or via pipeline as GH₂. In a hypothetical future global hydrogen-incorporated economy, ship and rail transportation could play an important role as well. While pipeline transportation requires high capital investment costs at limited flow rates it compensates for this with lower operational costs. Meanwhile, every truck trailer has a fixed number of storage units with fixed capacities. The main indicator for comparing different trailer delivery methods is the hydrogen storage capacity of the trailer, which is limited by its volume and weight limits.

3. Method and data

This study includes a well-to-tank analysis that estimates the GHG emissions for conditioning a transportation fuel. We analyse renewably-produced hydrogen as transportation fuel for FCEVs with respect to their different ways of storing and transporting hydrogen. The assumed feedstock for hydrogen production is available electricity from renewable energy sources and the fuel is hydrogen compressed to 700 bar. In between are four basic stages, namely: hydrogen production, storage, transport and fuelling at the station. These stages are displayed in Fig. 3. Chaining together these technologies will necessitate further interconnections to enable state changes such as liquefaction, compression, or hydrogenation and dehydrogenation. For this reason, the fuelling station stage includes the reconversion to gaseous hydrogen as well as the fuelling process. Each link of this process chain is analysed in the



Fig. 2. Systematic of liquid organic hydrogen carriers.

framework of one corresponding module and is evaluated as a static calculation.

As Table 1 indicates, we assume that the electrolyser and the conversion to the storage system have a flexible operation mode at 5300 full load hours per year with a levelised cost of electricity for wind power generation of 0.06 Ct/kWh [41]. Moreover, they have access to renewable electricity and a limited operating period to capitalize on lower electricity prices. The storage module then smooths the seasonal gap between hydrogen production and demand, with storage capacity based on 60 daily productions (18 days of seasonal storage and 42 days for strategic reserve) [10]. 30% of the overall annual production is assumed to be stored before being transported. This amounts to 1.85 charging cycles per year and an average residential time of 200 days. Thereby, we assume constant demand after the storage module and a continuous operation of all plants, whereas the electricity grid provides electric energy. The utilization rate of the fuelling station is assumed to be 70% of the daily capacity as an averaged value during its depreciation period.

The price of electricity incorporates the annual overall electricity consumption of the module as seen in Table 2. Eurostat offers three different values for industrial consumers:

- · Excluding all taxes and levies
- Excluding Value Added Tax (VAT) and all recoverable taxes and levies
- Including all taxes and levies

Agora Energiewende [42] claims that the German EEG Levy as the main levy in Germany will be at a similar level in 2035 as it is today. Therefore, the most probable electricity price for pre-tax calculations will correspond to exclude VAT and the recoverable taxes and still include the EEG levy.

The investment costs of chemical plants often scale with rising throughput, whereas specific investment costs are decreasing. Scaling functions can be used to estimate the resulting investment costs:

$$Invest_{Total} = Invest_{Base} \left(\frac{capacity}{Invest_{Compare}}\right)^{Invest_{Scale}}$$

Thereby, two phenomena influence the capacity. On the one hand, in the case of flexible plants with lower operational hours, an overcapacity factor f_{cap} is needed to supply the same amount of hydrogen per year. On the other hand, since losses of hydrogen in a specific module lead to higher hydrogen demand in a previous module, an overproduction f_{prod} is also included.

$$capacity = capacity_{nominal} * f_{cap} * f_{prod}$$

The specific capital expenditures per year are calculated by an annuity factor AF based on the depreciation period n and the Weighted Average Cost of Capital (WACC):

$$AF = \frac{(1 + WACC)^n \cdot WACC}{(1 + WACC)^n - 1}$$

For specific capital expenditure (*CAPEX*) per kg of hydrogen we must consider the real annual throughput:

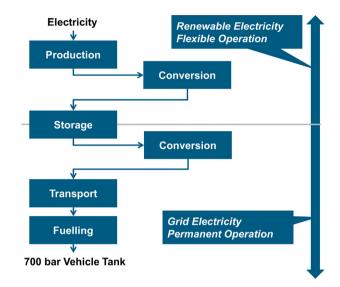


Fig. 3. Model setup.

Table 1General assumptions for all modules.

Weighted Average Cost of Capital (WACC)	8%	Utilization fuelling station	70%
Electricity cost RES ^a	0.06 €/kWh	Diesel cost	1.2 €/l
Op. Hours (RES) ^a	5300 h	Driver wage [26]	35 €/h
Storage Days ^a	60 days	Natural gas cost	0.04 €/kWh
Storage Partb	30%	Water cost	4 €/m³

^a IEK-3 energy concept 2.0 gives 5300 h electrolyser full load hours supplied by wind energy with levelised cost of electricity of 6 Ct/kWh, WACC of 8%, storage amount of 90 TWh = 60 days [41].

Table 2Electricity price industrial consumers (Average 2015, excluding VAT and other recoverable taxes and levies) [43].

Band IA: Consumption < 20 MWh	0.2190 €/kWh
Band IB: 20 MWh < Consumption < 500 MWh	0.1738 €/kWh
Band IC: 500 MWh < Consumption < 2 000 MWh	0.1501 €/kWh
Band ID: 2 000 MWh < Consumption < 20 000 MWh	0.1308 €/kWh
Band IE: 20 000 MWh < Consumption < 70 000 MWh	0.1118 €/kWh
Band IF: 70 000 MWh < Consumption < 150 000 MWh	0.0976 €/kWh

$$CAPEX = \frac{Invest_{Total} * AF}{throughput_{Nominal} * f_{prod}}$$

The operational costs are split into fix (fixOPEX) and variable (varOPEX) operational expenditures. The fixOPEX pertains to the fixed operational and maintenance costs (OM). These are represented by a percentage of the investment and are calculated similarly to the CAPEX:

$$\textit{fixOPEX} = \frac{\textit{Invest}_{\textit{Total}} * \textit{OM}}{\textit{throughput}_{\textit{Nominal}} * f_{\textit{prod}}}$$

^b Share of hydrogen that needs to be stored before transportation.

The *varOPEX* represents all specific energy demands like electricity, natural gas, diesel or hydrogen consumption. The overall expenditures of one module (*TOTEX*) constitute the sum of these three values.

$$TOTEX = CAPEX + fixOPEX + varOPEX$$

We calculate operational CO_2 emissions to add an ecological dimension to this analysis, as well as total energy demand. Therefore, we impinge the consumed energy with specific CO_2 -emissions from Table 3 and total energy demands from Table 4. CO_2 emissions caused by the production of the technologies are not considered in this study. All monetary values are in Euros (ϵ) . If conversion from US-Dollar (USD) is required, a rate of 1.1 USD/ ϵ is assumed. Inflationary adjustments are not included.

3.1. Production module

This study assumes hydrogen production by means of water electrolysis, whereby water is split into hydrogen and oxygen.

$$H_2O \rightarrow H_2 + \frac{1}{2}O_2$$

The most promising electrolysis systems for near to mid-term applications are alkaline and polymer electrolyte membrane (PEM) electrolysis. Based on the underlying energy concept of the IEK-3 [10,41], we assumed an overall PEM electrolysis efficiency of 70% which amounts to 47.6 kWh of electricity consumption per kg of hydrogen produced. The investment costs amount to $500 \, \epsilon / \mathrm{kW}$. Further assumptions are listed on Table 5.

3.2. Storage module

The storage module includes the costs for seasonal storage and calculates the expenditures based on the required capacity and losses concerning leakage and boil-off. The above-ground tanks have no implemented scaling function and fixed specific investment costs per kg of hydrogen. The LH₂ and GH₂ tank investment costs are based on 2015 U.S. Department of Energy values [21]. LOHC storage is assumed to be $50 \epsilon/kg_{H2}$, including the LOHC-carrier material $(2.5 \epsilon/kg_{LOHC})$ with $15.2 kg_{LOHC}/kg_{H2}$ as well as two tanks $(5 \epsilon/kg_{H2})$ per tank) [2].

In contrast, the cavern capital costs are based on their volume. To combine the mass-based required capacity with the volume-based cost function we use the density difference between maximal and minimal pressure given by CoolProp [49].

While GH_2 and LOHC storage are not subject to hydrogen losses during their storage time, the boil-off of liquid hydrogen must be considered. According to the U.S. DRIVE Team [47], state of the art large-scale liquid hydrogen tanks have boil-off rates of 0.03% per day. All assumptions are shown in Table 6.

3.3. Transport module

The transport module is split into two sub-calculations, as the pipeline and trailer delivery work in different ways and each system therefore needs some adjustments. Pipelines have high investment costs, but small operational costs. The pipe's diameter is the main parameter for cost calculations and depends on the required throughput and transportation distance. To specify the required diameter, an iterative thermodynamic approach as discussed by Tietze [50] calculates the minimal diameter *D* at a given demand, distance, and maximal allowable pressure drop. The specific costs per meter of pipeline are calculated in accordance with Krieg [45]:

$$Invest\left[\frac{\epsilon}{m}\right] = Invest_A * D^2 + Invest_B * D + Invest_C$$

Table 3 Specific CO₂-emissions per energy source.^a

Electricity RES	0 g/kWh	Natural gas ^b	238 g/kWh
Electricity grid	540 g/kWh	Diesel ^c	319 g/kWh

- ^a The specific CO_2 emissions as well as the total energy consumption of an energy source are taken from the JEC well-to-wheel analysis [44]: Due to uncertainties in the future energy mix and technological neutrality, they assumed similar emission values for the electricity grid after horizon 2020+ as that of 2009.
- ^b EU-mix natural gas supply without compression and dispensing at retail site (Code CMCG1).
- ^c Typical diesel supplied in the EU (Code COD1).

Table 4 Specific total energy consumption per energy source.^a

Electricity RES	3.60 MJ/kWh	Natural gas ^b	3.96 MJ/kWh
Electricity grid	10.62 MJ/kWh	Diesel ^c	4.32 MJ/kWh

- ^a The specific CO₂ emissions as well as the total energy consumption of an energy source are taken from the JEC well-to-wheel analysis [44]: Due to uncertainties in the future energy mix and technological neutrality, they assumed similar emission values for the electricity grid after horizon 2020+ as that of 2009.
- ^b EU-mix natural gas supply without compression and dispensing at retail site (Code CMCG1).
 - ^c Typical diesel supplied in the EU (Code COD1).

Table 5 Electrolysis assumptions.

Pressure Out	30 bar	Invest	500 €/kW
Water Demand	$0.01 \text{ m}^3/\text{kg}$	Depreciation Period	10 years
Electricity Demand	47.6 kWh/kg	OM	3%

Values derived from Robinius [41], Krieg [45] and Noack et al. [46].

Truck trailer delivery has a limited batch capacity and does not benefit from increasing hydrogen demand. Thus, the transportation costs depend on distance only. Similarly to Teichmann [2,26], the annualized investment costs for the truck and trailer are converted into hourly values. The specific cost of transportation can be calculated based on the driving speed, the time for loading/unloading the batch, and the fuel demand of the truck.

Today, GH₂ trailers are available as composite or steel tubes. While the composite material allows for higher storage capacities of up to 1150 kg per trailer at high capital costs above 1 mil USD, while the steel tubes require less investment but have less capacity [51]. LH₂ trailers are well established in the industry, with payloads of between 4000 and 4500 kg per trailer [21,47]. LOHC trailer could be utilized in the form of common steel tanks already used for diesel and gasoline delivery. Their capacity is limited by the wt.% of hydrogen in its loaded state. As can be seen in Table 7, we considered a net capacity of 680 kg for a compressed hydrogen composite trailer, 4300 kg for liquid hydrogen and 1800 kg of stored hydrogen for an LOHC. The loading and unloading operations as well as the trailer costs drawn from Elgowainy [18] and Teichmann [2]. Following Reddi and Elgowainy [18,51-53], we assume the GH2 trailer to remain at the fuelling station instead of being transferred to a low-pressure storage.

The difficulty in comparing pipeline and trucks is their different types of delivery. While trucks are flexible in endpoints and routes, a point-to-point consideration is reasonable between fuelling stations by computing an averaged distance. Conversely, pipelines are networks, wherein many different fuelling stations have access to the same pipeline sections, while some do not and require additional infrastructure. The system is therefore often divided into a transmission and a distribution grid [54]. While the transmission grid costs are split across the whole system of consumers, the distribution is tailored to each fuelling station. Robinius and Krieg [45] calculate the costs of a pipeline network to cover Germany.

Table 6 Storage module assumptions.

	Cavern ^a GH ₂	Tank			
		GH ₂ [21,47]	LH2 [21,47]	LOHC [2]	
Storage Pressure	60–150 bar	15-250 bar	1 bar	1 bar	
Invest _{Base}	81 M€	500 €	25 €	50 €	
Invest _{Compare}	500,000 m ³	1 kg	1 kg	1 kg	
Invest _{Scale}	0.28	1	1	1	
Depreciation Period	30 year	20 year	20 year	20 year	
Losses	0%/day	0%/day	0.03%/day	0%/day	
OM	2%	2%	2%	2%	

^a Assumptions in this study, derived from Acht [48].

Table 7 Transport module assumptions.

GH ₂ -Pipeline [45,50]		Truck [37]	
Pressure _{In} Pressure _{Out} Invest _A Invest _B Invest _C Depreciation Period OM Distribution Distance	100 bar 70 bar 0.0022 €/mm ² 0.86 €/mm 247.5 € 40 year 4% 3 km	Invest Depreciation Period Utilization OMfix Diesel Demand Speed ^a	160,000 € 8 year 2000 h/year 12% 35 l/100 km 50 km/h
Trailer	GH ₂ [21,51]	LH ₂ [21,47]	LOHC [2,55]
Invest Depreciation Period Utilization OM Payload Net capacity Loading time	550,000 € 12 year 2000 h/year 2% 720 kg 670 kg 1.5 h	860,000 € 12 year 2000 h/year 2% 4500 kg 4300 kg 3 h	150,000 € 12 year 2000 h/year 2% 1800 kg 1800 kg 1.5 h

^a The driving speed of Trucks is varying in different studies between 35 km/h [26] and 70 km/h [18] depending on rural or urban streets. This study doesn't differentiate between rural and urban streets and assumes a value of 50 km/h.

The average distribution length normalized against the quantity of fuelling stations was 3 km of additional distribution pipeline per station. We assume this value for this study. Especially for small distances, we will overestimate the costs in this way.

Liquid hydrogen boil-off during delivery does not correspond to the transport module, but will be considered at the station.

3.4. Fuelling module

The fuelling module maps all expenditures at the fuelling station, requiring a low-pressure storage system, conversion from the storage medium to ${\rm GH_2}$, compression to 900 bar fuelling pressure, 3 high pressure ${\rm GH_2}$ storage, and precooling to $-40\,^{\circ}{\rm C}$ to enable fast refuelling. Table 8 presents all assumptions for this module. The design capacity of 850 kg/day is drawn from previous studies at the IEK-3 for building a hydrogen refuelling network in Germany [41]. Currently, fuelling stations have capacities of up to 200 kg/day, while only serving about 2–3 vehicles per day. Thus, the assumed capacity in combination with the utilization rate of 70% represents refuelling within a further-developed hydrogen mobility.

We assume investment costs for pipeline fuelling stations of 2 million \in [41]. A calculation for GH₂ or LH₂ trailer-supplied fuelling stations with the HRSAM [56] yields a value of nearly 1.7 million \in . Nevertheless, these values are just rough assumptions, as the costs for different stations can vary a lot due to different site

locations, station designs and component cost reduction potentials. Elgowainy and Reddi [53] claim that the GH₂ tube trailer should remain at the fuelling station as low pressure storage instead of decanting the hydrogen for lower fuelling costs. Therefore, each tube-trailer-supplied fuelling station must invest in its own trailer (Table 7). The electricity demand of fuelling stations consists of compression/pump energy and precooling with the assumptions based on the 2015 DOE values [21].

LOHC fuelling stations are not available today and a demonstration of the combination of a dehydrogenation unit and a 700 bar refuelling compressor has yet to be conducted. The investment costs are therefore assumed to be 2 million €, like the pipeline fuelling station. Actual dehydrogenation units from Hydrogenious have operating pressures of 1–2 bar, but values up to 5 bar are feasible. The electricity consumption for the compression from 5 to 900 bar is assumed as 4.4 kWh/kg, wherein 4.0 kWh/kg are accountable to the compression energy demand.⁴ and 0.4 kWh/kg to precooling [23]. Linde [57] claims that at the same pressure difference its Ionic Compressor has an energy consumption of 2.7 kWh/kg, but with a lack of investment data the more conservative value is taken into account.

To supply the heat demand, different approaches are implemented in this study: Natural gas, electricity, hydrogen, and diesel. We implemented all options without regard to different investment costs for different supply systems. The fuels merely differ in their total costs and emissions to qualitatively estimate the potential benefits of different fuels.

The losses of hydrogen at the fuelling stations have different origins. The loss caused by the compression of hydrogen amounts to some 0.5% due to leakage [19,21]. Liquid hydrogen supply is thereby more difficult to estimate, since hydrogen losses can be induced by boil-off as well as by the procedure itself. According to the 2008 Nexant Report [19], boil-off during the trailer delivery is regulated to zero, but the boiled hydrogen is lost during the unloading process and accounts for 6% of the losses at the fuelling station. An additional compressor could recover the lost hydrogen, but the recovery process would be too cost-intensive. Similar assumptions were advanced by the US DRIVE Partnership [47] in 2013, with a value of up to 5% hydrogen loss per unloading operation. Teichmann [26] considers delivery losses at only 0.5% per truck. The HDSAM-Model [18] in its 2015 version also presented a default value for the unloading losses of 0%. In addition to the unloading process, the liquid storage tank causes boil-off at the station. The assumptions range from 0.25%/day [19] up to 0.5%/day [47]. Altogether, values of between 0% and 6% seem reasonable and as such we chose 3% losses for the fuelling station.

³ The pressure in the high pressure cascade must be between 875 bar and 950 bar based on the SAE J2601 worldwide hydrogen refuelling standard for 700 bar refuelling. In the case of lower vehicle tank pressures, e.g., for 350 bar fuel cell bus refuelling, the energy demand for the compression decreases, as well as precooling no longer being necessary.

⁴ The compression energy demand of 4.0 kWh/kg is calculated as a four stage compressor with an isentropic efficiency of 65%, which is the average compressor efficiency according to NREL [16]. The inlet temperature is set to 25 °C. After each stage, an intercooler cools the hydrogen to 40 °C. The thermodynamic properties of hydrogen are taken from the CoolProp Library [49]. The ESI contains the detailed calculation

Table 8 Assumptions for the considered fuelling modules.

Supply	Pipeline	Trailer			
	GH ₂ [18,21]	GH ₂ [18,21,53]	LH ₂ [18,21]	LOHC [2]	
Design Capacity	850 kg/day	850 kg/day	850 kg/day	850 kg/day	
Invest	2 M€	1.7 M€	1.7 M€	2 M€	
Depreciation Period	10 year	10 year	10 year	10 year	
OM	10%	10%	10%	10%	
Electricity Demand	2.0 kWh/kg ^a	1.9 kWh/kg ^a	0.6 kWh/kg	4.4 kWh/kg	
Heat Demand	0 kWh/kg	0 kWh/kg	0 kWh/kg	9 kWh/kg	
Losses	0.5%	0.5%	3%	0.5%	

^a Based on 2015 specific energy demand for compression of 1.6 kWh/kg (1.5 kWh/kg) for 100 bar pipeline (250 bar tube trailer) supply from DOE [21] plus additional 0.4 kWh/kg precooling demand [23].

3.5. Conversion module

The conversion module connects the production, storage and transport module and represents conversion between different storage methods. Since LH₂ and LOHC-storage are both highly energy intensive, a combination of these technologies, such as seasonal storage in LOHC and transport via LH₂, is not considered. Nevertheless, we consider seven conversion technologies. Based on a mass-based throughput capacity, capital expenditures are calculated with assumptions from Table 9. Only the compressor energy demand and power capacity depend on suction and outlet pressure as well as the compressor throughput. The calculation is similar to the one used for a refuelling station compressor, and uses an isentropic compressor efficiency and real gas properties given by CoolProp [49].

The 2008 Nexant Report [19] demonstrates that the efficiency of a liquefaction plant improves with higher plant capacities. The IdealHy-study [58] in particular claimed an energy demand of 6.78 kWh/kg at a plant capacity of 50 tons of hydrogen per day. For simplification, we set this value as fixed even though plants of these scales and efficiencies do not exist today. Besides the energy efficiency, the investment costs for the liquefaction plant are high. The past decade has seen an adjustment of optimistic assumptions on large-scale liquefaction plant costs. Fig. 4 shows that from 2005 to 2015 there was a steady increase in capital cost assumptions. However, the latest investigations were more conservative on this and as such the IdealHy cost function is utilized in this study.

The assumptions for hydrogenation and dehydrogenation are derived from Teichmann [2] and McClain [55] and customized for the investigation area up to hydrogen throughputs of 300 t/day. The heat supply used for centralized dehydrogenation units in this context is natural gas.

The liquefaction and hydrogenation plant assumptions are based on an inlet pressure of 30 bar. Since the electrolyzer outlet pressure is set to 30 bar and the cavern outlet is 70 bar, we do not include an additional compressor unit for the conversion to LH_2 and LOHC. In contrast, the outlet of the dehydrogenation units requires additional compression.

3.6. Model overview

Finally, the established model as shown in Fig. 3 can calculate hydrogen supply chains from production to refuelling with special attention on seasonal storage and transportation as a well-to-tank analysis. For this study, we considered electrolysis exclusively as the production method. Thus, each pathway is defined by its associated storage and transportation method. Only LOHC supplied fuelling stations require additional information about their heat supply (natural gas, hydrogen, electricity, and diesel). This accounts for 22 theoretical pathways as shown in Fig. 5.

4. Results and discussion

To reduce the number of investigated pathways, we first consider pathways that have the same storage method for seasonal storage and transportation (blue lines in Fig. 5). In a second analysis, we limit the possible pathways to competitive technologies and expand the investigation with different storage and transportation methods (grey lines in Fig. 5). Thereby, we use natural gas as the heat source for the dehydrogenation process at the fuelling station.

A sensitivity analysis of the main pathways reveals the most sensitive assumptions of the model. A well-to-wheel analysis determines the ecological impact and efficiency of FCEVs compared to conventional fuel technologies and integrates our results with the 2014 well-to-wheel analysis from the Joint Research Centre, EUCAR and CONCAWE (JEC) [44]. The last step will investigate different heat sources for this dehydrogenation to analyse their economic and ecological impact. The investigated hydrogen demand ranged from 0.4 to 100 t/day and the transport distance from 2 to 500 km. For a detailed analysis and direct comparison between different pathways, a transport distance of 250 km at 50 t/day demand is considered.

4.1. No conversion between storage and transport

Fig. 6 shows the results of the cheapest pathway costs considering those involving the same storage and transportation method similar to the illustration by Yang and Ogden [22]. The LOHC pathway with seasonal storage and transportation by LOHC is found to be the cheapest supply chain for small to medium-scale hydrogen demands, independent of transportation distance. With increasing hydrogen demand, GH_2 tube trailer delivery in combination with underground storage in salt cavern is the most cost-efficient supply chain for medium distances of up to 200 km. For larger distances, the pipeline/cavern combination is optimal.

Comparing the systematic scaling of Table 6 with these results, we see no scaling in the LOHC storage assumptions (fixed value of $50 \, \epsilon/\text{kg}$), but strong scaling in cavern investment costs (scaling factor 0.27). Thus, rising hydrogen demand leads to cheaper costs for the cavern systems compared to LOHC storage. For a comparison of the specific costs of each pathway, Fig. 7 show the total expenditures of each module at 250 km and 50 t/day.

An initial view confirms that the seasonal storage of hydrogen in GH_2 tanks is not competitive with other storage methods. The costs for cavern, liquid hydrogen and LOHC pathways are in the range of 8-10~€/kg for the whole supply chain, while just the storage in GH_2 tanks amounts to 10.01~€/kg due to the huge capacity of seasonal storage. Therefore, further investigation does not consider seasonal storage of hydrogen in pressurized tanks.

Another aspect of the results from Fig. 7 is the different contributions of station costs compared to storage and transportation

 Table 9

 Assumptions for the considered conversion modules.

	Compressor [18]	Liquefaction [58]	Evaporation [19]	Hydrogenation [2,26,55]	Dehydrogenation [2,26,55]	LH2Pump [19]	LOHCPump [59]
State _{In}	GH ₂	GH_2	LH ₂	GH_2	LOHC	LH ₂	LOHC
State _{Out}	GH_2	LH_2	GH_2	LOHC	GH_2	LH_2	LOHC
Invest _{base}	3.9 k€	105 M€	6 k€	40 M€	30 M€	30 €	50 €
Invest _{compare}	1 kW	50 t/day	1 t/day	300 t/day	300 t/day	1 kg/day	1 t/day
Invest _{scale}	0.8335	0.66	1	0.6	0.6	1	1
Depreciation Period	15 a	20 a	10 a	20 a	20 a	10 a	10 a
OM	4%/a	8%/a	3%/a	3%/a	3%/a	3%/a	3%/a
Electricity Demand	Variable	6.78 kWh/kg	0.6 kWh/kg	0.37 kWh/kg	0.37 kWh/kg	0.1 kWh/kg	0.1 kWh/kg
Heat Demand	0 kWh/kg	0 kWh/kg	0 kWh/kg	−9 kWh/kg ^a	9 kWh/kg	0 kWh/kg	0 kWh/kg
Losses	0.5%	1.65%	0%	3%	1%	0%	0%

^a Theoretically the hydrogenation reaction supplies 8.9 kWh of heat per kg of hydrogen at 180 °C, but selling this heat is not considered in this study; GH₂ = gaseous hydrogen; LH₂ = liquid hydrogen; LOHC = liquid organic hydrogen carrier.

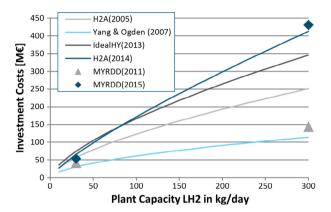


Fig. 4. Liquefaction capital cost assumptions from 2005 to 2015; Sources: H2A [18,19], Yang and Ogden [22], IdealHy [58], MYRDD [21].

costs. The specific production of hydrogen remains the same for all systems $(3.7\,\epsilon/kg)$. While LH₂ and GH₂ trailer-supplied pathways have less station expenditures $(2.27\,\epsilon/kg)$ than LOHC stations $(3.34\,\epsilon/kg)$, the cumulative costs for storage, transportation and the respective conversion are the cheapest for LOHC systems $(1.88\,\epsilon/kg)$. The liquefaction plant accounts for $1.89\,\epsilon/kg$, even under optimistic assumptions of an energy consumption of $6.78\,kWh/kg$.

4.2. Conversion between the storage and transportation modules

By excluding the GH₂ tank for seasonal storage and including additional pathways with different storage and transport methods, 10 pathways are available to analyse. In Fig. 8 it is obvious that the pathways without conversion between storage and transportation still dominate the investigated regime. However, the figure also includes two additional regimes compared to Fig. 6: For small hydrogen demands of below 20 t/day and distances of up to 100 km, the cheapest total pathway is found to be a seasonal LOHC storage system combined with tube trailer delivery.

Furthermore, a combination of LOHC delivery and seasonal cavern storage is the most cost-efficient supply chain at medium hydrogen demands of between 40 and 60 t/day and high distances above 300 km. Once again, liquid hydrogen does not play an important role in the investigated area. Fig. 9 shows the cost comparison at 250 km and 50 t/day. Seasonal storage with liquid hydrogen is the most cost-intensive option in all instances. Considering evaporation, liquefaction for storage-only is arguably less reasonable by comparison to LOHC-related pathways. Moreover, the liquefaction process by itself has higher expenditures than accumulated hydrogenation and LOHC-storage. The liquefaction of hydrogen after seasonal cavern storage is more expensive (1.98 ϵ /kg) than before seasonal storage (1.89 ϵ /kg) due to the rising electricity price for permanent operation compared to the flexible operation with renewable energies.

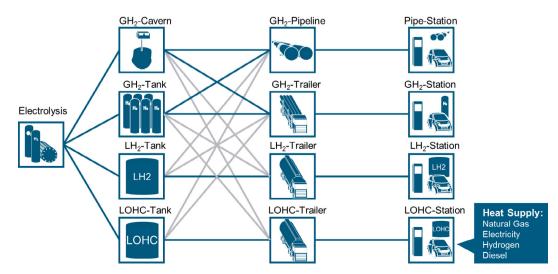


Fig. 5. Hydrogen pathways implemented in current study. Blue lines: pathways without conversion between storage and transport; grey lines: pathways with conversion between storage and transport. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

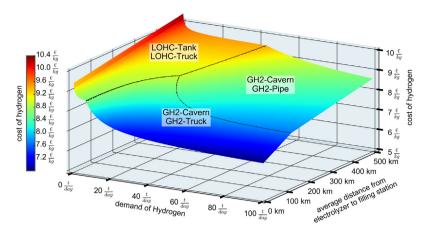


Fig. 6. Hydrogen cost at the fuelling station regarding the full supply chain (Electrolysis, seasonal storage, transportation, fuelling station) for transport = storage.

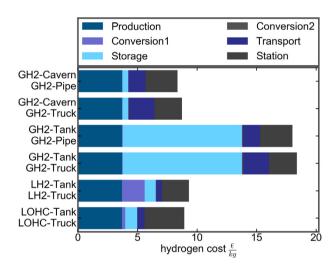


Fig. 7. Hydrogen costs for pathways without conversion between storage and transportation modules at 250 km distance and 50 t/day hydrogen demand.

The overall energy consumption per pathway is shown in Fig. 10 and is divided by source of energy. The pure GH_2 pathways require the least energy, between 51 kWh/kg (pipeline) and 54 kWh/kg (GH_2 -Truck), due to the small energy-conversion consumption. This amounts to an energy efficiency for the pipeline system of 64.8% based on the lower heating value (LHV) of hydrogen. Pathways including liquid hydrogen have cumulative energy

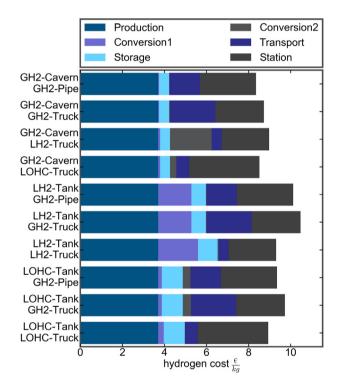


Fig. 9. Cost comparison of pathways without seasonal GH_2 -Tanks at 250 km distance and 50 t/day hydrogen demand.

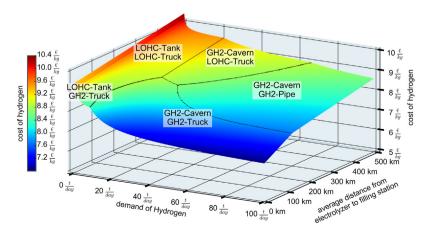


Fig. 8. Hydrogen cost at the fuelling station regarding the full supply chain (Electrolysis, seasonal storage, transport, fuelling station).

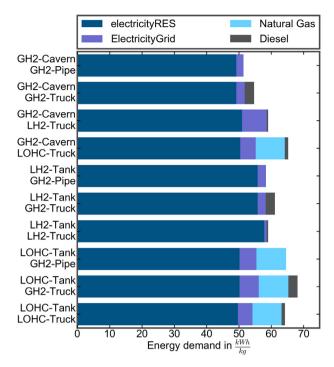


Fig. 10. Energy consumption of pathways without seasonal GH2-tanks at 250 km and 50 t/day hydrogen demand.

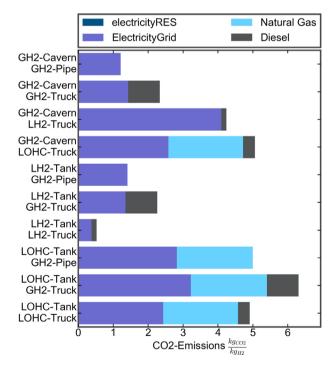


Fig. 11. Emission comparison between pathways without seasonal GH2-tanks at $250 \ \text{km}$ and $50 \ \text{t/day}$ hydrogen demand.

demands ranging between 58 and 61 kWh/kg and efficiencies between 55.1% and 57.3%.

The liquefaction process is more energy intensive than the pure compression pathways, but the liquid compression saves energy compared to gaseous compression. The LOHC pathways are then the most energy-intensive pathways, with an energy consumption up to 67.6 kWh/kg (efficiency: 49.3%). The heat produced during the hydrogenation process is thereby not included. Specifically,

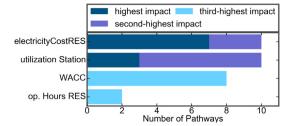


Fig. 12. Variables with the largest impact on hydrogen cost during sensitivity analysis of ±20%; for a detailed sensitivity analysis, see the supplementary information.

these pathways have a high energy demand for the dehydrogenation process and, furthermore, require compression up to 900 bar before fuelling. Comparing these results with Fig. 9, we can conclude that for small hydrogen demands the investment cost for highly efficient pathways are still high and cheaper pathways with LOHCs are more cost-efficient, even though they consume the most energy. For higher demands, the GH_2 pathways (GH_2 trucks for short distances and GH_2 pipelines for long distances) require smaller capital expenditures due to scaling effects and consequently the energy efficiency has more impact. Medium efficient pathways with liquid hydrogen are not preferred in either case.

Based on the energy consumption we determined the operational CO_2 emissions in Fig. 11. The LOHC pathways have the highest ecological impact, with CO_2 emissions up to 6.29 kg CO_2 per kg hydrogen. The pathway with the lowest CO_2 emissions is the pure liquid hydrogen path $(0.52~\mathrm{kg}_{\mathrm{CO}2}/\mathrm{kg}_{\mathrm{H2}})$, since it primarily requires renewable energy. In contrast, the liquefaction of hydrogen after seasonal cavern storage offers almost no benefit to the LOHC pathways since, with a continuous electricity demand; the liquefaction at this stage offers no grid service and does not acquire renewable energy for the liquefaction. The price of electricity is thereby higher, but the capital expenditure decreases due to more operational hours. As a result, this pathway is slightly less expensive as the pathway with "renewable" liquefaction and seasonal LH₂-storage.

4.3. Sensitivity analysis

The general input values may have a huge impact on the overall cost, even for small changes. To analyse the sensitivity of the model, we changed the values from Table 1 by ±20% and observed how the model reacted. Fig. 12 shows the variables with the highest impact within this analysis. The two most sensitive variables are the price of renewable electricity and the utilization of the station. While the renewable energy costs are the primary OPEX for production and the real source of hydrogen, the utilization of the station symbolizes the significant capital investment for the station. With increasing utilization, the CAPEX of the station substantially decreases. The third-highest values are the WACC and operating hours. The WACC represents the general investmentintensive system over the whole supply chain. The operating hours are only of special importance in two cases: seasonal storage with liquid hydrogen and GH₂ or LH₂ truck transportation. Thereby the high capital costs for the liquefaction plant can be reduced due to higher plant utilization.

4.4. Well-to-wheel analysis

To classify the results of this study with the existing literature, we expand the well-to-tank (WTT) analysis to a well-to-wheel (WTW) analysis. Therefore, we multiply the fuel consumption of FCEVs by WTT emissions, since the FCEV by itself has no further emissions. The JEC WTW analysis serves as a basis for comparison

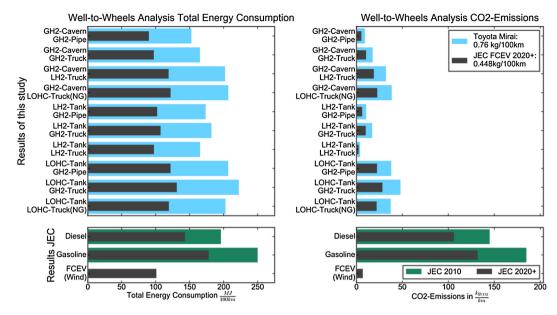


Fig. 13. Well to wheel analysis in comparison to JEC results from 2014 [44]; Notes: Own pathways at 250 km distance and 50 t/day hydrogen demand.

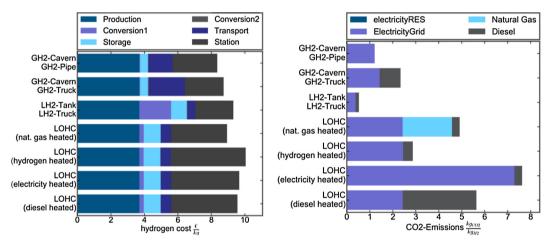


Fig. 14. Comparison of different heat sources for LOHC stations at 250 km and 50 t/day hydrogen demand (left side: cost comparison; right side: emission comparison).

in this study. They split their results into two sections: the total energy consumption in MJ/100 km and the GHG emissions in kilograms $\rm CO_2$ equivalent per km. In their study, they evaluate the hydrogen fuel consumption for the FCEV of 0.448 kg per 100 km for a horizon of 2020+ and 0.624 kg for 2010. Compared to today's Toyota Mirai, with a consumption of 0.76 kg/100 km [60], these values seem very low.

Fig. 13 shows the results of our WTW analysis (upper chart) in comparison to the JEC results for FCEV (wind-powered electrolysis with pipeline transmission) and conventional diesel and gasoline engines (lower chart). Like Fig. 10, the LOHC pathways feature the highest total energy consumption; nevertheless, the total energy consumption of all investigated pathways is smaller than that of conventional fuels. Regarding the right side of Fig. 13, all investigated pathways have less than 30% CO₂ emissions compared to conventional diesel; even the emission-intensive LOHC pathways. Comparing the cavern/pipeline path with the JEC FCEV result, we get slightly higher emissions, but a smaller total energy demand.

4.5. LOHC station comparison

The last aspect of this study investigates the dehydrogenation process of the LOHC-supplied fuelling station. Fig. 14 shows the

economic impact of different heat sources compared to the pure GH_2 and LH_2 pathways. Natural gas as a heat source is cheaper than liquid hydrogen and has the smallest cost of all investigated heat sources at $8.98~\rm e/kg$. A self-sufficient plant which burns a portion of the stored hydrogen is not competitive, with costs of $10.2~\rm e/kg$. Meanwhile, regarding the ecological impact indicated in Fig. 14 the hydrogen burner offers the lowest CO_2 emission of all heat sources with $2.9~\rm kg$ CO_2 per kg of hydrogen. The emissions from natural gas have the second lowest emissions due to the cleaner natural gas combustion compared to diesel. Electricity as a heat source has the highest impact on CO_2 emissions since we assumed the current grid mix.

Theoretically, there are many further options besides those analysed in this work. Biogas or biodiesel, for example, could displace gas and diesel as heat supply options, likely resulting in an improved ecological performance of LOHC pathways. In the case of a highly renewable grid, even the energy consumption for the pre-fuelling compression to 900 bar could be satisfied ecologically. Nevertheless, the total energy consumption will remain at a high level. To target this issue, the system components themselves must be reviewed. As already mentioned, the Linde ionic compressor claims to have achieved a higher efficiency, consuming just 2.7 kWh/kg, which accounts for energy saving of 1.3 kWh/kg (30% of total energy

demand for compression). Furthermore, the refuelling station needs heat as well as electricity. Thus, an optimized combined heat and power plant could operate with less primary energy consumption would be an interesting target for future investigation.

4.6. Summary of results

The results show that underground storage solutions and LOHCs offer an economic solution to the storage of large amounts of hydrogen at low charge cycles. In particular for regions without access to underground storage facilities, the LOHC technology presents itself as a reasonable alternative. The liquefaction of hydrogen is not found to offer any advantage in the analysed context. While seasonal storage is expensive due to the huge capital investments required, a continuous liquefaction plant entails higher electricity costs and CO₂ emissions. According to our results, liquid hydrogen could only be cost-efficient for long distance transportation of over 500 km. However, at these distances other relevant transportation methods, like ships or trains, were not considered here and so this application area will need to be addressed in future studies.

It is also apparent that LOHCs have an interesting economic impact on supplying hydrogen for the mobility sector in the context of seasonal storage and hydrogen transportation. These pathways can offer the cheapest storage and transportation option, especially in instances of low hydrogen demand. This is particularly the case when it is not economically feasible to store hydrogen in large-scale underground facilities. Nevertheless, pathways utilizing LOHCs as a storage or transportation medium correspond to the highest energy demands due to the low pressure of dehydrogenation. As hydrogen demand increases, gaseous pathways benefit from scaling due to the lower energy demand, in addition to benefitting from the usual economy of scale dynamics of pipelines and underground storage options.

On the basis of the sensitivity analysis, it is obvious that the most important factors are the price of renewable electricity and the utilization rate of the station. Regarding the price of electricity, locations with major potential for cheap renewable energy production should be further investigated. Nevertheless, this topic is limited to the technical potential for renewables within a country. Regarding the utilization rate, the building of new refuelling stations necessitates diligent planning to maximize the utilization.

The well-to-wheel analysis clarified that the outlined results are well-suited to similar approaches from the JEC well-to-wheel analysis and all investigated pathways offer significant advantages compared to conventional fuels regarding GHG emissions.

The analysis of different heat sources for the dehydrogenation of LOHC-supplied fuelling stations shows that dehydrogenation heat sources other than natural gas are more expensive and offer no advantage on overall emissions. Nevertheless, for the realistic setup of LOHC stations, the dehydrogenation heat source should be the focus of a more detailed analysis.

5. Conclusion

We developed a model for calculating the costs, energy consumption and GHG emission for supplying hydrogen to FCEVs that we present as a well-to-tank analysis. Instead of focussing on single supply chain parts like storage or transportation, we considered the full supply chain so as to obtain a holistic view of the advantages and drawbacks of different technologies. The established model gives a strong overview of relevant infrastructure technologies and combinations from ecological and economic aspects. The evaluation of these technologies and their application area is an essential step for implementing them in highly spatial and

temporal resolved energy system models. Due to the easy implementation of alternative infrastructure technologies besides the state of the art of compression and liquefaction, the potential of future disruptive technologies – like LOHC – can be estimated quickly. The results indicate that the LOHC technology is very promising for future hydrogen supply chains from an economic point of view, even if there remain questions to be resolved, such as the final system design of the LOHC-supplied fuelling station or the heat source for dehydrogenation.

Furthermore, seasonal fluctuations of renewable power generation were considered as additional storage demand. We showed that seasonal storage may have a high economic impact, as well as on GHG emissions, especially with regard to the liquefaction of hydrogen.

Acknowledgements

This work was supported by the Helmholtz Association under the Joint Initiative "EnergySystem 2050 – A Contribution of the Research Field Energy". The authors furthermore gratefully acknowledge funding by the German Federal Ministry of Education and Research (BMBF) within the Kopernikus Project P2X: Flexible use of renewable resources – exploration, validation and implementation of 'Power-to-X' concepts.

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.apenergy.2017. 05.050.

References

- UN. Adoption of the Paris agreement. In: Framework convention on climate change. United Nations: Paris; 2015.
- [2] Teichmann D. Konzeption und Bewertung einer nachhaltigen Energieversorgung auf Basis flüssiger Wasserstoffträger (LOHC). In: Berichte aus der Energietechnik. Erlangen-Nürnberg: Aachen; 2015. p. XIV, 190 Seiten.
- [3] BMWi and BMU. Energiekonzept für eine umweltschonende, zuverlässige und bezahlbare Energieversorgung. Bundeministerium für Wirtschaft und Technologie (BMWi); 2010.
- [4] BMWi. EEG in Zahlen; 2015.
- [5] Adamek F et al. Energiespeicher für die Energiewende Speicherungsbedarf und Auswirkungen auf das Übertragungsnetz für Szenarien bis 2050, 2012.
- [6] Schill W-P. Residual load, renewable surplus generation and storage requirements in Germany. Energy Policy 2014;73:65–79. http://dx.doi.org/10.1016/j.enpol.2014.05.032.
- [7] AgEE-Stat. Entwicklung der Erneuerbaren Energien in Deutschland im Jahr 2015; 2016.
- [8] Minkos A et al. Luftqualität 2015 Vorläufige Auswertung, 2016.
- [9] McKinsey. A portfolio of power-trains for Europa: a fact-based analysis; 2010.
- [10] Schiebahn S et al. Power to gas: technological overview, systems analysis and economic assessment for a case study in Germany. Int J Hydrogen Energy 2015;40(12):4285–94. http://dx.doi.org/10.1016/j.iihydene.2015.01.123.
- [11] Ausfelder F et al. Energiespeicherung als Element einer sicheren Energieversorgung. Chem Ing Tec 2015;87(1–2):17–89. http://dx.doi.org/10.1002/cite.201400183.
- [12] Léon A. Hydrogen technology. In: Green energy and technology. Berlin Heidelberg: Springer-Verlag; 2008.
- [13] Guandalinia G, Robiniusb M, Grubeb T, Campanaria S, Stoltenb D. Long-term power-to-gas potential from wind and solar power: A country analysis for Italy. Int J Hydrogen Energy 2017. http://dx.doi.org/10.1016/j.ijhydene.2017.03.081. in press.
- [14] Edwards R et al. Well-to-Tank Report Version 4.a. In: Godwin S et al., editors. JEC Well-to-Wheels Analysis. Joint Research Centre; 2014. p. 148.
- [15] Chen T-P. Hydrogen delivey infrastructure option analysis. Nexant; 2010.
- [16] Remick R. Hydrogen station compression, storage, and dispensing technical status and costs: systems integration, 2014.
- [17] Dodds PM, Will. A review of hydrogen delivery technologies for energy system models. University College London: UCL Energy Institute; 2012.
- [18] Reddi K et al. Hydrogen delivery scenario analysis model. Department of Energy USA; 2015.
- [19] Nexant. H2A hydrogen delivery infrastructure analysis models and conventional pathway options analysis results; 2008.

- [20] Cipriani G et al. Perspective on hydrogen energy carrier and its automotive applications. Int J Hydrogen Energy 2014;39(16):8482–94. http://dx.doi.org/10.1016/i.jihydene.2014.03.174.
- [21] DOE. Multi-Year research, development, and demonstration plan 3.2 hydrogen delivery. Available from: http://energy.gov/eere/fuelcells/downloads/fuel-cell-technologies-office-multi-year-research-development-and-22; 2015 August [08.04.2016].
- [22] Yang C, Ogden J. Determining the lowest-cost hydrogen delivery mode. Int J Hydrogen Energy 2007;32(2):268–86. http://dx.doi.org/10.1016/j.ijhydene. 2006.05.009.
- [23] DOE. DOE hydrogen and fuel cells programm annual progress report 2015; 2015.
- [24] Gardiner M. Energy requirements for hydrogen gas compression and liquefaction as related to vehicle storage needs. In: Satyapal S, editor. DOE hydrogen and fuel cells program record.
- [25] Niaz S, Manzoor T, Pandith AH. Hydrogen storage: materials, methods and perspectives. Renew Sustain Energy Rev 2015;50:457–69. http://dx.doi.org/10.1016/j.rser.2015.05.011.
- [26] Teichmann D, Arlt W, Wasserscheid P. Liquid Organic Hydrogen Carriers as an efficient vector for the transport and storage of renewable energy. Int J Hydrogen Energy 2012;37(23):18118–32. http://dx.doi.org/10.1016/j.iihydene.2012.08.066.
- [27] Teichmann D et al. A future energy supply based on Liquid Organic Hydrogen Carriers (LOHC). Energy Environ Sci 2011;4(8):2767. http://dx.doi.org/10.1039/c1ee01454d
- [28] Dagdougui H. Models, methods and approaches for the planning and design of the future hydrogen supply chain. Int J Hydrogen Energy 2012;37(6):5318–27. http://dx.doi.org/10.1016/j.ijhydene.2011.08.041.
- [29] Samsatii S. Optimal design and operation of integrated wind-hydrogenelectricity networks for decarbonising the domestic transport sector in Great Britain. Int J Hydrogen Energy 2016;41(1):447–921.
- [30] Klell M. Storage of hydrogen in the pure form. In: Handbook of hydrogen storage. Wiley-VCH Verlag GmbH & Co. KGaA; 2010. p. 1–37.
- [31] Di Profio P et al. Comparison of hydrogen hydrates with existing hydrogen storage technologies: energetic and economic evaluations. Int J Hydrogen Energy 2009;34(22):9173–80. http://dx.doi.org/10.1016/j.ijhydene. 2009.09.056
- [32] Sartbaeva A et al. Hydrogen nexus in a sustainable energy future. Energy Environ Sci 2008;1(1):79. http://dx.doi.org/10.1039/b810104n.
- [33] He T, Pei Q, Chen P. Liquid organic hydrogen carriers. J Energy Chem 2015;24 (5):587–94. http://dx.doi.org/10.1016/j.jechem.2015.08.007.
- [34] Okada Y, Shimura M. Development of large-scale H2 storage and transportation technology with Liquid Organic Hydrogen Carrier (LOHC). Chiyoda; 2013.
- [35] Dalebrook AF et al. Hydrogen storage: beyond conventional methods. Chem Commun (Camb) 2013;49(78):8735–51. http://dx.doi.org/10.1039/c3cc43836h.
- [36] Krasae-in S, Stang JH, Neksa P. Development of large-scale hydrogen liquefaction processes from 1898 to 2009. Int J Hydrogen Energy 2010;35 (10):4524–33. http://dx.doi.org/10.1016/j.iihydene.2010.02.109.
- [37] Bruckner N et al. Evaluation of industrially applied heat-transfer fluids as liquid organic hydrogen carrier systems. Chemsuschem 2014;7(1):229–35. http://dx.doi.org/10.1002/cssc.201300426.
- [38] Durbin DJ, Malardier-Jugroot C. Review of hydrogen storage techniques for on board vehicle applications. Int J Hydrogen Energy 2013;38 (34):14595–617. http://dx.doi.org/10.1016/i.jihydene.2013.07.058.
- [39] Aardahl CL, Rassat SD. Overview of systems considerations for on-board chemical hydrogen storage. Int J Hydrogen Energy 2009;34(16):6676–83. http://dx.doi.org/10.1016/j.jihydene.2009.06.009.

- [40] Converse AO, Converse AO. Seasonal Energy Storage in a Renewable Energy System. Proc IEEE 2012;100(2):401–9. ISSN: 0018-9219 (print), CODEN: IEEPAD Publisher: IEEE Country of Publication: USA, 2012. 100(2): p. 401. http://dx.doi.org/10.1109/iproc.2011.2105231..
- [41] Robinius M. Strom- und Gasmarktdesign zur Versorgung des deutschen Straßenverkehrs mit Wasserstoff. In: Institut für elektrochemische Verfahrenstechnik. Aachen: RWTH Aachen; 2015.
- [42] Öko-Institut. Die Entwicklung der EEG-Kosten bis 2035; 2015, study in order of Agora Energiewende.
- [43] Eurostat. Strompreise nach Art des Benutzers. http://ec.europa.eu/eurostat/; 2016
- [44] Edwards R et al. Well-to-Wheels Report Version 4.a. In: Godwin S et al., editors. JEC well-to-wheels analysis. Joint Research Centre; 2014.
- [45] Krieg D. Konzept und Kosten eines Pipelinesystems zur Versorgung des deutschen Straßenverkehrs mit Wasserstoff. In: Forschungszentrum Jülich. RWTH Aachen; 2012.
- [46] Noack C. et al. Studie über die Planung einer Demonstrationsanlage zor Wasserstoff-Kraftstoffgewinnung durch Elektrolyse mit Zwischenspeicherung in Salzkavernen unter Druck. DLR – Deutsches Zentrum für Luft und Raumfahrt; Ludwig Bölkow Systemtechnik; Fraunhofer ISE; KBB Underground Technologies; 2015.
- [47] Barckholtz T. et al. Hydrogen delivery technical team roadmap, U.S.D. Partnership, Editor; 2013.
- [48] Acht A. Salzkavernen zur Wasserstoffspeicherung. RWTH Aachen; 2013. p. 98.
- [49] Bell IH et al. Pure and pseudo-pure fluid thermophysical property evaluation and the open-source thermophysical property library coolprop. Ind Eng Chem Res 2014;53(6):2498–508. http://dx.doi.org/10.1021/ie4033999.
- [50] Tietze V. Techno-ökonomischer Entwurf eines Wasserstoffversorgungs systems für den deutschen Straßenverkehr, 2016.
- [51] Reddi K et al. Challenges and opportunities of hydrogen delivery via pipeline, tube-trailer, LIQUID tanker and methanation-natural gas grid. In: Stolten D, Emonts B, editors. Hydrogen science and engineering: materials processes, systems and technology. Wiley; 2016. p. 849–74.
- [52] Reddi K, Elgowainy A, Sutherland E. Hydrogen refueling station compression and storage optimization with tube-trailer deliveries. Int J Hydrogen Energy 2014;39(33):19169–81. http://dx.doi.org/10.1016/j.ijhydene.2014.09.099.
- [53] Elgowainy A et al. Tube-trailer consolidation strategy for reducing hydrogen refueling station costs. Int J Hydrogen Energy 2014;39(35):20197–206. http://dx.doi.org/10.1016/j.ijhydene.2014.10.030.
- [54] Baufumé S et al. GIS-based scenario calculations for a nationwide German hydrogen pipeline infrastructure. Int J Hydrogen Energy 2013;38 (10):3813–29. http://dx.doi.org/10.1016/j.iihydene.2012.12.147.
- [55] McClain AW, Brown K, Bowen DDG. Magnesium hydride slurry: a better answer to hydrogen storage. J Energ Resour Technol 2015. http://dx.doi.org/10.1115/1.4030398.
- [56] Elgowainy A et al. Hydrogen refueling station analysis model. Argonne National Laboratory; 2015.
- [57] Mayer M. From prototype to serial production. In: A3PS Conference 2014. Vienna: Linde; 2014.
- [58] Stolzenburg K, Mubbala R. Hydrogen liquefaction report whole chain assessment. In: Integrated design for demonstration of efficient liquefaction of hydrogen (IDEALHY). FCH JU; 2013.
- [59] Loh HP, Lyons J, White CW. Process equipment cost estimation final report. DOE; 2002.
- [60] Toyota. Toyota Mirai ist verbrauchsärmstes Serien-Brennstoffzellenfahrzeug. Available from: https://www.toyota.de/news/details-2015-43.json; 2015.
- [61] Gesetz für den Ausbau erneuerbarer Energien (Erneuerbare-Energien-Gesetz -EEG 2014). Bundesministerium der Justiz und für Verbraucherschutz; 2014.