Liquid hydrogen as prospective energy carrier: A brief review and discussion of underlying assumptions applied in value chain analysis

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ABSTRACT

In the literature, different energy carriers are proposed in future long-distance hydrogen value chains. Hydrogen can be stored and transported in different forms, e.g. as compressed dense-phase hydrogen, liquefied hydrogen and in chemically bound forms as different chemical hydrides. Recently, different high-level value chain studies have made extrapolative investigations and compared such options with respect to energy efficiency and cost. Three recent journal papers overlap as the liquid hydrogen option has been considered in all three studies. The studies are not fully aligned in terms of underlying assumptions and battery limits. A comparison reveals partly vast differences in results for chain energy efficiency for long-distance liquid hydrogen transport, which are attributable to distinct differences in the set of assumptions. Our comparison pinpoints the boiloff ratio, i.e. evaporation losses due to heat ingress, in liquid hydrogen storage tanks as the main cause of the differences, and this assumption is further discussed. A review of spherical tank size and attributed boiloff ratios is presented, for existing tanks of different vintage as well as for recently proposed designs. Furthermore, the prospect for further extension of tanks size and reduction of boiloff ratio is discussed, with a complementary discussion about the use of economic assumptions in extrapolative and predictive studies. Finally, we discuss the impact of battery limits in hydrogen value chain studies and pinpoint knowledge needs and the need for a detailed bottom-up approach as a prerequisite for improving the understanding for pros and cons of the different hydrogen energy carriers.

1. Background and motivation

Recent studies of hydrogen-carrier conversion and transport options and the highly differing results and conclusions thereof, calls for a review and discussion of underlying assumptions and methodologies leading to such divergences. Three recently published papers overlap as they consider liquid hydrogen (LH_2) as a long-distance seaborne hydrogen transport option. Niermann et al. [1] have investigated hydrogen export from Algeria to Hamburg and are focussing mainly on different liquid organic hydrogen carriers (LOHC) and consider compressed pipeline transport and liquid hydrogen bulk transport, as well as electric transmission, as benchmarks. In a similar type of study Ishimoto et al. [2], hereunder the authors of this paper, have estimated the chain energy efficiency and cost for LH_2 and ammonia transport from Northern Norway in a regional (Rotterdam) and global (Tokyo) case. In a recent work by d'Amore-Domenech et al. [3], the energy balance and life-cycle cost of energy transported by subsea electrical high-voltage

cables was compared with that of pipeline hydrogen transport and ship transport of compressed or liquid hydrogen for different distances.

A full comparison of all assumptions and sensitivities thereof, choice of methodology, technical abstraction levels and details levels lies beyond the ambitions and scope of this piece. This is not only a comprehensive task but also a very difficult one as the data sets and volumes are vast, and full methodology descriptions are not necessarily fully available. We will therefore shed some light on and discuss the most obvious differences in assumptions for liquid hydrogen transport, which is the only overlapping energy carrier option for the three above-mentioned recent works. Finally, we will discuss some general methodological issues among various high-level, top-down energy chain studies seeking to predict and extrapolate scaling-up of technologies and solutions that are yet to be realised in large scale.

2. Comparison of energy efficiency results from chain studies

The study by d'Amore-Domenech et al. [3] does not report explicit

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Abbreviations

BoP	Balance of plant
BOR	Boiloff ratio
CAPEX	Capital expenditures
HHV	Higher heating value
IEA	The International Energy Agency
KHI	Kawasaki Heavy Industries Ltd.
LH_2	Liquid/liquefied hydrogen
LHV	Lower heating value
LNG	Liquefied natural gas
LOHC	Liquid organic hydrogen carriers
NASA	The National Aeronautics and Space Administration
PEM	Polymer electrolyte membrane
REFPROP	NIST Reference Fluid Thermodynamic and Transport Properties Database
TRL	Technology readiness level

results for overall chain energy efficiency from production to reception of shipment, but rather absolute values of delivered energy and energy losses over the system's defined life cycle. The chain energy efficiency can thus be approximated as the delivered energy as a fraction of the total energy input, which equals sum of delivered energy and lost energy. These values can be read from bar diagrams and for the LH_2 chain across 3000 km distance, the chain energy efficiency is so estimated to almost 69 % on a higher heating value (HHV) basis. The studies in Refs. [1,2] present the energy efficiency figures explicitly, although the battery limits cannot be fully equalized. In the study by Ishimoto et al. [2] the overall LH_2 chain energy efficiency for Rotterdam (2539 km sea distance) and Tokyo (23 407 km sea distance) is estimated to 61 % and 53 %, respectively, on a lower heating value (LHV) basis. On a HHV basis, the corresponding efficiencies are approximately 67 % and 58 %.[1] The efficiency of the 2539-km Rotterdam case (67 %) is very close to that of the 3000-km case in Ref. [3] (69 %), although battery limits, distance and other assumptions are not fully equalized. However, the fact that distances are similar and that the work by Ishimoto et al. [2] includes further processing of LH_2 upon offloading with inherent parasitic losses, indicates that the results seem rather consistent. Differences in battery limits are in general terms discussed later.

To relate the results for the two investigated distances in Ref. [2] to the definition of distance applied in Ref. [1], they must first be converted from sea distance to the defined equivalent linear distance.[2] The corresponding energy efficiencies from Ref. [1] are approximately 47 % and 21 % on a HHV basis. Although the battery limits are not equal and the estimates rely on extensive sets of underlying assumptions, the differences are still substantial. If the results from Ref. [1] are used as reference, the chain efficiencies calculated in Ref. [2] are approximately 41 % higher (1.41 x) and 178 % higher (2.78 x) than what is calculated in Ref. [1] for equivalent sea distances.

Without evaluating all input factors impacting the chain efficiency, it therefore seems clear that a main cause of these substantial deviations in sensitivity to transport distance is attributed to the ditto differences in

assumed boiloff ratio for seaborne LH_2 cargo tanks. An additional indicator thereof is that the relative differences in chain energy efficiency escalates with distance. It is therefore natural to take a closer look at this particular underlying assumption and how it differs between the studies. Ishimoto et al. [2] assume 0.2 % per day boiloff ratio (BOR) for seaborne LH_2 transport (assumed carrier capacities: 86 000 m^3 for the Rotterdam case and 172 000 m^3 for the Tokyo case) and correspondingly 0.1 % per day for onshore terminal tanks. d'Amore-Domenech et al. [3] assume 0.1 % per day (BOR) for LH_2 storage tanks [3], but do not explicitly declare the boiloff ratio on the LH_2 carrier, which is assumed to carry 180 000 m^3. Niermann et al. [1] assume 1.35 % per day BOR for LH_2 storage tanks, regardless of size. As techno-economic assumptions for LH_2 storage, Ref. [1] adopts the same set of assumptions used in Ref. [4], save for the two parameters BOR and operations-and-maintenance percentage. Instead of adopting the same BOR, 0.03 % per day [4], a 44 times higher value, 1.35 % per day, is assumed. Moreover, even though this boiloff ratio would cause vast excesses of gaseous hydrogen, far beyond what a full-scale tanker would require for propulsion, no assumption for cargo-retaining reliquefaction ancillaries has been made, and evaporated hydrogen surpluses must therefore be vented or flared directly to the atmosphere once the loading arms have been disconnected and for the full duration of the voyage. For the LH_2 ship capacity, if assumed to be 173 400 m^3, the practical implication of 1.35 % per day boiloff ratio (relative to maximum inventory) is a continuous boiloff rate and thus blowoff rate of about 164 ton per day,[3] or almost 230 MW on a LHV basis. In addition to being, in our opinion, a significant overestimation as discussed in the next section, it is for different reasons difficult to envision large-scale LH_2 transport at such boiloff rates unless high-capacity reliquefaction ancillaries are installed, drawing power from onboard gas-to-power conversion.

3. Boiloff rates from liquid hydrogen tanks – state of the art and extrapolations

The main concern regarding the high assumed boiloff ratio in Ref. [1] is that it, in our opinion, may be overestimated by potentially up to an order of magnitude based on interpolations as well as extrapolations from state-of-the-art technology. A brief review of state-of-the art LH_2 storage tank technology can provide some insights:

- Linde can provide different types of LH_2 tanks, the BOR of which depend on, inter alia, size and intended usage pattern. Examples are cylindrical tanks with 300 m^3 capacity and <0.3 % per day boiloff, and spherical tanks with 1100–2300 m^3 capacity and <0.1 % per day BOR [5].
- In the 1980s Kawasaki Heavy Industries (KHI) built spherical LH_2 tanks with 600 m^3 volume and 540 m^3 LH_2 capacity with a boiloff ratio of 0.18 % per day [6]. No degradation of performance has been detected after 30 years of operation [7]. In 2020, a 1250 m^3 cylindrical seaborne tank and a 2500 m^3 spherical onshore terminal tank have been put in operation [8], the latter with ≤0.1 % per day boiloff ratio [9]. Recently, KHI announced the completion of the basic design of an 11 200 m^3-sized spherical LH_2 tank with 10 000 m^3 storage capacity and ≤0.1 % per day boiloff ratio [10].
- NASA has operated the world's hitherto largest spherical LH_2 storage tanks (LC-39A and -B, each with approximately 3200 m^3 LH_2 capacity), built in the 1960s. The boiloff rate is reportedly 563 kg/d in absolute terms [11], which translates to a boiloff ratio of around 0.25 % per day.

Based on this brief review, it is reasonable to argue that 1.35 % per day BOR is excessive, provided that low boiloff rates can be realised also

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[1] Both hydrogen output and natural gas input are transformed from LHV basis to HHV basis.

[2] Distances used in Ref. [2] are divided by the sea-to-linear-distance factor 1.88 assumed in Ref. [1], resulting in linear-equivalent distances of 1351 km and 12 451 km, respectively. The resulting LH_2 chain energy efficiency can then be read from the chart in Fig. 8 found in [1] for these distances. While the shorter distance is directly readable in the chart, which ranges up to 10 000 km, a slight but linear extrapolation is required to estimate the long-distance chain efficiency.

[3] 164 ton per day equals roughly 50 % of the hydrogen liquefaction capacity installed worldwide as of ultimo 2019 [12].

for LH$_2$ tank sizes close to those deployed in present-day liquefied natural gas (LNG) transport. Since 0.18–0.25 % per day is already realised for tanks in the magnitudes 100s and 1000s m^3, a natural question is what further scaling-up will imply for achievable performance – whether it will facilitate or impede the realisation of lower boiloff ratios. Under the assumption of a nearly full, well-insulated tank with a uniform internal temperature distribution, the heat ingress from the surrounding environment is proportional to the difference between ambient temperature and the LH$_2$ bulk temperature [K], the tank's surface area [m^2] and the overall heat transfer coefficient [Wm^{-2}K^{-1}]:

$$\dot{Q}_{ingress} = (T_{\infty, \, ambient} - T_{\infty, \, LH2}) \, A_{surface} \, U_{overall} \tag{1}$$

The boiloff ratio or evaporation ratio, defined as the hydrogen evaporation rate [kg/s] relative to a full tank inventory [kg], has the SI unit [s^{-1}] but [% per day] is a more common unit for cryogenic storage tanks. The evaporation ratio can be expressed as:

$$BOR = \frac{(T_{\infty, \, ambient} - T_{\infty, \, LH2}) \, A_{surface} \, U_{overall}}{\rho_{LH2} \, V_{tank} \, (1 - ullage) h_{evap,LH2}} \tag{2}$$

From Eq. (2), it can be observed that the evaporation ratio is proportional to the ratio between surface area and volume, commonly referred to as the specific surface area. For a spherical tank, Fig. 1 shows how the specific surface area varies with size. As can be observed, it decreases substantially with volume, while it on the other hand escalates steeply for sufficiently low volumes.

To further illustrate what implications size may pose on the realisable design-point boiloff ratio, Fig. 2 shows estimates for the daily evaporation ratio as function of overall heat transfer coefficient and spherical tank volume. The ambient temperature is set to 288 K and the tank is for simplicity assumed to contain pure, saturated liquid parahydrogen at 1.2 bar(a) with 10 % ullage. The thermophysical properties retrieved from REFPROP [13] give a liquid density (ρ_{LH2}) and temperature ($T_{\infty,LH2}$) of 70.15 kg/m^3 and 20.86 K, respectively, and a specific evaporation heat ($h_{evap,LH2}$) of 443.17 kJ/kg. For any fixed value of $U_{overall}$, the daily evaporation ratio drops significantly with tank size and is reduced by roughly 54 % per tenfold increase of volume, e.g. for 1000 m^3 relative to 100 m^3. In addition to these equation-based correlations, rough data for three existing spherical tanks of different size and vintage [6,9,11], as well as two other indicated tank performances [5, 10], are plotted in the same diagram. The assumptions from the value chain studies in Refs. [1–3] are also included, under the assumption that [3] applies equal BOR for terminal and seaborne LH$_2$ tanks. An estimate of the overall heat transfer coefficient $U_{overall}$ for each tank can be read from the curve intersecting with the data point, and indicates roughly 0.004 Wm^{-2}K^{-1} for [5,6,9], and approximately 0.010 Wm^{-2}K^{-1} for [11]. If, theoretically, tank [5,6] or [9] could be scaled further with unchanged $U_{overall}$, the daily evaporation ratio would drop to e.g. around 0.07 % per day for 10 000 m^3 volume. The corresponding figure for [11] would end up at around 0.17 % per day.

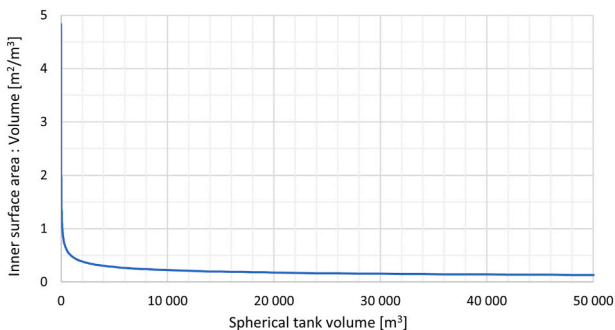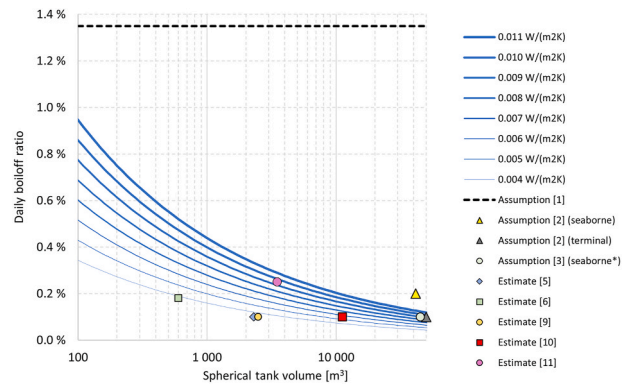As shown by these illustrative examples, size and diameter increases

Fig. 2. Estimates for the relation between spherical LH$_2$ tank volume (10 % ullage assumed), overall heat transfer coefficient and daily evaporation ratio. Ambient temperature is assumed to be 288 K.

are generally beneficial for low-pressure, insulated systems due to decreasing surface-to-volume ratio. From this perspective, relative evaporation ratios in the 0.1–0.2 % per day range may be attainable also for spherical tanks with volumes in the >10^4 m^3 magnitude. A prerequisite for this prediction is that high insulation standard and thus low overall heat transfer coefficients can be realised for increased tank sizes. This assumption is yet to be proven a posteriori and will require a gradual advancement of TRL for the tank size in consideration. Among examples of challenges faced in scaling-up are e.g. thermal contractions and scaling-up of vacuum-reliant insulation concepts, which for different reasons become increasingly challenging with size. What is the techno-economically optimal boiloff ratio for prospective large-scale LH$_2$ carriers, of sizes equal to present-day LNG tankers, remains to be identified. This is a holistic R&D task which must weigh tank designs and insulation layouts against an array of trade-offs, some of which may relax certain constraints, such as balancing evaporation rates with energy demand for propulsion and auxiliary systems as one example. Multidisciplinary competence is required on different levels, ranging from construction and materials technology, thermo- and fluid dynamics, mass and heat transfer, thermal processes, naval architecture, and power and propulsion systems. LH$_2$ containment and insulation, cargo loading and offloading, and marine hydrogen reliquefaction processes are all topics in focus in the SINTEF-led research project "LH$_2$ Pioneer – Ultra-insulated seaborne containment system for global LH$_2$ ship transport" [14].

4. Impact of techno-economic assumptions

4.1. Scaling exponents and the validity ranges thereof

The use of transparent and reasonable assumptions in techno-economic evaluations is essential to ensure proper quality and credibility of results. An example of important cost-related assumptions are scaling exponents (n) used for estimation of cost of specific equipment or process plant sections by scaling from a reference according to Eq. (3):

$$C_i = C_{ref} \left(\frac{S_i}{S_{ref}}\right)^n \tag{3}$$

where C_i is the cost of the specific equipment or process plant section, C_{ref} is the reference cost and S is the size-defining variable used to scale cost, such as flowrate, volumes, etc. When scaling costs, it is important to consider a proper scaling exponent as well as the range of which the specific variable is being scaled for. Chemical engineering handbooks [15] can give an indication on suitable scaling exponents for different type of equipment when more detailed data is unavailable. As an example, tanks are commonly scaled with a scaling exponent in the range of 0.6–0.7. Although scaling laws and proper scaling exponents

Fig. 1. Relation between inner surface area and volume for spherical tanks for the volume range 1–50 000 m^3.

can be derived based on the relation between physical properties such as surface area (due to cost of e.g. of steel or insulation) and volume (e.g. tank capacity), these equations are approximations with higher validity at capacities close to the reference point.

The reference cost should therefore generally be in the same order of magnitude as the target variable to ensure a viable estimate. Going far beyond this validity range would lead to high uncertainties on the estimates to the point that the resulting estimate may be of little value. To address this concern, studies sometimes assume no economies of scale which is another unreasonable extreme. This is, for example, the case in Ref. [1], where a LH$_2$ receiving terminal (primarily comprised of spherical tanks) with reference capacity of 30 kg/h (720 kg/d) is specified and extrapolated with a scaling exponent of 1.0 to full-scale capacities presumably at least 100 times, potentially 1000 times, that of the reference capacity. Similarly, scaling exponent issues are observed in Ref. [3] where a scaling exponent of 1.334 is assumed for spherical tanks. Although it is reasonable to assume that the steel cost of spherical tanks evolves with such an exponent, the steel cost usually represents a fraction (in the range of 30 %[4]) of the cost of a spherical tank and the other costs traditionally benefit from economies of scale. For this reason, a scaling exponent of 0.7 and a reasonable ratio between the considered and reference capacities are recommended for such tanks [16].

Another concern regarding techno-economic analyses across large capacity spans, is the risk of missing out on necessary transitions in process layout and technology selection. As one example, the technology commonly applied for a 720 kg/d hydrogen liquefaction plant is liquid nitrogen-precooled helium reverse Brayton cycles. This solution is, however, only preferrable up to 2000–3000 kg/d plant capacity [17] and has, at such small scales a substantially higher specific power requirement than assumed in Refs. [1–3] due to a high specific CAPEX share and few incentives for high energy efficiency. Beyond this output, the generally preferred technology is a hydrogen Claude cycle [17]. Hydrogen liquefaction plants have so far only been realised to capacities in the rough range of 15–30 ton/d due to idiosyncratic demands from particular industries. For substantial scaling-up for energy commodity markets, additional process modifications and scaling benefits will be enabled. According to Cardella [18], a 68 % reduction in specific liquefaction cost, to below 1 €/kg, can be obtained when scaling up the output from the well-established 5 ton/d block capacity to 100 ton/d. Even at 100 t/d output, it must be emphasised that this represents a very low capacity compared to natural gas liquefaction plants. Depending on unit (output expressed in the form of mass, volume or energy rate), the throughput of a full-scale LNG plant is at least 2–3 orders of magnitude higher than that of existing hydrogen liquefiers. A single-train natural gas liquefaction plant can have a capacity of at least 5.5 Mt/a, or 15 kt/d, which on a mass basis is roughly 150 times that of a prospective 100 t/d plant, which would be a large-scale LH$_2$ plant by today's measures. If hydrogen liquefiers are to be scaled up by another order of magnitude, e.g. to 1 kt/day, new cryogenic refrigeration technology relying on centrifugal compressors may provide further scaling benefits. Hence, to determine a scaling factor for existing block sizes, the reference capacity, reference investment as well as the choice of technology for different capacities must be carefully considered.

Regarding the scalability of LOHC-related technology, a potential limiting factor in the scaling-up of e.g. cylindrical catalyst-filled reactors with substantial requirement for heat ingress or heat rejection is the heat transfer through the pipe wall, either from the outside-in or from the inside-out, which must be effective to the full reaction zone in the cross section. Thus, it can be expected that a reactor unit will have a maximum realisable diameter and capacity, beyond which parallelisation is required, which in turn implies a higher scaling exponent.

4.2. Economic lifetime and operation and maintenance cost

Unclear assumptions on other economic parameters are commonly observed in the literature, for example when it comes to assumed lifetime and fixed operating costs of technologies in comparative studies. Should assumptions differ significantly between different technologies, the reason should be clearly motivated to understand why specific technologies appear more or less feasible from this perspective. To exemplify, Refs. [1,4] both assume a fixed operation and maintenance cost to 8 % of the investment cost for a hydrogen liquefaction plant while half this value is assumed for other technologies in comparison, for unclear reasons. A reference is made to the IDEALHY project [19], where on the contrary, a 4 % fixed operation and maintenance cost is used. Correspondingly, Ref. [3] assumes a value of 3 % for the hydrogen liquefaction plant as well as for the LH$_2$ storage tanks. Similarly, in Ref. [1] the economic lifetime of the LH$_2$ receiving terminal (denoted "Hydrogen release") is set to 10 years, half of that for the other alternatives in the comparison, also for unclear reasons and contrary to the reference studies of [4,19]. d'Amore-Domenech et al. [3] assume a 30-year lifespan for all investigated infrastructures, conditional to technology-specific mid-life refurbishment costs. Although these assumptions might not, in the specific studies exemplified here, have a strong enough impact on their own to alter the conclusions it is still important to understand and motivate the reasons for these differences and the general impact they can have in comparative studies through sensitivity or uncertainty analyses [15].

5. Impact of battery limits and other central assumptions

In addition to the differences attributable to the different assumptions for LH$_2$ boiloff ratios, there are several other differences and concerns attributable to various central assumptions that should be briefly mentioned, although further detailed evaluations and discussions thereof lie beyond the scope of this piece.

5.1. Power-to-hydrogen efficiency for water electrolysers and power demand for hydrogen liquefaction

The efficiency of hydrogen production by water electrolysis differs between the considered value chain studies:

- Niermann et al. [1] assume 81.5 % efficiency on HHV-basis for PEM electrolysers. The discharge pressure is not explicitly given, but is presumably around 20 bar, based on stated compression work figures required to reach different elevated pressure levels.
- Ishimoto et al. [2] assume 52.3 kWh electricity per kg hydrogen produced and compressed based on state-of-the-art alkaline electrolysis with additional compression from atmospheric pressure to 20 bar. This is equivalent to an overall HHV-based efficiency of 75.3 %.
- d'Amore-Domenech et al. [3] assume 80 % efficiency, presumably on HHV-basis, for PEM water electrolysers with 30 bar hydrogen discharge pressure.

Estimates for the overall efficiency of water electrolysis varies with electrolyser type, between review studies, and between expectations to future development. A review of the most recent technical specifications for scaled-up PEM electrolyser modules from commercial vendors, shows rated power requirements and efficiencies of e.g.:

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[4] Although it increases with capacity.

- 10 070 kW for 4050 kg hydrogen per day at 20 bar discharge [20], which translates to 66 % HHV-based energy efficiency.[5]
- 5.3 kWh/Nm^3 at 40 bar discharge [21], indicating 67 % HHV-based energy efficiency.[5]
- 5 kWh/Nm^3 at 15 bar discharge [22] and 68 % HHV-based energy efficiency[5].
- > 75.5 % efficiency, presumably HHV basis [23] (associated discharge pressure not disclosed).

The first three efficiency figures correspond well with the definition of state of the art in IEA's recent report "The Future of Hydrogen" [24], while 80 % HHV efficiency is in the higher end of 2030 predictions/extrapolations found in the same report. Since there is high spread among the assumptions for the efficiency of conversion from primary energy to compressed hydrogen, whether reflecting state of the art or future extrapolations, this must be devoted particular attention when comparing results from different energy chain studies. This can reduce the risk of any ambiguities regarding the specific energy demand of electrolyser units, whether it reflects that of the stand-alone stack or that including the full balance-of-plant (BoP).

Another production step with a considerable power demand is the liquefaction of hydrogen. For this step, all studies have assumed values that are rather close in absolute value: 6.78 kWh/kg [1], 6.5 kWh/kg [2], and 6.1 kWh/kg [3]. These efficiency figures are yet to be realised but are expected to be so with the scaling-up of liquefier capacities [18, 24].

5.2. Battery limits – final hydrogen state in the receiving end

Due to the low molecular mass of hydrogen, compression is generally highly power-intensive. In all practical aspects, only displacement-type compressors such as piston and screw compressors are applicable. Having considerably lower throughput capabilities than axial and centrifugal compressors, displacement compressors have limited capacities and must be stacked for applications exceeding maximum single-machine capacity, which indicates a high scaling exponent for large scale capacities. Due to the relatively high energy requirement and unfavourable scaling characteristics, hydrogen compression, particularly from low pressures where the specific volumes are the highest, may in many cases be a significant cost driver in large-scale hydrogen logistics. The deeper analyses of such issues pertaining to the process intensification and scaling capabilities of hydrogen conditioning in the receiving end of envisaged large-scale logistics chains are arguably rather absent in the literature, hereunder hydrogenation/dehydrogenation or cracking reactor kinetics, heat supply and space velocity, product separation/purification and final pressurisation. As one example, several of these issues are superficially approached, while others are not mentioned, in the IEA report [24].

For liquid hydrogen, the conditioning between final LH$_2$ storage tanks and the end users will depend on the application. From a principal thermodynamic perspective, the conversion from LH$_2$ to pressurised state can be made simple and energy efficient with low-to negligible parasitic energy requirement. As one case-specific example for distribution of imported LH$_2$ out to decentralised vehicle refuelling stations, LH$_2$ can be pressurised directly to around 900 bar by a submerged cryogenic piston pump [5], which reduces the compression power substantially relative to gaseous compression. For this end-use specification, a portion of the thermomechanical exergy added to liquefy hydrogen is recuperated indirectly through reduced compression power

in the receiving end. For low-to-medium pressure demands, LH$_2$ can in principle, like other cryogenic fluids, be compressed isochorically in various arrangements through heat ingress from the ambient surroundings. Compression without any significant exergy input is possible since the thermomechanical exergy level of LH$_2$ is significantly higher than that of compressed hydrogen at normal temperatures.[6] Hence, elevated pressures are attainable despite exergy destruction caused by heat ingress from the ambient air or water heat sources.

Ishimoto et al. [2] assume 182 kWh/t$_{LH2}$ of electricity consumption at the receiving terminal, which includes power consumption of envisaged equipment pieces such as submerged LH$_2$ pumps and boiloff gas compressors. The hydrogen pressure at the battery limits is 60 bar(g), aligned with [25], and is assumed to be supplied to the demand side by LH$_2$ pumps combined with pressurised vaporizers as well as boiloff gas compressors. No further conditioning or pressurisation of LH$_2$ is assumed in Ref. [3] after offloading. In Ref. [1], it is not clear how the conditioning path for LH$_2$ to compressed hydrogen at 200 bar is configured, but it is reasonable to believe the least efficient option possible is chosen, that is, isobaric regasification at atmospheric pressure followed by multi-stage gaseous compression.[7] We would not recommend this processing path from LH$_2$ to pressurised hydrogen as it does not recuperate any of the thermomechanical exergy, neither directly nor indirectly. The evaporation increases the specific volume by a factor of roughly 800 and creates vast, yet partly or wholly unavoidable, compression requirements. Instead, different pumping strategies at liquid and dense-phase conditions should be pursued also for large-scale applications, to the extent possible derived from LNG logistics, possibly in combination with auxiliary cycles for exergy recovery during reheating of pressurised hydrogen.

6. Further knowledge needs for hydrogen value chains

We have compared some of the overlapping assumptions related to long-distance LH$_2$-based value chains and shown that these can differ substantially between studies. The examples show that the energy results for chain studies can be highly sensitive to central input parameters, and that they must be interpreted with great care. The main example on this, highlighted in our comparison, is the assumed boiloff ratio for LH$_2$ transport. We have also indicated that technology and scaling extrapolations must be handled with care, and that such issues will require significant attention if the confidence in predictive chain studies, which involve hitherto unproven technologies and scales, is to be improved in the future. In our view, as considered in other applications [26], the main tool for improving the precision in such studies is to take a distinct bottom-up approach, starting with high-detail modelling of critical chain components that are yet to be realised and/or scaled up to relevant magnitudes, e.g. gigawatt-scale. Examples of such components are reactors, separators/purification units, transfer and storage units, and compression and conditioning units. The detailed bottom-up approach can so contribute to identifying limiting factors for the scaling-up of various carrier options and technology solutions, which can be related to e.g. intensification barriers related to heat transfer, reaction kinetics, and reactant retention time, separation and purification units, high-temperature and cryogenic heat exchangers, compressors or cryogenic pumps. To identify and overcome barriers specific for the different hydrogen carrier technologies, deeper investigations are required. This is a field where, in our opinion, a substantial amount of work remains to improve the understanding and quantification of the pros and cons of energy carriers in a true large-scale setting, that is, close

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[5] Only considers the input power and output hydrogen energy flow (HHV). A hydrogen density of 0.0899 kg/m^3 at normal conditions is assumed. An exergy-based efficiency definition will also account for the hydrogen discharge pressure level since the output will be defined as the sum of chemical exergy and thermomechanical exergy of the hydrogen product.

[6] The thermomechanical exergy of para-hydrogen at 1.2 bar and saturated-liquid conditions equals roughly that of gaseous hydrogen at $\geq$ 3000 bar pressure.

[7] Ref. [1] states: "Due to the evaporation unit and the final compression, LH$_2$ has comparatively high costs in this segment."

to or well into the gigawatt magnitude in terms of time-averaged energy transfer rates.

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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