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Hydrogen liquefaction and storage: Recent progress and perspectives

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ABSTRACT

The global energy sector accounts for ~75% of total greenhouse gas (GHG) emissions. Low-carbon energy carriers, such as hydrogen, are seen as necessary to enable an energy transition away from the current fossil-derived energy paradigm. Thus, the hydrogen economy concept is a key part of decarbonizing the global energy system. Hydrogen storage and transport are two key elements of hydrogen economy. Hydrogen can be stored in various forms, including its gaseous, liquid, and solid states, as well as derived chemical molecules. Among these, liquid hydrogen, due to its high energy density, ambient storage pressure, high hydrogen purity (no contamination risks), and mature technology (stationary liquid hydrogen storage), is suitable for the transport of large-volumes of hydrogen over long distances and has gained increased attention in recent years. However, there are critical obstacles to the development of liquid hydrogen systems, namely an energy intensive liquefaction process ($\sim$13.8 kWh/kg$_{LH2}$) and high hydrogen boil-off losses (liquid hydrogen evaporation during storage, 1–5% per day). This review focuses on the current state of technology development related to the liquid hydrogen supply chain. Hydrogen liquefaction, cryogenic storage technologies, liquid hydrogen transmission methods and liquid hydrogen regasification processes are discussed in terms of current industrial applications and underlying technologies to understand the drivers and barriers for liquid hydrogen to become a commercially viable part of the emerging global hydrogen economy. A key finding of this technical review is that liquid hydrogen can play an important role in the hydrogen economy - as long as necessary technological transport and storage innovations are achieved in parallel to technology demonstrations and market development efforts by countries committed liquid hydrogen as part of their hydrogen strategies.

1. Introduction

Hydrogen is one of the most promising energy vectors to assist the low-carbon energy transition of multiple hard-to-decarbonize sectors [1, 2]. More specifically, the current paradigm of predominantly fossil-derived energy used in industrial processes must gradually be changed to a paradigm in which multiple renewable and low-carbon energy sources are leveraged in industrial processes via multiple (and interconvertible) chemical energy carriers [3]. Yet, wider-scale adoption of hydrogen-based energy systems must address specific challenges across the technology value chain, where both the commercial-scale adoption of existing demonstration-scale technologies and further

development of low-to mid-TRL processes are accelerated [1].

Notably, while ensuring that hydrogen production systems are able to meet expected demand under forecast scenarios is important (in particular concerning the need to integrate variable renewable energy sources into the mix [4] and to guarantee additive CO_2 emissions reduction from existing fossil-derived processes [5]), widespread adoption of hydrogen systems in the industrial sector relies on solving another crucial problem, namely the lack of compatible infrastructure [6]. Whether via retrofits or greenfield deployment, extensive investments in the transmission, distribution and storage of hydrogen for end-use applications will be required [7,8] to meet even the most conservative hydrogen adoption targets [9,10].

As such, addressing the issues related to infrastructure is particularly important in the context of global hydrogen supply chains [8], as determining supply costs for low-carbon and renewable hydrogen will depend on the means by which hydrogen is transported as a gas, liquid or derivative form [11]. Further, the choice of transmission and storage medium and/or physical state is directly tied to lifecycle environmental impacts of supply chains [12], which are in turn affected by both spatial and temporal considerations (e.g., distance of supply routes, storage capacity, storage duration, throughput, among others) [13].

Thus, most hydrogen value chains currently being explored are considered more complicated than other comparable energy value chains [14], and three main physical conversion processes are envisioned to increase the volumetric energy density of hydrogen transport given that hydrogen energy density per unit volume is around four times less than that of liquid fuels [15]. The three main physical forms of volumetrically densified hydrogen are compressed gaseous hydrogen (CGH_2), cryo-compressed hydrogen (CcH_2), and liquefied hydrogen (LH_2). Besides these three main forms, slush hydrogen (hydrogen in a cryogenic solid–liquid two-phase form), featuring greater density and refrigerant heat capacity than liquid hydrogen, has also gained much attention for applications in rockets and fuel cells [16].

The selection of a physical state or a chemical energy carrier for the deployment of hydrogen supply chains is far from being a solved problem, as not only continued advances across processes and technologies have rapidly changed performance metrics of tentative supply chains (and thus altering highly sensitive central input parameters in current studies) [17], but also different geographical locations may have institutional and structural drivers favouring specific choices [18]. For instance, LH_2-based transmission systems may be more cost-effective than ammonia-based ones (NH_3), which rely on the chemical translation of hydrogen into ammonia and then back into hydrogen via ammonia cracking at consumer-gate [19]. An example of this is the Norway–Japan maritime shipping route. In addition, the CO_2 intensity of the grid electricity used for cracking (i.e., in Japan) also makes NH_3-based transmission less appealing from an emissions perspective. With energy efficiency improvements across the NH_3 value chain being possible, however, both the cost-effectiveness and CO_2 intensity of NH_3-based systems may surpass LH_2-based ones in specific offshore production scenarios [20]. Thus, understanding the potential advantages and disadvantages associated with each alternative is important to technology adoption and policymaking supportive of these novel hydrogen value chains. Also for consideration is the use of chemical conversion of hydrogen into liquid organic hydrogen carriers (LOHC) to achieve the necessary volumetric energy densification for cost-effective storage and transport. Using this approach, a chemical molecule is used as a hydrogen carrier with suitable LOHC molecules being those capable of undergoing reversible hydrogenation and dehydrogenation reactions. As such, each LOHC is effectively a pair of related chemical molecules that differ in their hydrogen "loading" states (i.e., unloaded and loaded LOHC molecules). Release of hydrogen for use at consumption sites results in the regeneration of the LOHC molecule from its loaded to its unloaded state. Again, the use of an LOHC rather than either liquid hydrogen or ammonia is a context dependent choice.

Given these considerations, hydrogen as a future energy vector is highly dependent on the means taken for its storage and transport. Liquid hydrogen, the focus of this work, has been mentioned in previous publications, particularly in some comprehensive hydrogen technical reviews [21], but their discussion on technical developments in liquid hydrogen only scratches the surface. There have also been some reviews in the scholarly literature focusing on liquid hydrogen, but they have mainly concentrated on only one dimension of liquid hydrogen, either development of hydrogen liquefaction processes [22–25], cryogenic storage for liquid hydrogen [26], or liquid hydrogen as fuel in the transport sector (e.g., vehicle, aviation and maritime) [27,28]. In the more comprehensive reviews of liquid hydrogen [29,30], the entire liquid hydrogen supply chain, from liquefaction at the production side to

regasification at the user side, was not addressed. Liquid hydrogen transmission and regasification are either not included or are presented in a very limited way. Furthermore, the interplay between technical developments and opportunities for liquid hydrogen in the emerging global hydrogen economy are not typically highlighted in the published technical reviews. Therefore, the present work aims to bridge these gaps by providing a comprehensive and critical review on the technical aspects of liquid hydrogen supply chains, a brief comparison between liquid hydrogen and alternative liquid-phase hydrogen carriers, and the pathways for liquid hydrogen to achieve impact in the broader hydrogen economy landscape.

2. Materials and methods

2.1. Literature review approach

A parallel document search was undertaken through the search engines Scopus and Web of Science. To capture the relevant literature, research queries were conducted in both databases for the words "hydrogen storage", "hydrogen economy", "liquid hydrogen storage", "liquid hydrogen", "hydrogen liquefaction", "hydrogen transportation", "hydrogen carrier", "ammonia", and "LOHC", in the title, abstract and keywords. From these initial search queries, irrelevant sources were further discarded based on factors such as scope, availability, and language. No temporal restriction was adopted to limit results based on the date of publication, however documents containing outdated information or supplanted by more recently reported data were also excluded. Forward and backward references were then made across the documents to retrieve more publications deemed suitable for inclusion. Although systematic search and selection strategies were adopted, we consider our approach to fall under the "hybrid review" theoretical type as discussed by Paré et al. [31] and Xiao and Watson [32], incorporating elements of both descriptive and critical reviews. In total, around 200 publications have been selected for inclusion in this manuscript. A summary of the criteria used in the literature search and selection of results is presented in Table 1.

2.2. Structure of this review article

The structure of this article comprises eight sections. Section 1 introduces the research background, namely the growing importance of hydrogen as an energy carrier in energy awareness and its main implications. Section 2 explains the literature review method and structure for this article, followed by five main sections (section 3-7).

Section 3 presents the basic hydrogen characteristics and compares different hydrogen storage forms. The potential opportunity of liquid hydrogen in the hydrogen economy is identified.

Sections 4, 5 and 6 focuses on the state-of-the-art developments of hydrogen liquefaction processes, liquid hydrogen storage/transmission methods and liquid hydrogen regasification processes, respectively, where the challenges they face, and the corresponding solutions are discussed.

Section 7 discusses the emerging opportunities of liquid hydrogen as a hydrogen carrier compared to ammonia and LOHCs, specifically in the context of long-distance maritime transport, and provides a roadmap to capture these opportunities.

Finally, Section 8 summarizes and highlights the important findings.

3. Comparison of hydrogen storage technologies

3.1. Hydrogen characteristics

3.1.1. General properties of hydrogen

Hydrogen is the lightest and the most abundant element, which in its most prevalent form (^1H, protium) has a proton, an electron, with no neutrons. Other two naturally occurring isotopes, ^2H (deuterium,

Table 1

Overview of literature search and evaluation methodology of results adopted in this review.

Literature search criteria	Description	Comment
Inclusion criterion	English-language publications retrieved from search engines Scopus and Web of Science.	Search results were not temporally limited based on date of publication.
Literature identification	Search results obtained from using the search strings "hydrogen storage", "hydrogen economy", "liquid hydrogen storage", "liquid hydrogen", "hydrogen liquefaction", "hydrogen transportation", "hydrogen carrier", "ammonia", "LOHC".	Initial search results represent the cumulative pool of positively identified documents via each databases' own search engine. Initial results include non-unique entries (either via positive selection from multiple search strings used in the same database or due to the original document being indexed in more than one database, thus being included in the initial results pool of both database searches).
Screening for inclusion	Screening of all manuscripts identified in the literature identification step for thematic fitness to the topic of liquid hydrogen. Screening of title, abstract and key words at this step.	Positively included results discuss the topic of liquid hydrogen production, storage, transmission and use, or address actors, interventions, technology value chains, drivers and barriers to this technology pathway.
Quality and eligibility assessment	Full scanning of pre-screened results for thematic fitness, high-quality outputs from reputable sources.	Documents from non-reputable publishers or low-quality, non-peer reviewed outputs were excluded at this step. Documents with outdated information or supplanted by more recent data were also excluded.
Iterations	After evaluation of fully scanned results, duplicates resulting from indexing across both databases were also removed.	Grey literature outputs where yearly reports were published during the drafting of this review were manually updated to reflect the most up-to-date information where applicable.

Table 2

Hydrogen properties. Adapted from [29].

| Properties	Value
Lower heating value (MJ/kg)	118.8
Higher heating value (MJ/kg)	143
Boiling temperature at 1 atm (°C)	−253
Melting temperature (°C)	−259
Critical temperature (°C)	−240.01
Critical pressure (MPa)	1.3
Density of gaseous hydrogen at 0 °C (kg/m^3)	0.08987
Density of liquid hydrogen at −253 °C (kg/m^3)	70.85
Heat capacity of gaseous hydrogen at 0 °C (kJ/(kg·K))	14.3
Heat capacity of liquid hydrogen at −253 °C (kJ/(kg·K))	8.1
Heat of vaporization at −253 °C (kJ/kg)	447
Thermal conductivity of liquid hydrogen at −253 °C (W/(m·K))	0.104
Thermal conductivity of gaseous hydrogen at 0 °C (W/(m·K))	0.173
Volumetric energy density of liquid hydrogen at −253 °C (kWh/L)	2.36
Gravimetric energy density of hydrogen (kWh/kg)	33.3
Heat of ortho-to-para hydrogen at −253 °C (kJ/kg)	703
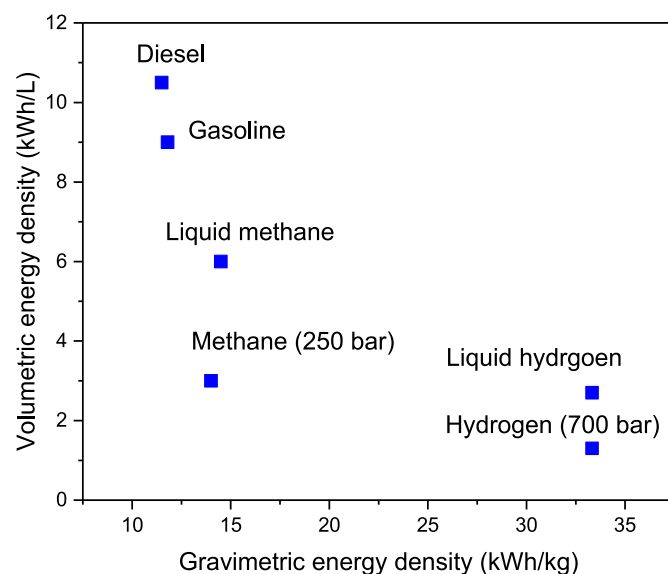
Fig. 1. The energy density of hydrogen compared to other common gases/liquids (data collected from Ref. [36]).

containing one proton and one neutron in the nucleus) and ^{3}H (tritium, with one proton and two neutrons in the nucleus) are utilized in nuclear fusion devices [33]. At ambient conditions (i.e., 20 °C, 1 atm), hydrogen is a colourless, odourless, tasteless, non-toxic, and highly combustible gas of diatomic molecules. The small and light hydrogen molecule (2.016 g/mol) shows a high mobility with a very high diffusion rate of 0.61 cm^2/s [34].

Compared to gaseous hydrogen, liquid hydrogen is characterised by its cryogenic temperature and high density. Hydrogen has a very low critical temperature of −240.01 °C, which indicates difficulties in condensing hydrogen. At the ambient pressure (1 atm), the liquefaction temperature of hydrogen is −253 °C. The lower heating value (LHV) of hydrogen is as high as ~120 kJ/g, which is the highest gravimetric energy density of all known substances [35]. Table 2 lists some common physical properties of hydrogen. On a mass basis, hydrogen has quite a high energy density, which is almost 3 times that of gasoline (see Fig. 1). However, on a volume basis, although liquid hydrogen has a higher energy content than compressed hydrogen, it is still much lower than most of traditional fossil fuels.

3.1.2. Ortho-para conversion

Diatomic hydrogen occurs in two spin isomeric forms, one with its

two proton nuclear spins aligned parallel (i.e., ortho-hydrogen), and the other with the two proton spins aligned antiparallel (i.e., para-hydrogen). The two forms of molecular hydrogen were first proposed by Werner Heisenberg and Friedrich Hund in 1927 [37]. Two years later, pure para-hydrogen was first synthesized by Paul Harteck and Karl Friedrich Bonhoeffer at the Kaiser Wilhelm Institute for Physical Chemistry and Electrochemistry [38].

The associated magnetic moment of a proton is related to the proton's spin. The chemical properties of these spin isomers are equivalent. However, there are slight differences in thermal, magnetic, and optical properties between the two types of hydrogen. Compared with para-hydrogen, ortho-hydrogen is in a higher energy state. The concentrations of the ortho-hydrogen and para-hydrogen depends mainly on temperature; see Fig. 2. Hydrogen consists of 75% ortho-hydrogen and 25% para-hydrogen at the room temperature, while liquid hydrogen contains 100% para-hydrogen. Under the cryogenic environment, ortho-hydrogen is thermodynamically unstable and spontaneously converts into para-hydrogen. Such a conversion process is exothermic, which is one of the critical reasons for the boil-off during liquid hydrogen during storage. Thus, the coexistence of two spin isomeric forms must be considered when handling liquid hydrogen.

Given the heat generated from the complete conversion of ortho-hydrogen to para-hydrogen of 703 kJ/kg, one can estimate that, in the
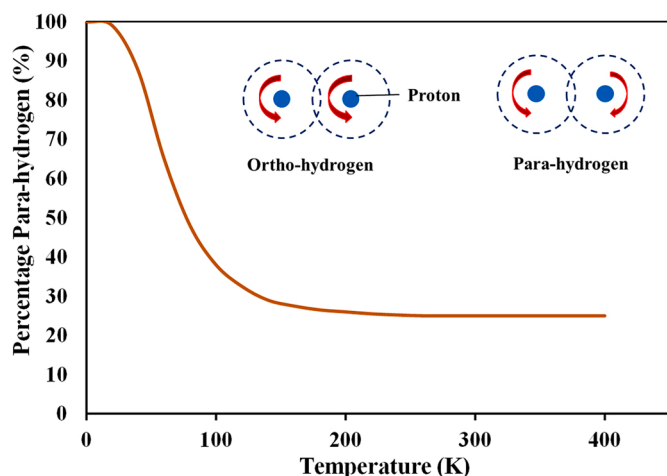
Fig. 2. Ortho-hydrogen and Para-hydrogen composition at equilibrium.

case of normal gaseous hydrogen liquefaction, the conversion of the para-hydrogen from 25% (at room temperature) to 100% (when liquefied), gives out 527 kJ/kg of heat. However, the latent heat of liquid hydrogen vaporization is only 446 kJ/kg [29]. The heat absorbed via liquid hydrogen evaporation is lower than the heat generated via ortho-to-paraconversion, leading to the continuous boil-off of liquid hydrogen, and hence, a low storage efficiency at this state. When the gaseous hydrogen with 75% ortho-hydrogen is liquefied, up to 50% stored hydrogen evaporates after 100 h and 65% of stored hydrogen evaporates after 1000 h due to the exothermic ortho-to-para conversion [39,40]. Thus, to realize the long-term liquid hydrogen storage, a higher concentration of para-hydrogen (95–98%) is essential prior to hydrogen storage [41]. Measures, such as the use of catalysts (e.g., iron hydroxides and chromium oxides), are often employed to accelerate the ortho-to-para conversion during hydrogen liquefaction, and hence reducing the boil off during the storage.

3.2. Basic comparison of different forms of hydrogen

As already discussed, storing and transporting hydrogen in large amounts is one of the most significant challenges for establishing a robust hydrogen economy. Hence, this section presents the state of knowledge concerning the three main physical forms of hydrogen carriers (CGH_2, CcH_2, LH_2) and their typical pressure and temperature values used in commercial systems (Fig. 3). CcH_2 technology can thermodynamically be considered a combination of compression (CGH_2) and cooling/liquefaction (LH_2). Pursuing the combination of both systems (from this thermodynamic perspective) makes sense only if some of

the disadvantages of the individual technologies are eliminated or new, additive advantages are provided to offset the inherent added complexity of such integrations. Therefore, only CGH_2 and LH_2 as standalone technologies will be discussed and compared in this section.

CGH_2 storage is at the commercial stage for both stationary and mobility applications. Unlike LH_2 storage, CGH_2 storage may leverage natural formations with limited porosity, such as salt caverns, as physical media and storage space. The underground caverns, which may further include rock caverns and depleted gas/oil fields, are mainly used for bulk stationary CGH_2 storage with storage pressure of 50–200 bar, leading to a relatively low volumetric hydrogen storage density (~10 g/L) [42]. The salt cavern-based hydrogen storage has many advantages, such as negligible hydrogen loss, large capacity and low capital cost requirements when compared to the construction of dedicated storage vessels. The need for compatible natural and engineered geological formations, however, limits the widespread adoption of this storage mechanism, due to uneven distribution and availability of such formations around the world. Further, only select locations may be suitable for use, whether due to physical limitations on storage capacity or due to potential environmental impacts. Transmission and distribution pipelines are required if the hydrogen stored in underground caverns need to be transported elsewhere for use, which additionally makes this approach not very flexible to serving an emerging set of hydrogen supply and demand centres. Furthermore, the construction of a such pipeline infrastructure is highly capital intensive, with long implementation timelines.

Pressurized tanks are another option for CGH_2 storage. There are typically four types of compressed gas storage pressure vessels/tanks. Type I and II are usually used for stationary applications due to their high weight, while Type III and IV tanks can perform much better in portable applications. Among them, Type IV, i.e., a tank made of composite materials such as carbon fibre with a non-metallic composite liner, can withstand pressures up to 700–1000 bar [6], which can increase the volumetric hydrogen storage density to ~40 g/L (at 700 bar) [42]. The relatively low hydrogen storage density and high cost of Type IV tanks makes compressed hydrogen expensive to transport via road or ship and also limits it to small-scale and short-term applications given that tanks do not benefit from economies of scale.

LH_2 storage is a way to convert gaseous hydrogen to its pure liquid form to increase its energy density for storage and transport. Such a storage method must have three key components: a hydrogen liquefaction unit to cool down and liquefy gaseous hydrogen, a liquid hydrogen storage tank, and a regasification unit to convert the liquid hydrogen back into gaseous form. The advantages of LH_2 storage lies in its high volumetric storage density (>60 g/L at 1 bar). However, the very high energy requirement of the current hydrogen liquefaction process and high rate of hydrogen loss due to boil-off (~1–5%) pose two critical challenges for the commercialization of LH_2 storage technology. Thus, LH_2 storage is currently at the late development stage.

Chemical hydrogen storage provides an alternative to physical forms of hydrogen storage, and the most investigated forms of chemical storage of hydrogen are also currently at the development stage at the latest. Ammonia and liquid organic hydrogen carriers (LOHC) are two most promising candidates, which can be used for long-term hydrogen storage. However, if pure hydrogen is required, neither of them can be used directly and energy-intensive hydrogen extraction processes must be performed.

Slush hydrogen, a mixture of solid and liquid state hydrogen, is produced by first liquefying hydrogen, and then further cooling the liquid to the triple point (13.81 K) or below [43]. Slush hydrogen is usually utilized for space and rocket fuel applications [43], which are beyond the scope of this review. Therefore, the details of slush hydrogen will not be discussed in this paper.

Table 3 summarizes the characteristics of different hydrogen storage methods and Fig. 4 provides their current and projected future levelized storage costs (LCOS). It can be seen that due to the very high energy
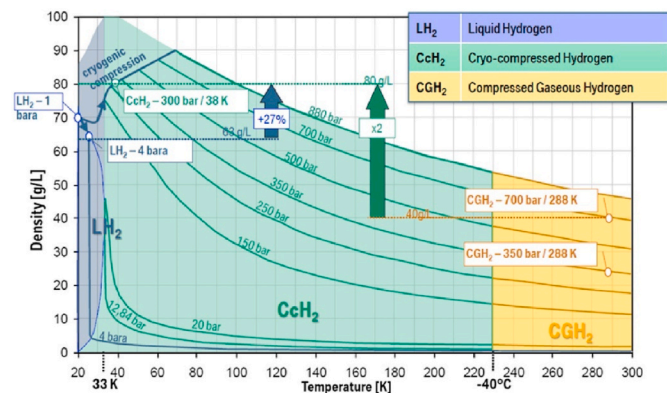

Fig. 3. Hydrogen density (y axis) as a function of temperature (x axis) and pressure (iso-lines) for physical based storage [6].

Table 3

Features of different hydrogen storage methods (data collected from Ref. [45]).

| Storage medium state	Storage method	Volume	Volumetric hydrogen storage density (g H$_2$/L)	Cycling	Geographical constraints
Gaseous state	Salt caverns	Large	~10 g/L (50–200 bar)	Month-weeks	Limited
	Pressurized containers	Small	~40 g/L (at 700 bar)	Daily	Not limited
Liquid state	Liquid hydrogen	Small-medium	~66 g/L (at 1 bar)	Days-weeks	Not limited
	Ammonia	Large	107 g/L (at 1 bar)	Month-weeks	Not limited
	LOHCs	Large	55 g/L (benzyltoluene at 1 bar)	Month-weeks	Not limited
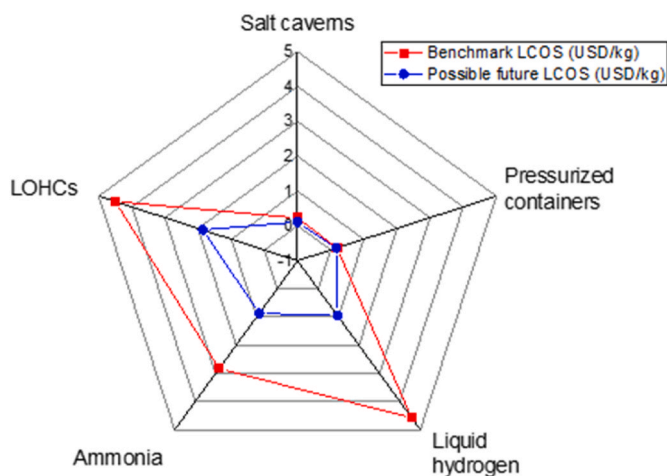
Fig. 4. Current and possible future LCOS of different hydrogen storage methods (data collected from Ref. [45]).

penalty of hydrogen liquefaction and high boil-off loss during transport, LH$_2$ storage shows the highest LCOS currently. It is for this reason that it is not considered an economically viable hydrogen carrier for the time being. However, LH$_2$ storage demonstrations are happening because liquid hydrogen still has some unique advantages over compressed hydrogen, especially its high density and flexible transportation, which makes it perform well in some specific applications. As shown in Fig. 5, hydrogen pipelines have the lowest transport costs for both short and medium-distance hydrogen transmission. However, due to lack of flexibility and long construction timelines for new pipelines, road transport via trucks and marine transport via ships are still needed for hydrogen transmission. Liquid hydrogen in particular can provide a much lower
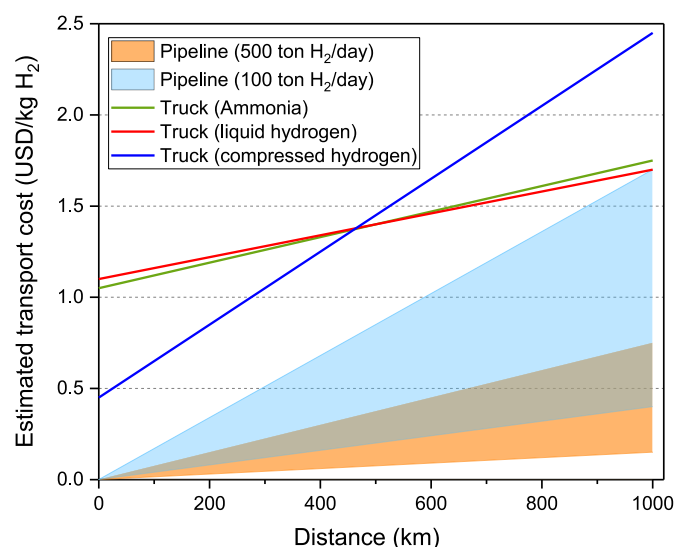
long-distance transport cost than compressed hydrogen (LH$_2$ transportation can potentially be even more cost-effective than pipeline transportation when transported volumes are high and the transmission distance is longer than the 2000–3000 km [44], although these distance are not depicted in Fig. 5). Based on this assessment, if synergistic opportunities for minimization of liquefaction energy consumption and of transmission/transport-associated boil-off losses are possible, liquid hydrogen still holds great promise for future applications in the hydrogen economy. As seen in Fig. 4, the possible future LCOS of LH$_2$ still shows a huge potential for reduction. An in-depth understanding of the approaches, technical barriers and pathways of LH$_2$ storage technology can even help accelerate this process.

4. Hydrogen liquefaction processes

Hydrogen liquefaction, one of the key processes in the overall liquid hydrogen supply chain, is a very energy intensive process. Although hydrogen liquefaction is considered a proven technology, improvements are still being made to further reduce energy consumption and enhance efficiency, which is critical to decrease the cost of the liquid hydrogen supply chain and make it more competitive for use in the hydrogen economy.

Many hydrogen liquefaction methods have been proposed/practised, and these can be broadly divided into two categories in terms of thermodynamic cycles: common liquefaction cycles (including Linde-Hampson cycles, refrigerant liquefaction cycles, Claude cycles, and expander liquefaction cycles), and magnetic refrigeration cycles. So far, some common hydrogen liquefaction cycles have been commercialized for practical use. This section will introduce the fundamental principles of these cycles, compare their working performances, and summarise their current status and future potentials in terms of commercial applications.

4.1. Basics of hydrogen liquefaction

Fig. 6 gives a flowsheet of a baseline hydrogen liquefaction process. One can see that if the hydrogen gas is fed at low pressure, it needs to go through a compression process first, before being pre-cooled to around 80 K. The hydrogen then needs to be further cooled to a temperature lower than 30 K via a closed-loop refrigeration cycle, during which an adsorption process (to remove impurities) and a catalytic conversion process (to achieve ortho-to-para conversion) are carried out. Finally, liquid hydrogen can be obtained by a typical adiabatic expansion process (J-T expansion or turbine expansion) and stored at 20–30 K (0.1–0.2 MPa). As can be seen from this baseline hydrogen process, there are several key points related to the overall efficiency: (1) the hydrogen feed pressure, (2) the efficiencies of precooling system and main refrigeration system, (3) the efficiencies of key components (hydrogen compressor, heat exchanger, expansion unit).

Before looking at different hydrogen liquefaction cycles, some key parameters/assessment indicators have been defined and explained in Table 4, which help to understand the working performance of the different cycles.
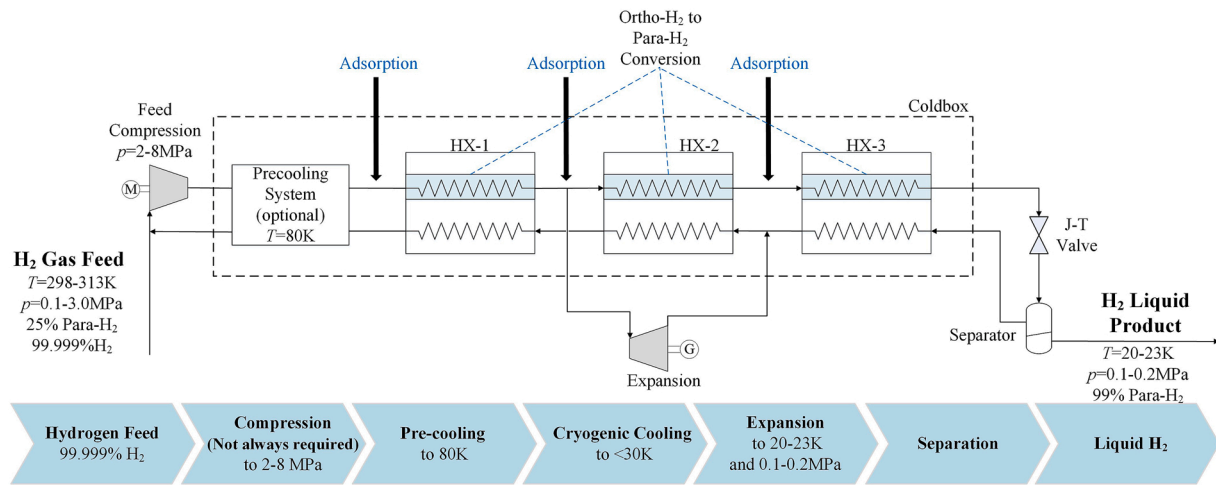

Fig. 5. Estimated transport costs of hydrogen (data collected from Ref. [46]).

Fig. 6. Flowsheet of a baseline hydrogen liquefaction cycle (adapted from Ref. [30]).

Table 4
Key assessment indicators for hydrogen liquefaction cycles.

Assessment indicators	Definition
Specific energy consumption (w, kWh/kg$_{H2}$)	The energy required to liquefy 1 kg of hydrogen: $w = W/m_{LH2}$ (W - net power consumption of one liquefaction cycle, m_{LH2} – mass flow rate of liquid hydrogen product)
Exergy efficiency (η_{ex}, %)	The ratio of theoretical minimum specific energy consumption to the actual specific energy consumption [30]: $\eta_{ex} = w_{ideal}/w$ (w_{ideal} - theoretical minimum specific energy consumption[a])
Yield or liquefaction rate (Y_{LH2}, %)	Ratio of liquid hydrogen flow to total input hydrogen flow: $Y_{LH2} = m_{LH2}/m_{H2}$ (m_{H2} – mass flow rate of fed hydrogen)

[a] w_{ideal} depends on the initial state of the input hydrogen (temperature and pressure), inlet and outlet para-hydrogen mole fractions, and the state of the selected reference ambient condition (2.7–3.9 kWh/kg$_{H2}$ [30,47,48]).

4.2. Theoretical hydrogen liquefaction methods

4.2.1. Linde-Hampson liquefaction cycle and its variants

The Linde-Hampson cycle is the most basic and straightforward gas liquefaction cycle [49], invented by Carl von Linde and William Hampson in 1895 for liquefying air [22]. A schematic of the Linde-Hampson cycle is shown in Fig. 7 (a). In this cycle, the gas is compressed to a high pressure first and then cooled down to a low temperature by the cold return gas in the main heat exchanger. Then, the low-temperature and high-pressure gas is expanded via a throttling valve (called Joule Thomson valve or JT valve) to a low-pressure state with temperature reduced during this isenthalpic process, resulting in the formation of liquid phase [50,51]. After expansion, the remaining gaseous portion is recirculated to the main heat exchanger for cooling power supply. This simple Linde-Hampson cycle is not suitable for large-scale gas liquefaction due to high energy consumption and low efficiency. Further, the Linde-Hampson cycle can only be used to liquefy gases that can be cooled by expansion at ambient temperature, e.g., nitrogen. However, the temperature of hydrogen increases after expansion at ambient temperature, which indicates that the Linde-Hampson cycle cannot be used for hydrogen liquefaction unless a pre-cooling process or other pre-treatment measures are used [40,52].

Pre-cooled Linde-Hampson cycle Pre-cooling hydrogen to its inversion temperature (200–205 K) or lower is a common way to make the simple Linde-Hampson cycle possible for hydrogen liquefaction. The pre-cooling can be done by, for example, adding a liquid nitrogen bath [53–55]. Fig. 7 (b) shows a simple flowsheet of a liquid nitrogen

pre-cooled Linde-Hampson cycle, in which a pressurized hydrogen stream is cooled down by an external cooling source (i.e., liquid nitrogen) in the first heat exchanger [51]. Nandi et al. [56] investigated the Linde-Hampson cycle with liquid nitrogen pre-cooling for hydrogen liquefaction, and obtained a liquid yield of 12–17%, with a specific energy consumption of 72.8–79.8 kWh/kg$_{H2}$ (i.e., energy consumption to produce 1 kg of liquid hydrogen), and an exergy efficiency of 4.5–5.0% depending on inlet pressure.

Dual-pressure Linde-Hampson cycle The Linde-Hampson cycle can be modified by increasing the number of compression and expansion stages; see example in Fig. 7 (c), where a dual-pressure Linde-Hampson system is shown. In such a cycle, the compression work is reduced by employing a two-stage compression, and, accordingly, a two-stage expansion is utilized. Instead of expanding to atmospheric pressure directly, the high-pressure gas is expanded through the first Joule-Thomson (JT) valve (denoted as main valve) to an intermediate pressure with liquid collected by a receiver and remaining gas returned to the inlet of the second-stage compressor. Then, the liquid from the receiver is further expanded to atmospheric pressure via a second JT valve, yielding more liquefied product compared to a single-stage expansion process [51]. Although this dual-pressure Linde-Hampson cycle can lead to a significant liquid yield enhancement for air liquefaction, the performance improvement is limited for hydrogen liquefaction [56].

Pre-cooled dual-pressure Linde-Hampson cycle To further enhance liquid hydrogen yield and reduce energy consumption, a combination of the pre-cooling and the dual-pressure cycles has been proposed, leading to a cycle known as pre-cooled dual-pressure Linde-Hampson cycle. A process flow diagram of such a cycle is shown in Fig. 7 (d). According to Peschka et al. [57], the pre-cooled dual-pressure Linde-Hampson cycle can achieve a liquid hydrogen yield of 41%, a specific energy consumption of 12.14 kWh/kg$_{H2}$ and an exergy efficiency of 27%.

4.2.2. Refrigerant liquefaction cycle (without isentropic expansion)

The working principle of a basic closed refrigeration cycle is shown in Fig. 8 (a) and is briefly explained here. Heat is removed by vaporization of a low-pressure refrigerant in the evaporator (for cooling). The refrigerant is then compressed and condensed at a higher pressure condition against a heat sink. Based the high reliability of cascade refrigerant liquefaction cycles in LNG liquefaction plants, Ho-Myung et al. [48] proposed to use similar configuration for hydrogen liquefaction, as shown in Fig. 8 (b). Exergy efficiency and liquid yield were analyzed, and their results showed that a dual-pressure system with two refrigerant cycles in cascade would be the best choice in terms of efficiency and operability. To enable the refrigerant cycle supply the
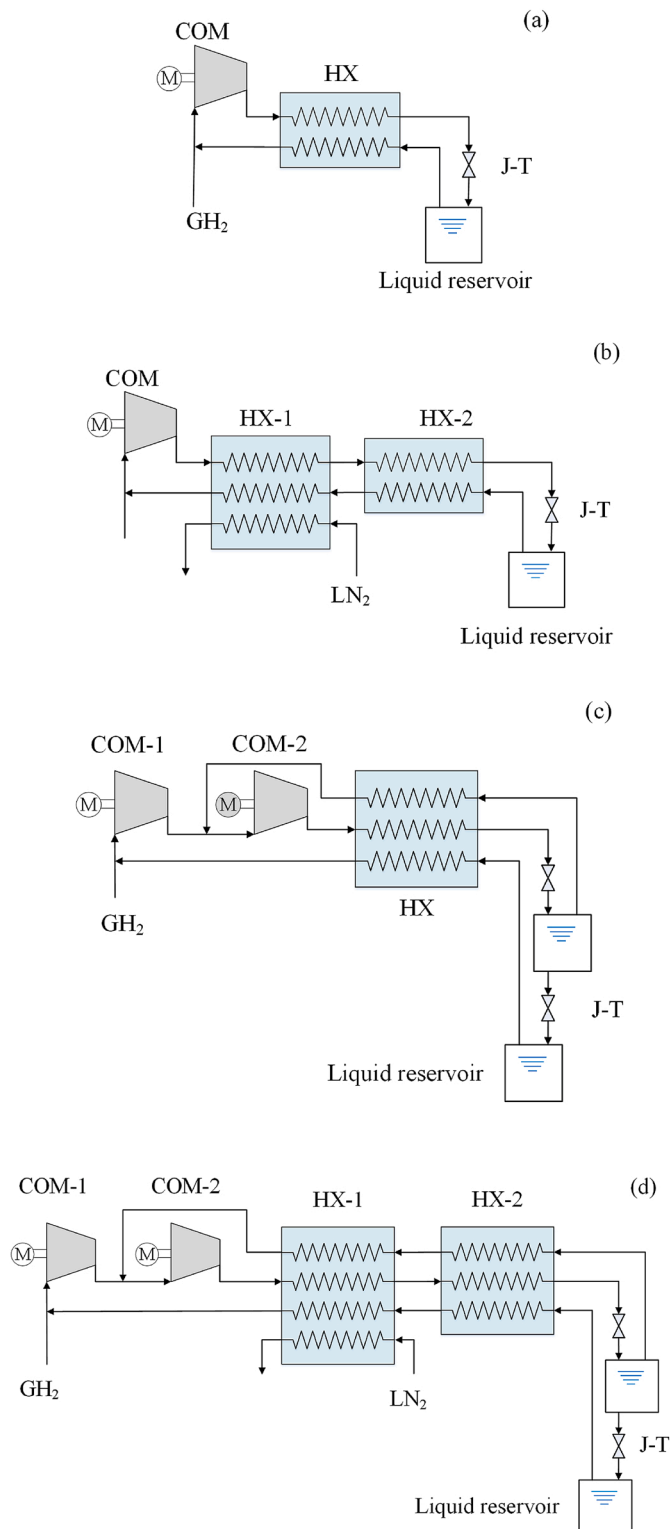
Fig. 7. Basic working principle of (a) simple Linde-Hampson cycle, (b) liquid nitrogen pre-cooled Linde-Hampson cycle, (c) dual-pressure Linde-Hampson cycle, and (d) pre-cooled dual-pressure Linde-Hampson cycle for hydrogen liquefaction (COM: compressor, HX: heat exchanger, J–T: JT valve).

cooling power along a wide temperature range, a multi-stage refrigerant system can be utilized, as shown in Fig. 8 (c) [24]. Mixed refrigerants are typically used that can undergo isobaric phase change through a range of temperature between the dew and bubble points (also known as temperature gliding), hence, providing a good match between the

process and refrigerant temperature distribution and a low exergy loss. The use of the mixed refrigerants can provide a simpler, more reliable system with greater efficiency than the use of a single refrigerant [58]. This type of refrigerant liquefaction process is usually used for liquified natural gas (LNG) production but not liquid hydrogen production [59].

4.2.3. Claude liquefaction cycle and its variants

Claude cycle was invented by George Claude in 1902, based on the Linde-Hampson cycle, to further improve the working performance of air liquefaction, as shown in Fig. 9 (a) [40]. The difference between the Linde-Hampson and Claude cycles is that the Claude cycle uses an additional isentropic expander [25]. In contrast to the isenthalpic process in the JT valve, the isentropic process in the expander can result in a higher gas temperature drop and lower exergy destruction. The expanded gas is then mixed with the regenerated return gas to supply the cooling load in an isobaric heat exchanger [51]. Due to the additional cooling effect provided by the isentropic expansion process, the Claude cycle can be used for hydrogen liquefaction without adding any pre-cooling measures [40]. The simple Claude cycle for liquid hydrogen production was reported to give a liquefaction yield of 8%, a specific energy consumption of 22.1 kWh/kg$_{H2}$ and an exergy efficiency of 18.1% [24].

Pre-cooled Claude cycle Several modifications based on the simple Claude cycle have been proposed to improve the hydrogen liquefaction performance further, among which the combination with a pre-cooling process is regarded as a promising option [51]. In 1989, Timmerhaus et al. [60] found that, with a liquid nitrogen pre-cooling process integrated to the Claude cycle, the exergy efficiency of the cycle could be increased to 50–70%. This is illustrated in Fig. 9 (b), where the compressed hydrogen is first cooled in a liquid nitrogen bath before entering subsequent heat exchangers. In 1993, Nandi et al. [56] investigated the effect of the hydrogen pressure after compression on the liquid nitrogen pre-cooled Claude cycle for hydrogen liquefaction. Their results showed that, at a hydrogen pressure of 1–3 MPa, the liquid hydrogen yield, energy consumption and exergy efficiency can reach 16–20%, 28–39.2 kWh/kg$_{H2}$, and 9.2–13%, respectively. In 2006, Kramer et al. [61] proposed the integration of a large hydrogen liquefaction plant with an LNG regasification terminal, using the cold energy released from the LNG regasification process as a pre-cooling load in hydrogen liquefaction. Their results showed that this integration could significantly reduce the specific energy consumption of hydrogen liquefaction to 3.2–8.5 kWh/kg$_{H2}$ with different LNG inputs (43.9–3.2 kg$_{LNG}$/kg$_{H2}$). However, this integration will reduce the flexibility of the hydrogen liquefaction plant because it must be co-located and co-operated with an LNG terminal.

Pre-cooled dual-pressure Claude cycle In 1978, Baker et al. [62] proposed a dual-pressure Claude cycle for hydrogen liquefaction with liquid nitrogen as hydrogen pre-cooling load, as shown in Fig. 9 (c). This pre-cooled dual-pressure Claude cycle with a designed production capacity of 250 US tons/day (or approximately 227 tonnes/day) can achieve a specific energy consumption of 10.85 kWh/kg$_{H2}$ and an exergy efficiency of 36%.

Thus, the Claude cycle based liquefaction cycle is very suitable for large-scale liquid hydrogen production plants in terms of both plant economics and operational economics, especially for plants with a liquefaction capacity of 3 tons/day or more [24]. Currently, the Claude cycle is the basis for almost all large-scale hydrogen liquefaction plants worldwide [63].

4.2.4. Expander liquefaction cycle

Expander liquefaction cycle uses one or more isentropic processes through expansion devices, for instance turbines, rather than the isenthalpic processes (i.e., JT valve) in a cascade liquefaction cycle. The expansion devices can produce a temperature much lower than that generated by an isenthalpic throttling process. According to this definition, Claude cycles can also be considered as an expander liquefaction
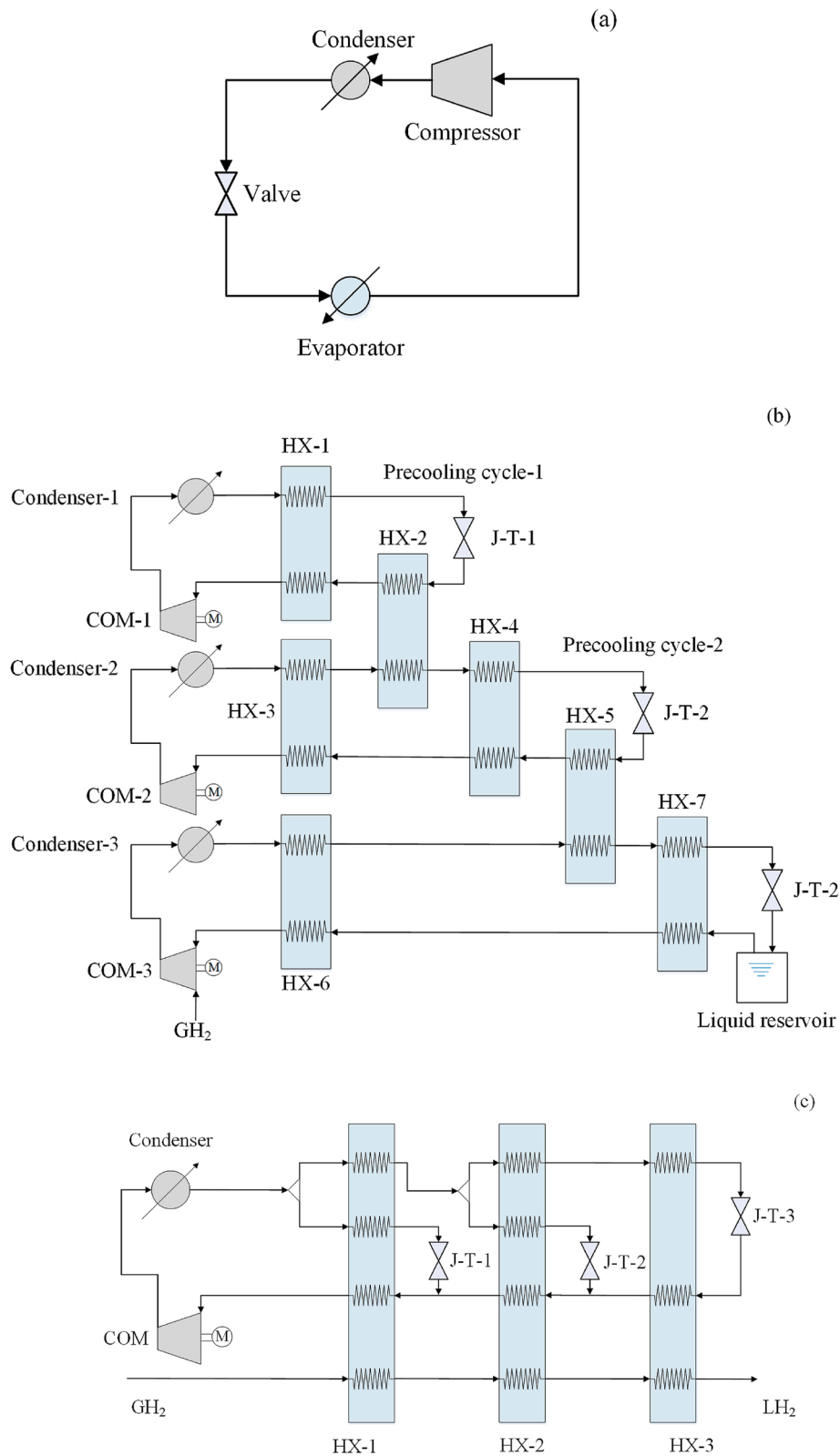
Fig. 8. Basic working principle of (a) simple refrigerant cycle, (b) cascade refrigerant cycle, and (c) multi-stage mixed refrigerant cycle without isentropic expansion for hydrogen liquefaction (COM: compressor, HX: heat exchanger, J–T: JT valve).

cycle.

Brayton cycle A flow diagram of a simplest Brayton cycle is given in Fig. 10 (a), in which helium or neon is usually used as the refrigerant (working fluid) to drive the refrigeration cycle. In such a cycle, the refrigerant is compressed to high pressure first, which then expands in

an isentropic expander to obtain the desired low temperature condition (i.e., lower than the temperature of liquid hydrogen). The very cold refrigerant then condenses the hydrogen that has already been pre-cooled by liquid nitrogen. In all Brayton cycles, the temperature pinch point (i.e., the minimum temperature difference between the hydrogen
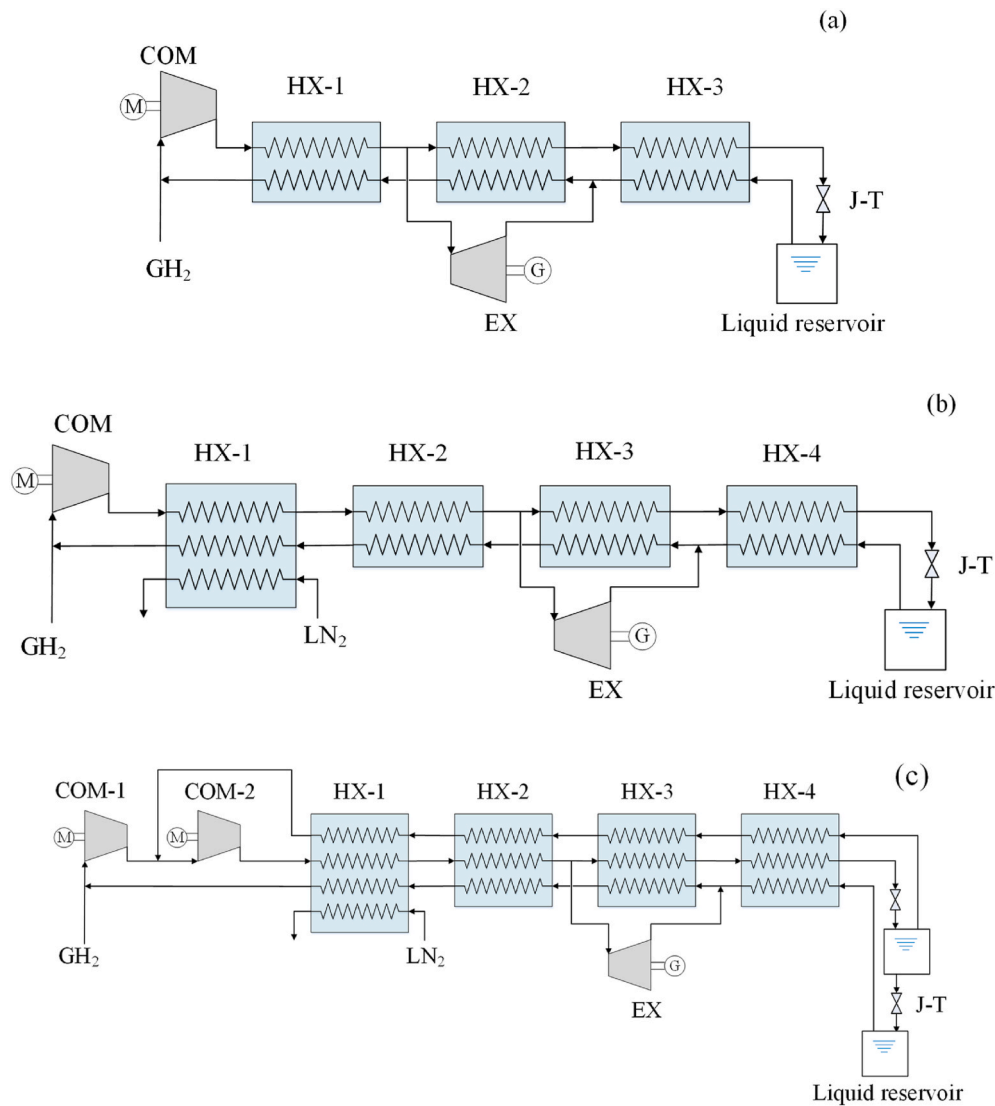
Fig. 9. Basic working principle of (a) simple Claude cycle, (b) liquid nitrogen pre-cooled Claude cycle, and (c) pre-cooled dual-pressure Claude cycle for hydrogen liquefaction (COM: compressor, EX: expander, HX: heat exchanger, J–T: JT valve).

and the refrigerant) of the coldest heat exchanger occurs at the saturated vapor point of hydrogen [64]. The Brayton hydrogen liquefaction cycle normally uses a tube-in-tube heat exchanger, which has advantages of simple construction, compact size, light weight and high safety [65]. Chang et al. [64] compared the thermodynamic performances of helium Brayton cycles with different configurations. Their results showed that a helium Brayton cycle without and with a liquid nitrogen pre-cooling process can obtain maximum liquefaction efficiencies of 8.7% and 21.1%, respectively. Besides, among different variants of the Brayton cycle, the helium Brayton cycle with two-stage expansion showed the highest FOM of 21.5% and 24.5% without and with liquid nitrogen for pre-cooling, respectively. Nandi et al. [56] reported that liquid hydrogen yields could reach 100% and 54% when the helium Brayton cycle was used for normal hydrogen and parahydrogen liquefaction, respectively.

Collins Cycle (multi-stage Brayton cycle) While multiple heat exchangers are employed with refrigerant stream splitting prior to expansion, the system could be regarded as a Collins cycle based liquefaction process. A flow diagram of such cycle is shown in Fig. 10 (b). The refrigeration loop bypass turbines in the Collins cycle are organized in such a manner that the inlet temperature of each expander is higher than the outlet temperature of the previous expander by an

amount equal to the temperature difference of the heat exchanger, which illustrates that the cooling powers in different temperature ranges can be supplied by different expansion processes. Therefore, a better matching between the cold fluid (i.e., refrigerant) and hot fluid (i.e., hydrogen) temperature profiles than the Brayton cycle could be achieved, leading to an improved thermodynamic efficiency [59]. If the feed gas (i.e., hydrogen) is used to replace the refrigerant in the Collins cycle, the system could be classified as a Claude cycle or one of its modifications. Valenti et al. [66] analyzed a four-stage helium expansion hydrogen liquefaction cycle with a liquefaction capacity of 867 tons/day, yielding a specific energy consumption of 5.04 kWh/kg_{H2} and an exergy efficiency of 47.7%. In 2017, Yuksel et al. [67] proposed an optimised four-stage helium expansion hydrogen liquefaction system with eight heat exchangers, in which the power generated by helium expanders is used to power compressors for hydrogen and helium compression. This system with a liquefaction capacity of 50 tons/day can achieve an exergy efficiency of 57.13%. Tang et al. [68] proposed a Collins cycle with liquid nitrogen precooling for liquid hydrogen production at 50 tons/day, and found that exergy efficiencies of 38.52% and 40.17% could be achieved by using a throttled expansion and a liquid expander at the final stage, respectively.

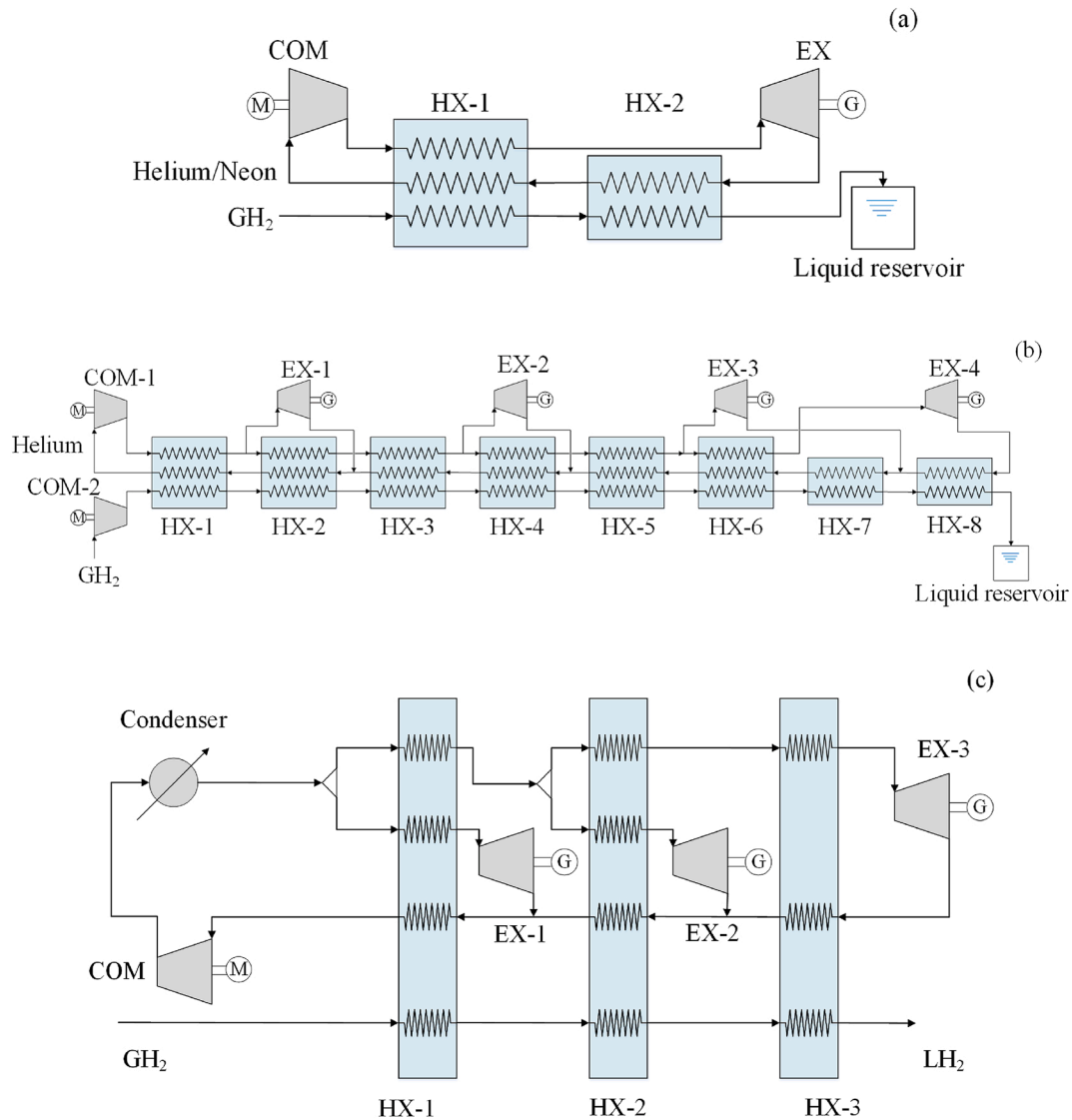*Mixed refrigerant cycle (with isentropic expansion)* A mixed

Fig. 10. Basic working principle of (a) Brayton cycle, (b) Collins cycle, and (c) mixed refrigerant Brayton cycle (COM: compressor, HX: heat exchanger).

refrigerant cycle is usually used as a pre-cooling process for liquid hydrogen liquefaction, in which a refrigerant mixture instead of a single refrigerant is used. The working principle of this type of cycle is similar to the refrigerant liquefaction cycle introduced in Section 3.2. The main difference is that isentropic expanders are utilized to replace the expansion valves to achieve cooling effect, as shown in Fig. 10 (c). Ansarinasab et al. [69] proposed a two-stage mixed refrigerant cycle system in which the first refrigeration stage uses a refrigerant mixture consisting of nine components to cool hydrogen to −195 °C and the second refrigeration stage uses a refrigerant mixture consisting of 6.19% hydrogen, 83.61% helium and 10.20% neon to further cool the hydrogen to −254.55 °C. The efficiency of the overall system is as high as 55.47% with a specific energy consumption of 1.10 kWh/kg$_{H2}$. Krasae-In et al. [70] proposed a Brayton cycle for large-scale hydrogen liquefaction using a mixed refrigerant pre-cooling system and four hydrogen Brayton refrigeration systems for liquefying hydrogen. Their results showed specific energy consumptions of 2.89 kWh/kg$_{H2}$ and 5.35 kWh/kg$_{H2}$ respectively under ideal and realistic operating conditions, and an exergy efficiency of equal or greater than 54.02% depending on the compressor and expander efficiencies. In 2014, Krasae-In Ref. [71] proposed a simplified five-component refrigerant mixture (4% hydrogen, 18% nitrogen, 24% methane, 28% ethane, 26% butane) combined with four optimised hydrogen Brayton cycle systems.

Their results showed an efficiency of 48.9% and a specific energy consumption of 5.91 kWh/kg$_{H2}$.

4.2.5. Magnetic refrigeration

Magnetic refrigeration is based on the magneto-thermal effect of magnetic materials. Such a magneto-thermal effect refers to the fact that during an excitation process, the orderliness of a magnetic material increases and the magnetic entropy decreases, and hence heat is released; during demagnetisation, the orderliness of the magnetic material decreases, while the magnetic entropy increases, leading to absorption of heat from outside and hence the creation of a cooling effect. The magnetic refrigeration system does not require a low-temperature compressor but uses solid materials as the working medium, so the application of magnetic refrigeration in hydrogen liquefaction has (claimed) potential advantages of low cost, simple and compact structure, light weight, no noise, easy maintenance and no pollution [72].

Fig. 11 (a) shows a simple flow diagram of a magnetic refrigerator for hydrogen liquefaction. The figure shows that hydrogen is cooled down from ambient temperature to −251 °C through 7–9 AMR (Active Magnetic Refrigerator) and CMR (Carnot Magnetic Refrigerator) stages to be liquefied. To improve the liquefaction performance of the magnetic refrigeration, liquid nitrogen can be used to pre-cool the hydrogen to −153 °C or −196 °C, as shown in Fig. 11 (b) and (c). The pre-cooled
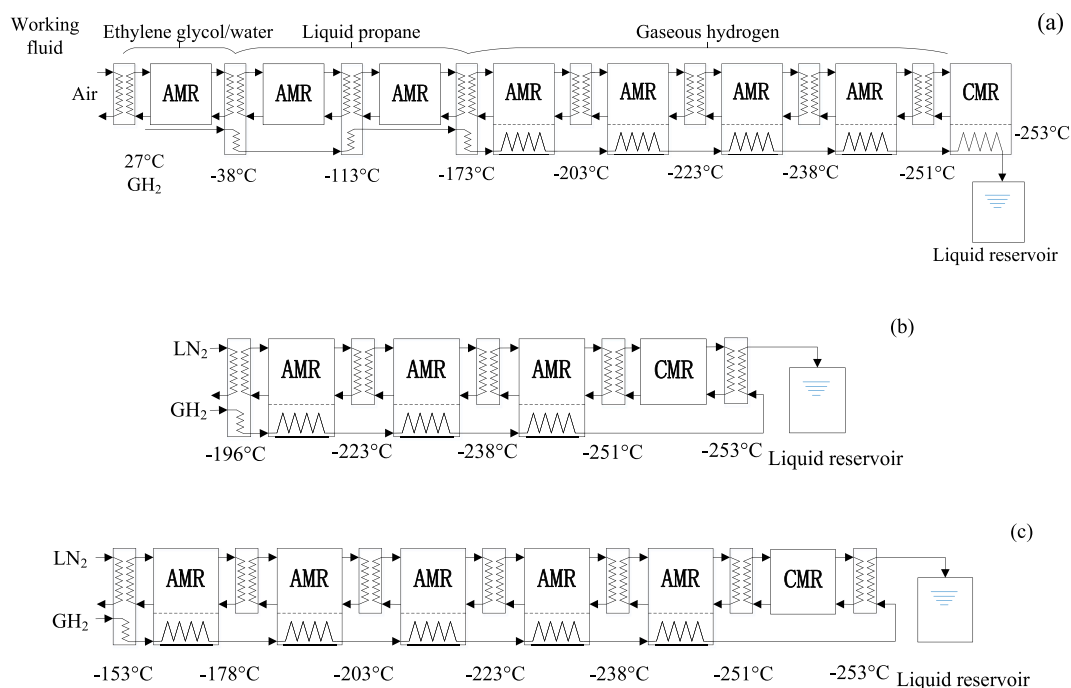
Fig. 11. Working principle of (a) Basic magnetic refrigerator, (b) pre-cooled magnetic refrigerator with three-stage AMR, and (c) pre-cooled magnetic refrigerator with five-stage AMR for hydrogen liquefaction.

hydrogen is further cooled to −251 °C by five AMR (Fig. 11 (b)) or three AMR (Fig. 11 (c)) stages with CMR to achieve liquefaction. It was reported that a liquid nitrogen pre-cooled magnetic refrigerator with CMR plus three-stage AMR could achieve a maximum FOM of 46.9% [72].

4.2.6. Summary of liquefaction processes

Table 5 summarizes the main studies on hydrogen liquefaction technologies, including the main features and important parameters, such as liquid hydrogen capacity, liquid yield, specific energy consumption and exergy efficiency. For a clearer illustration, the energy consumption and exergy efficiency of various types of hydrogen liquefaction cycles are shown and compared in Fig. 12. One can see, from Table 5, that the Linde-Hampson cycle typically has the highest energy consumption and the lowest efficiency, followed by the Claude cycle. Compared with these basic cycles, the more advanced expander cycles, i. e., Brayton cycle, Collins cycles and mixed refrigerant cycle, give a significantly enhanced performance. Among these advanced cycles, the mixed refrigerant cycle (with isentropic expansion) is the most researched hydrogen liquefaction process, and has been shown to have the lowest energy consumption and the highest efficiency, with the best performance reported as of 2019 (energy consumption of 1.1 kWh/kg_{H2} and an efficiency of 55.47%) [69]. Although magnetic refrigeration cycle has the advantages of low cost and simple structure, its liquefaction capacity is very low and so far it cannot be used for large-scale hydrogen liquefaction applications.

4.3. Commercial hydrogen liquefaction plants

The first hydrogen liquefier in the USA was built in 1904 using a Dewar hydrogen liquefier which was exhibited at the St Louis World's Fair and finally purchased by the National Bureau of Standards (NBS) [77]. In 1957, Air Products built a large-scale hydrogen liquefaction plant in Ohio with a similar design to the NBS system, and it could produce three tons of liquid hydrogen per day [78]. The third hydrogen liquefaction plant, which was equipped with three separate parallel purification units and two conventional parallel liquefaction units to ensure continuous operation, was located in California, began

construction in 1957 by Stearns-Roger of Denver and was fully operational in January 1958 [79].

Air Products supplies the largest quantity of liquid hydrogen in North America, followed by Praxair. Praxair currently has five liquid hydrogen production plants in the USA with a capacity of 6–35 tons/day [22]. The liquefaction processes of the Praxair large-scale hydrogen liquefaction plants typically employ a modified Claude cycle with pre-cooling [80]. As reported, the specific energy consumption and the exergy efficiency of this system are 12.5–15 kWh/kg_{H2} and 19–24%, respectively [22,80].

Linde has two installed hydrogen liquefaction plants in Germany: the first was built in Ingolstadt in 1991 and the second was built in Leuna in 2008. The Linde hydrogen liquefaction production plant in Ingolstadt, which uses feed gas produced by refineries, used to be the largest hydrogen liquefaction plant in Germany. The liquefier is based on a modified liquid nitrogen pre-cooled Claude cycle. Liquid nitrogen is used to precool the hydrogen to 80 K and a hydrogen expander refrigeration system is used to cool down the hydrogen further to 30 K before being liquefied by a throttling process (JT valve) [81]. This liquefier has a liquefaction capacity of 4.4 tons/day, and a specific energy consumption of 13.58 kWh/kg_{H2} [82]. In September 2007, Linde built its second hydrogen liquefaction plant at Leuna in Germany with an investment of €20 million. This plant can produce five tons liquid hydrogen per day with a specific energy consumption of 11.9 kWh/kg_{H2} [22]. The commercialized hydrogen liquefaction plants typically have a specific energy consumption within a range of 10–20 kWh/kg_{H2} [29, 83]. A plot of specific energy consumption versus the capacity of hydrogen liquefaction plants is given in Fig. 13 with the data collected from US Department of Energy [84]. The specific energy consumption decreases with increasing plant capacity with a minimum specific energy consumption of approximately 10 kWh/kg_{H2}. Table 6 summarizes hydrogen liquefaction plants built globally since the early 1950s, whereas a list of hydrogen liquefaction plants under construction or under planning is given in Table 7.

Table 5
Summary and comparison of conceptual hydrogen liquefaction cycles.

Cycle		Ref.	Capacity (tons/day)	Yield (%)	Specific energy consumption kWh/kg$_{LH2}$	Exergy efficiency (%)	Remarks
Linde-Hampson liquefaction cycle	Pre-cooled	Nandi et al., 1992 [56]	–	12–17	72.8–79.8	4.5–5.0	Inlet pressure: 6–10 MPa LN$_2$ precooling
	Pre-cooled dual-pressure	Peschka 1992 [57]	–	41	12.14	27	–
Claude liquefaction cycle	Simple	Majid et al., 2018 [24]	–	8	22.1	18.1	–
	Precooled	Nandi et al., 1992 [56]	–	16–20	28–39.2	9.2–13	Inlet pressure: 1–3 MPa LN$_2$ precooling
		Kuendig et al., 2006 [61]	50	100	3.2–8.5	–	LNG input for precooling: 43.9–3.2 kg$_{LNG}$/kg$_{LH2}$ Near seaport
	Pre-cooled dual-pressure	Baker et al., 1978 [62]	–	–	10.85	36	–
Expander liquefaction cycle	Brayton cycle	Nandi et al., 1993 [56]		100 for normal hydrogen (ratio of ortho- to para-hydrogen is 3: l) 54 for para-hydrogen	33.3–55.6	6.5–11	Inlet pressure (He): 1.5–2.5 MPa Mass ratio: 7–10
		Bian et al., 2004 [73]	120	–	6.60	47.0	LNG precooling and dual-pressure
	Collins cycle	Shimko 2008 [74]	50	100	7.4	44	–
		Yuksel et al., 2017 [67]	50	100	–	57.13	Four helium expanders
		Valenti et al., 2008 [66]	864	100	5.04	47.7	Four helium expanders
		Tang 2012 [68]	50	100	–	38.52	LN$_2$ precooling with throttle expansion at the final stage
		Tang 2012 [68]	50	100	–	40.17	LN$_2$ precooling with expander at the final stage
	Mixed refrigerant cycle (with isentropic expansion)	Ansarinasab et al., 2019 [69]	300	100	1.10	55.47	Refrigerant: 6.19% hydrogen, 83.61% helium and 10.20% neon (with inlet feed hydrogen pressure: 21 bar)
		Krasae-in et al., 2010 [70]	100	100	5.35	54.02	–
		Krasae-in 2014 [71]	100	100	5.91	48.9	Refrigerant: 4% hydrogen, 18% nitrogen, 24% methane, 28% ethane, and 26% butane
		Sadaghiani et al., 2017 [75]	300	100	4.41	55.47	Refrigerant: 10% neon, 6.5% hydrogen and 83.5%helium;
		Asadnia et al., 2017 [63]	–	100	7.69	39.5	–
		Aasadnia et al., 2018 [76]	90	100	6.47	45.5	–
Magnetic refrigeration		T. Utaki et al., 2007 [72]	0.01	–	8.45	–	3 stages AMR with CMR and precooled by LN$_2$

4.4. Outlook for hydrogen liquefaction

4.4.1. Gaps between theoretical and commercial hydrogen liquefaction

The minimum energy required for the thermodynamically ideal hydrogen liquefaction cycle was reported approximately 3 kWh/kg$_{LH2}$ [22,24]. The classical theoretical cycles (mainly Linde-Hampson and Claude cycles) developed a long time ago showed high energy consumption (>10 kWh/kg$_{LH2}$). Today, some optimised liquefaction cycles (mainly expander liquefaction cycles) can deliver much lower energy consumptions, below 10 kWh/kg$_{LH2}$ and in some cases even below 6 kWh/kg$_{LH2}$. However, the average specific energy consumption for large plants in-service today around the world is 13.83 kWh/kg$_{LH2}$ [24], a significant gap from the optimised theoretical value. The ultimate energy consumption target for large-scale hydrogen liquefaction plants (with a capacity of 300 ton/day) has been set to 6 kWh/kg$_{LH2}$ by the US Department of Energy (DOE) [89]. Both system-level and component/material level measures can be implemented to accelerate the achievement of this target:

System-level optimisation

- Upgrade the hydrogen liquefaction configurations currently used in the industry to more efficient and energy-saving configurations.
- Optimise the scale/capacity of the hydrogen liquefaction plants (large-scale hydrogen liquefiers are expected to be more cost-effective and efficient).
- Co-locate the hydrogen liquefaction plants with the high-grade waste cold, e.g., LNG regasification terminals.
- Recover the high-grade cold energy released during liquid hydrogen transportation and utilization (boil-off, regasification, and para-to-ortho conversion), and reuse it in the hydrogen liquefaction process.

Component/material-level optimisation

- Design and adopt more efficient components, including compressors, expanders, heat exchangers, and insulation material etc.
- Using expanders instead of JT valves in the hydrogen liquefaction process.
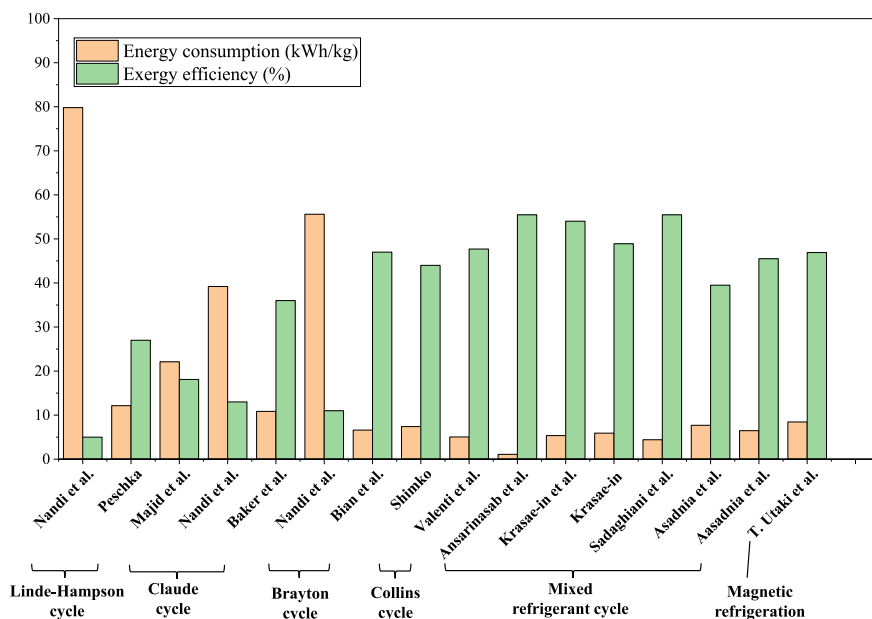- Develop novel mixed refrigerants.

Fig. 12. Summary of energy consumption and exergy efficiency of hydrogen liquefaction cycles reported in the literature.
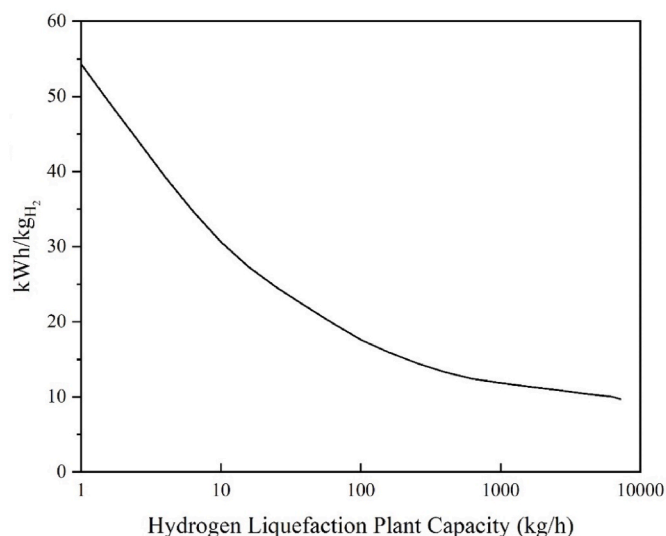

Fig. 13. Typical energy requirements for the hydrogen liquefaction plant as a function of plant size.

- Develop novel cold energy storage materials which can recovery and store the high-grade cold of liquid hydrogen.

4.4.2. Future opportunities

Large-scale hydrogen liquefaction plants adopting state-of-the-art technology to improve exergy efficiency and reduce energy consumption represent the future development of liquid hydrogen production. However, this goal needs to be achieved through a step-by-step process. A summary of stepwise implementation of large-scale hydrogen lique-faction technology is given in Table 8. It can be seen that with scaling up of hydrogen liquefaction plants and the upgrading of refrigeration cycles and components in the future, the focus shifts from operating cost to investment cost. Despite the increase in investment costs, the reduction in operating costs resulting from the significant reduction in energy consumption can bring significant economic benefits [90]. A projected trend of specific hydrogen liquefaction costs is shown in Fig. 14.

5. Liquid hydrogen storage and transmission

5.1. Categories of liquid hydrogen storage

Hydrogen liquefaction is typically considered for meeting long-distance transportation requirements, such as intercontinental hydrogen shipping [92]. However, small-scale liquid hydrogen storage is also needed for some specific application scenarios, such as on-board storage for fuel cell vehicles and aviation [93]. Based on the application areas of hydrogen, the storage of liquid hydrogen can be divided broadly into two main groups: stationary storage and mobile storage, as shown in Fig. 15. Stationary storage refers to the on-site liquid hydrogen storage at a production site, an end-user site and a hydrogen-fuelled power generation site. Mobile storage usually encompasses the storage of liquid hydrogen storage during transport via a truck or ship, or for fuel supply to vehicles or aircraft.

5.2. Conditions and challenges for storage and transmission

All cryogenic infrastructure, such as storage tanks/vessels, pipelines and refrigeration systems, are designed to ensure that liquid hydrogen can be stored and transported at its characteristic temperatures. During liquid hydrogen storage and transmission, heat leakage is unavoidable, which is particularly important for long duration storage or for storage for long distance transport. The evaporation of liquid hydrogen during storage is referred to as boil-off [94]. Compared with gaseous hydrogen, liquid hydrogen possesses a higher volumetric energy density, which indicates that more liquid hydrogen can be stored in a fixed-volume tank. The boil off gas (BOG) should be released when it generated, otherwise, the increasing pressure in the infrastructure may lead to safety issues, such as risk of explosion. As a small amount of hydrogen gas exhausted to the environment is harmless and the easiest way to dispose of the boil-off hydrogen during storage is to release it directly into the atmosphere. However, to avoid waste and for safety reasons, many producers choose to collect and reuse the boil-off hydrogen rather than exhausting it.

Boil-off occurs due to one or more mechanisms, including ortho-hydrogen to para-hydrogen conversion (spin isomer conversion), heat leakage [95], thermal stratification, sloshing, and flashing. The details of each of the mechanisms are discussed below.

Table 6

Global overview of hydrogen liquefaction plants [22,24,40,77,85,86].

Country	Location	Operated by	Capacity (tons/day)	Year established	Still in service	SEC[a] (kWh/kg$_{LH2}$)
USA	Colorado	NBS	0.5	1952	–	–
USA	Ohio	APCI	1	1956	–	–
USA	Painsville	Air Products	3	1957	No	–
USA	West Palm Beach	Air Products	3.2	1957	No	–
USA	Florida	APCI	3.5	1957	–	–
USA	California	Stearns-Roger Mfg. Co.	1.5	1957	–	–
USA	Painsville	Air Products	3	1957	No	–
USA	West Palm Beach	Air Products	3.2	1957	No	–
USA	Florida	APCI	30	1958	–	–
USA	West Palm Beach	Air Products	27	1959	No	–
USA	Mississippi	Air Products	32.7	1960	No	–
USA	California	Stearns-Roger Mfg. Co.	7	1960	–	–
USA	Ontario, CA	Praxair	20	1962	Yes	–
USA	California	Stearns-Roger Mfg. Co.	26	1962	–	–
USA	California	APCI	32.5	1963	–	–
USA	Sacramento	Union Carbide	54	1964	No	–
USA	New Orleans, LA	APCI	34	1977	Yes	–
USA	New Orleans, LA	APCI	34	1978	Yes	–
Japan	Amagasaki	Iwatani	1.2	1978	No	–
USA	Niagara Falls, NY	Praxair	18	1981	Yes	–
Canada	Sarnia Ontario,	APCI	29	1982	Yes	–
Japan	Tashiro	MHI	0.6	1984	No	–
Japan	Akita Prefecture	Tashiro	0.7	1985	Yes	–
USA	Sacramento, CA	APCI	5	1986	Yes	–
Japan	Tane-Ga-Shima	Japan Liquid Hydrogen	1.4	1986	Yes	–
Japan	Oita	Pacific Hydrogen	1.4	1986	Yes	–
Canada	Monterial	Air Liquide	10	1986	Yes	–
Netherlands	Rosenburg	APCI	5	1987	Yes	–
France	Waziers, Lille	Air Liquide	10.5	1987	Yes	–
Japan	Minamitane	Japan Liquid Hydrogen	2.2	1987	Yes	–
Canada	Becancour Quebec	Air Liquide	11	1988	Yes	–
Canada	Magog, Quebec	BOC	15	1989	Yes	–
French Guyana	Kouru F.	Air Liquide	2.5	1990	Yes	–
Canada	Monterial	BOC	14	1990	Yes	12
Germany	Ingolstadt	Linde	4.4	1991	Yes	13.58
India	Mahendragiri	ISRO	0.3	1992	Yes	–
USA	Pace, FL	APCI	30	1994	Yes	–
USA	McIntosh, AL	Praxair	24	1995	Yes	–
China	Beijing	CALT	0.6	1995	Yes	–
USA	East Chicago, IN	Praxair	30	1997	Yes	–
Japan	Kimitsu	Air Products	0.3	2003	Yes	–
India	Saggonda	Andhra Sugars	1.2	2004	Yes	–
Japan	Osaka	Iwatani	11.3	2006	Yes	–
Germany	Leuna	Linde	5	2008	Yes	11.9
Japan	Tokyo	Iwatani	10	2008	Yes	–
Japan	Ichihara	Iwatani	5.1	2009	Yes	–
Japan	Shunan	Yamaguchi	5.1	2013	Yes	–
Japan	Harima	Kawasaki Heavy Ind.	4.2	2015	Yes	–
Germany	Leuna	Linde	5	2021	Yes	–
USA	La Porte, TX	Air Products	30	2021	Yes	–
China	Beijing	101 Institute	1.7	2021	Yes	–
India	India	Asiatic Oxygen	1.2	–	Yes	–
India	India	Andhra Sugars	1.2	–	Yes	–
USA	California	Stearns-Roger Mfg. Co.	62.5	–	–	–
USA	New Jersey	Air reduction Sales Co.	6	–	–	–
USA	Ashtabula, OH	Praxair	–	–	No	–

[a] SEC: specific energy consumption.

- **Ortho-hydrogen to Para-Hydrogen Conversion** As discussed in section 2.1, ortho-to-para conversion is an exothermic process that can lead to hydrogen evaporation. This transformation is a barrier for long-term liquid hydrogen storage and transportation. To reduce the energy loss, proper catalysts need to be used before storage (i.e., during hydrogen liquefaction process), which can force this conversion to accelerate. A higher concentration of para-hydrogen leads to less heat generation caused by ortho-to-para hydrogen conversion, and hence, less hydrogen loss during long-term storage and transportation.

- **Heat exchange with surrounding environment** Heat leak during storage is another cause of energy loss. Liquid hydrogen is stored under a very low temperature (~ -253 °C), which is far less than

ambient conditions. Thus, an effective cryogenic insulation system is needed. However, the thermal loss is unavoidable due to the heat transferred from the surrounding environment. Depending on the insulation quality and the surface-to-volume ratio of the storage tank, the boil-off gas generation can be on the order of 0.4% per day for a 50 m^3 cryogenic tank and 0.06% per day for a 20,000 m^3 liquid hydrogen tank [94]. The strategies for improving the efficiency of thermal management are very important in practical liquid hydrogen storage. First, to decrease the surface-to-volume ratio, large-size spherical or cylindrical adiabatic tanks are usually adopted for liquid hydrogen storage [96]. Second, to improve the insulation quality, the material selection for the storage tank should be optimised by using materials with low heat conductivity. Also, the

Table 7

Commercial hydrogen liquefaction plants worldwide under construction/planning [85,87,88].

Country	Location	Operated by	Capacity (tons/day)	To be onstream in
USA	Carson, CA	Air Products	10	–
USA	USA	Chart Industries	28	2022
USA	Las Vegas, NV	Air Liquide	30	2022
China	Zhejiang	Air products	30	2022
China	Beijing	FULLCRYO	1.5	2022
China	Shaanxi	–	8.5	–
Japan	Chubu Pref	Ituchu-Air Liquide	30	2025
ROK	Ulsan	Hyosung/Linde	35.6	–
ROK	Changwon	Doosan Heavy Ind./Air liquids	5	2023

double-wall structure with the vacuum space can help to reduce thermal convection and conduction, and multilayer insulation and variable density multilayer insulation can help to reduce the radiation heat transfer. In addition, self-evaporation vapor cooled shield (VCS) has been also introduced into such systems to enhance the storage effectiveness [97]. More details about the advanced thermal management methods for liquid hydrogen storage are discussed in Section 5.3.

- **Sloshing** Sloshing of liquid hydrogen may damage the tank wall and worsen the boil-off. Movement, such as acceleration and shaking, of liquid hydrogen inside of a tank or vessel caused by transport increases boil-off gas generation by (a) transferring heat to the tank system in the form of dissipated kinetic energy and (b) increasing the surface area between the liquid and vapor phases [98].
- **Flashing** Flashing of liquid hydrogen occurs during the transfer from a high-pressure tank to a low-pressure tank. This pressure difference leads to the evaporation of part of the liquid hydrogen.
- **Thermal stratification and overfill** When thermal leaks occurs in the storage tank, hydrogen evaporates and natural convection generated by buoyancy raises the pressure inside the tank [99]. The difference of liquid and gas densities makes a relatively stable liquid-vapor interface.

5.3. Liquid hydrogen storage

5.3.1. Materials selection for cryogenic application

Materials employed for liquid hydrogen storage must meet a series of special requirements, including resistance to hydrogen embrittlement, resistance to hydrogen permeation, mechanical strength, thermal robustness, and fire and heat resistance.

Hydrogen embrittlement refers to the cracking of metal from hydrogen exposure under both cryogenic and ambient temperature conditions. The hydrogen embrittlement characteristics are affected by temperature, pressure, and alloy strength [100]. While the main mechanism for hydrogen entrainment in the metal lattice occurs at the

atomic level [101], hydrogen embrittlement leads to macroscopic effects such as compromised material strength of steel alloys. Due to the lower hydrogen material penetration at low temperature, liquid hydrogen causes less hydrogen embrittlement than does gaseous hydrogen [29].

Stainless steels are the most widely used materials for liquid hydrogen storage. Stainless steels with different alloy components in varying grades have different properties and applications. For example, the 316 L stainless steel with added Molybdenum (Mo) has a higher resistance to chloride ion corrosion, making it more suitable for applications in marine regions. The 321 stainless steel with added Titanium (Ti) shows a relatively high performance in resistance to intergranular corrosion and high strength under high temperature, allowing it to be applied for heat resistance applications. In addition, there are also a number of studies focused on surface coating, cathodic protection, ion implantation and laser shot peening to improve the resistance of stainless steel against hydrogen embrittlement corrosion [26]. Table 9 lists several types of commonly used materials in cryogenic environments. Hydrogen permeation is a main barrier for application of lightweight composite materials.

5.3.2. Thermal management for liquid hydrogen storage

As discussed in Section 4.2, the temperature of the liquid hydrogen is as low as ~20 K, which indicates a large temperature difference relative to ambient temperature, and hence, heat transfer from surroundings is unavoidable. Thermal management is therefore a critical aspect of liquid hydrogen storage, especially for the long-term storage.

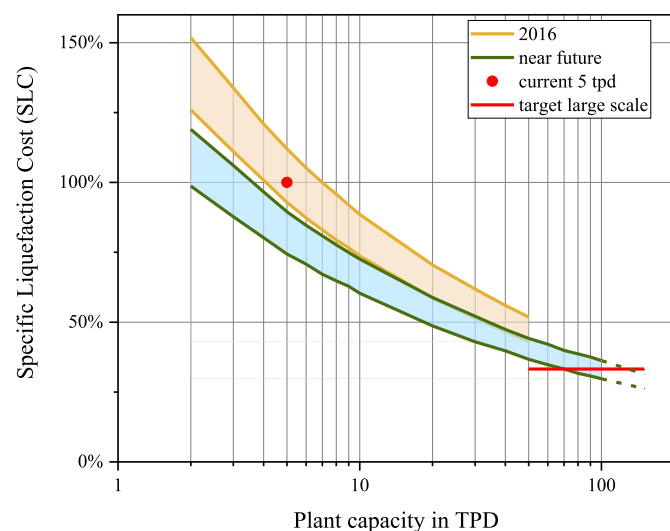##### 5.3.2.1. Shape and structure of storage tanks. Cryogenic tanks/vessels

Fig. 14. Future trend of specific hydrogen liquefaction costs (specific liquefaction cost of a 5 tons/day liquefier is used as a reference) (data collected from Ref. [47]).

Table 8

Future trend of hydrogen liquefaction (data collected from Refs. [47,90,91], (+) Strength (−) Weakness).

| Parameters	Today		Short to medium term	Long term
Liquefaction capacity	<3 tons/day	<50 tons/day	up to 150 tons/day	≥100 tons/day
Main refrigeration cycle	Brayton	Claude	High-pressure Claude	High-pressure Claude
Refrigeration medium	Helium	Hydrogen	Hydrogen	Hydrogen
Precooling cycle	Liquid nitrogen	Liquid nitrogen	Liquid nitrogen or mixed refrigerant	Mixed refrigerant
Feed pressure	10–15 bar	15–20 bar	20–25 bar	>20 bar
Compressor type	Reciprocating	Reciprocating	Reciprocating	Centrifugal
Specific energy consumption	>12.3 kWh/kg$_{LH2}$	>10.8 kWh/kg$_{LH2}$	7.7–10.8 kWh/kg$_{LH2}$	<9 kWh/kg$_{LH2}$
Investment cost (CAPEX)	++	O	−	−
Operating cost (OPEX)	−	O	+	++
CAPEX & OPEX	−	O	+	++
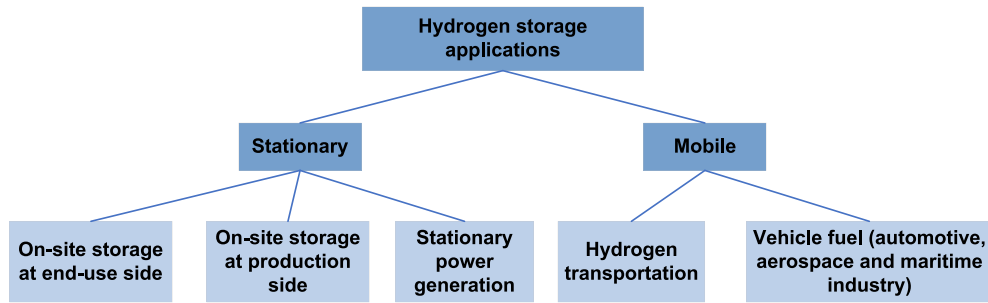
Fig. 15. Categories of liquid hydrogen storage based on its applications.

Table 9
Commonly used materials in liquid hydrogen storage (adapted from Ref. [26]).

Materials	Advantages	Challenges and research focus areas	Applications
Austenite stainless steel	Excellent hydrogen embrittlement resistance, good low temperature performance, weldability, and corrosion resistance	Low-temperature mechanical properties under specific environmental requirements need to be further improved	Widely used in liquid hydrogen storage and transportation, especially in ground liquid hydrogen storage and transportation
Aluminum Alloy	Lightweight, excellent formability, weldability, and good corrosion resistance	Optimisation of its welding method and low temperature mechanical properties after welding, corrosion resistance, and stress corrosion	Widely used in the space industry
Titanium Alloy	High strength, lightweight and low temperature performance	Advantages in forming and welding performance are not obvious, and the cost is relatively high	Initially used in the field of liquid rocket engines
Cryogenic Composites	Lightweight	Hydrogen permeation	Used in space industry for launch vehicles or in-orbit space trajectory or path adjustments

are a relatively mature technology and have been in use for more than 50 years across industrial applications. Vacuum insulated vessels are the most common type of cryogenic storage tank in use (see Table 10).

For the same volume of liquid hydrogen storage, a smaller surface area can result in less energy loss. Good insulation and low surface-to-volume ratio (such as for spherical tanks) can limit the daily boil-off to less than 0.1% [26]. A cylindrical vessel has a higher surface-to-volume ratio and therefore a larger surface area compared to a spherical vessel with the same volume. Moreover, to make the pressure well distributed, cylindrical vessels usually require thicker walls [103]. Therefore, spherical vessels are regarded as a better choice for liquid hydrogen storage. Fig. 16 shows a typical spherical liquid hydrogen storage vessel, which generally adopts double-walled structure with a high vacuum space in the middle to minimize heat convection and heat conduction [104]. Multilayer insulation materials are used to reduce heat transfer through radiation and convection.

5.3.2.2. Insulation technologies. In order to minimize the hydrogen boil-off, intensive research has been performed on both passive and active insulation technologies. The passive technology relies on the optimal selection of insulation materials. There are several good candidates for insulation materials, including multi-layer insulation materials (MLI, including reflector material such as aluminized polyester film and spacer materials such as nylon net), variable density multi-layer insulation (VDMLI), spray on foam insulation (SOFI), hollow glass microspheres (HGMs), perlite and aerogel [105,106]. MLI is the most common insulation method, which consists of low emissivity reflectors and low thermal conductivity spacers arranged layer by layer in an orderly way
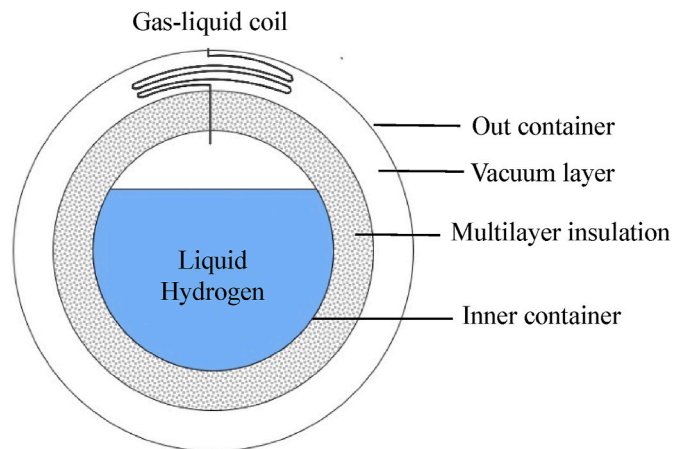

Fig. 16. Simplified structure of liquid hydrogen storage vessel.

under high vacuum. The thermal conductivity of MLI can be as low as 10^{-6}–10^{-5} W/(m·K). Based on the general MLI, variable intermedia density multi-layer insulation (VDMLI) has been proposed in recent years. To minimize the overall heat flow, the thickness of the spacer and the number of reflector layers are variable in VDMLI, which can further reduce the thermal conductivity by 10%–50% [107–109]. To solve the problem that the vacuum level gradually decreases with long-term transportation, the spray on foam insulation (SOFI, with thermal

Table 10
Number of different types of cryogenic vessels being used in some regions, 2012 [102].

Type of vessels	Units					
	Vacuum insulated			Non-vacuum insulated		
	Australia	Europe	US	Australia	Europe	US
Static vessels	2000	40,000	50,000	200	20,000	20,000
Small transportable vessels (no more than 1000 L)	3000	100,000	250,000	–	–	–
Large transportable vessels	200	5000	5000	40	1000	1000

conductivity of 10^{-3}–10^{-2} W/m·K) and hollow glass microspheres (HGMs, with thermal conductivity of 10^{-3}–10^{-4} W/m·K) have also been implemented as insulation materials for long duration liquid hydrogen storage due to their relatively high insulation performance at atmospheric pressure. In addition to simple use of one insulation material, numerical calculations and experimental studies have also been carried out on the composite systems of these thermal insulation materials [109–112]. Fig. 17 illustrates schematically the composite passive thermal insulation systems.

Besides the passive insulation method discussed above, some active technologies, such as vapor cooled shield (VCS), have been studied to improve the thermal insulation performance further. Compared with the heat of vaporization of hydrogen, the sensible heat of liquid hydrogen from the boiling point to 300 K (i.e., around ambient temperature) is much larger (more than 7 times) [106]. Thus, the self-evaporation vapor cooled shield (VCS) technology has been developed and commonly used, using sensible heat provided by the gaseous hydrogen from the tank to further reduce the boil-off loss without any additional power input. The additional adoption of VCS can further reduce the heat flux significantly (up to 50%) [97,113]. Considering the huge combustion heat of hydrogen, a fuel-cell-driven refrigerator was proposed to improve the insulation efficiency (up to 80%, compared with traditional MLI), as shown in Fig. 18 [106,107]. Liquid nitrogen was also proposed to be used for liquid hydrogen vessel cooling, because this approach can minimize the heat transfer between the liquid hydrogen and the atmosphere, and reduce the loss of liquid hydrogen greatly. Such a system with liquid nitrogen was reported to be capable of achieving zero boil-off for approximately 12 days of storage [49]. Furthermore, to realize zero boil-off liquid hydrogen storage, a cryocooler (i.e., a refrigerator designed to reach cryogenic temperatures for small-scall systems) can be implemented. The adoption of cryocooler combined with passive insulation technology is widely used by NASA for cryogenic propellant storage to improve the storage duration [114].

5.4. Liquid hydrogen transmission

As discussed in Section 5.1, the storage of liquid hydrogen can be divided into stationary storage and mobile storage. Mobile storage is used to transport liquid hydrogen from one place to another via road, railway or maritime.
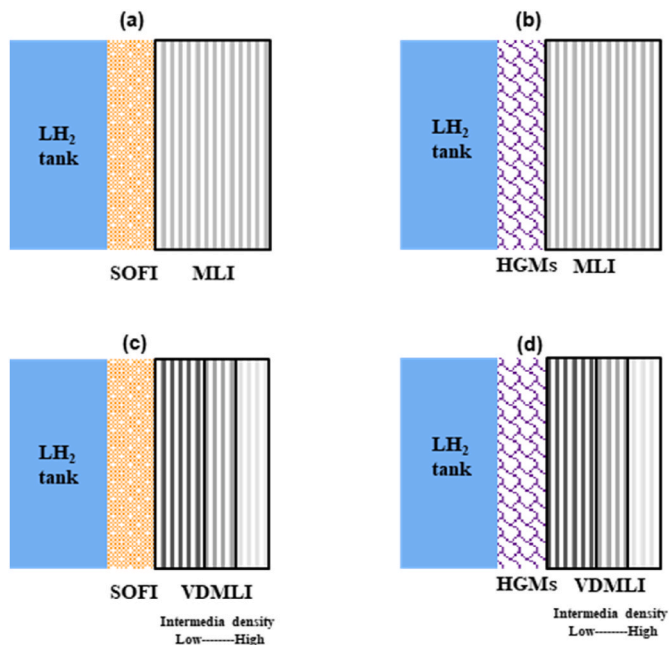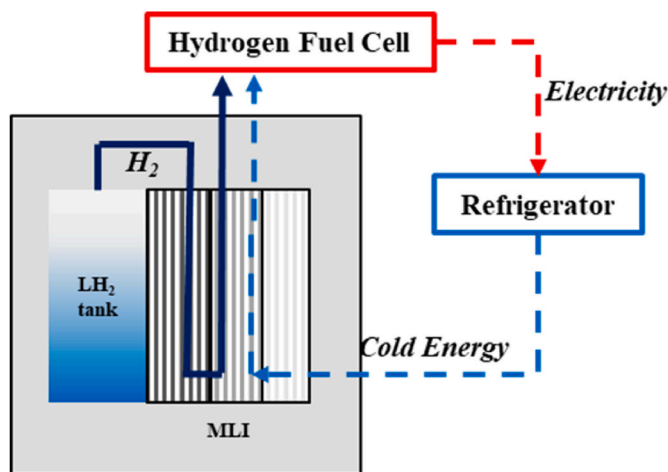
Fig. 18. Schematic diagram of a fuel-cell-driven refrigerator for liquid hydrogen storage.

When road transport is adopted, liquid hydrogen needs to be stored on board of road tankers (contained in the double walled tank). As mentioned before, the BOG formation is the main drawback of this storage and transport method, which impedes long distance shipments. Railway transport of liquid hydrogen is also not widely used due to a lack of liquid hydrogen tank cars availability and railway time scheduling factor that can increase the BOG formation [27]. Although maritime transport is suitable for very long-distance transport of large quantities of liquid hydrogen, the only hydrogen ship currently in existence is a 1250 m^3 pilot vessel demonstrated by the CO_2-free Hydrogen Energy Supply-chain Technology Research Association (HySTRA, established by four Japanese companies). This pilot hydrogen ship has completed its first operation in January 2022, loading its first cargo of liquefied hydrogen in Australia and returning to Japan (~9000 km) [115,116]. The most advanced liquid hydrogen carrier ship design so far is from Kawasaki Heavy Industries, and has a volume of 160,000 m^3 (four tanks of 40,000 m^3 each), which means a scale up of 64 times from the pilot is needed [116]. Liquid hydrogen transport methods are summarised in Table 11. One can also see that the maritime transport shows the largest load capacity of liquid hydrogen and the lowest BOG formation per day (<0.2 vol%). However, the BOG losses remain a major challenge for the long-duration storage and long-distance transport of liquid hydrogen. There are several approaches that can be implemented throughout the liquid hydrogen supply chain to reduce the negative impact of BOG losses.

- **Hydrogen liquefaction process:** using catalysts (e.g., iron hydroxides and chromium oxides) to accelerate the ortho-to-para conversion during hydrogen liquefaction, and hence reducing the boil off during the storage and transmission.
- **Liquid hydrogen storage:** adopting large tanks that have relatively low surface-to-volume ratios for liquid hydrogen storage during transmission (tanks with larger volume usually have lower evaporation rate [117]); using multi-layer insulation in combination with high vacuum, and actively cooled radiation shields for liquid hydrogen storage tanks [118]; pre-cooling cryogenic storage tank [119].
- **Liquid hydrogen transmission:** adopting an additional refrigeration unit during transmission [120]; using boil-off hydrogen gas as fuel for the ship.
- **Liquid hydrogen loading/unloading:** installing a compression and recovery system to compress and recover the boil-off hydrogen during loading/unloading [117].

Fig. 17. Schematic diagrams of passive thermal insulation systems of (a) SOFI-MLI, (b) HGMs-MLI, (c) SOFI-VDMLI, and (d) HGMs-VDMLI.

Table 11

Liquid hydrogen transport methods (adapted from Ref. [21]).

Transportation method	Transportation distance	Pressure	Hydrogen amount	Tank volume	BOG formation (per day)	Application example
Road	Mid-range distance [117]	≤7 bar	4 ton per truck	≤64 m³ [121, 122]	0.5 vol% [122]	Air Products [121] transports liquid hydrogen via liquid semi-trailers with a capacity of 12,000 to 17,000 gallons (45–64 m³).
Railway	>1000 km	≤7 bar	7 ton per rail car	105 m³ [122]	0.2 vol% [122]	National Renewable Energy Laboratory [123] estimated that LH₂ rail delivery cost is likely to be lower than the cost of CGH₂ and LH₂ trucks/pipelines delivery for long-distance and large-scale application.
Maritime	Transoceanic delivery	≤7 bar	60 ton per tank	1250–40,000 m³ [116]	<0.2 vol% [117]	A pilot-scale liquid hydrogen supply chain between Australia and Japan (HySTRA Project [116], 1250 m³ ship) has been completed in 2022.

6. Liquid hydrogen regasification

Liquid hydrogen regasification is the process by which liquid hydrogen is returned to its natural (gaseous) state. Compared with hydrogen liquefaction and liquid hydrogen storage/transport, liquid hydrogen regasification consumes relatively much less energy. The liquid hydrogen can be simply re-gasified and heated up by heat exchange with seawater or air. This process can be done in open rack vaporisers, shell and tube vaporisers, or intermediate fluid vaporisers with some additional cost due to the cryogenic conditions and corrosivity of seawater. If the hydrogen needs to be transported/distributed to user site via transmission pipelines eventually, the low-pressure liquid hydrogen (~1–4 bar) can be pressurized to the required pipeline pressure (~70–100 bar) via cryogenic pumps before the regasification process. It is much easier and more energy-saving to pump liquid hydrogen to high pressure than to compress gaseous hydrogen to the same pressure [116]. Another option to achieve the high pressure is to just re-gasify the liquid hydrogen at a constant volume, which can avoid the use of pumping equipment.

However, for liquid hydrogen regasification, the most challenging part is the recovery of the high-grade cold energy released during liquid hydrogen evaporation. During regasification, all the energy that was put into the hydrogen to make it cold and liquefied is released, and in most cases, wasted. There is a great need for cryogenic energy recovery measures to avoid or mitigate this waste, however, these measures have not been adequately researched and applied. Therefore, lessons can be learned from the cold recovery of LNG regasification processes and potentially applied to hydrogen (see Fig. 19).

Some of these cold energy recovery methods have been applied or demonstrated in real LNG terminals, including power generation, air separation, traditional desalination, and cryogenic carbon dioxide capture. Therefore, these measures are more mature and more likely to be replicated technologically at liquid hydrogen terminals. As the liquid
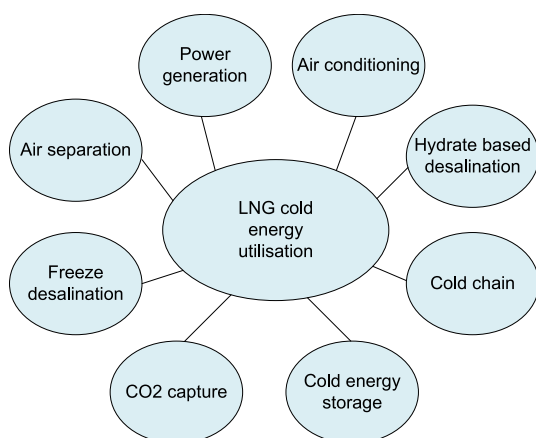
hydrogen market grows, the remaining as yet unproven methods of LNG cold energy recovery/utilization, e.g., air conditioning (data centre cooling), hydrate-based desalination, cold chain transportation, cold energy storage etc., are also potential candidates for future use in liquid hydrogen terminals. However, it must be stressed that, despite several applications, most of the high-grade cold energy recovery methods described above have not yet been applied on a large scale. Most of the existing LNG terminals still use seawater to heat LNG directly without any cold recovery facilities [116]. The main obstacles lie in the distance from the waste cold sources to the potential users (a cost-effective way to transport the heat transfer fluid is needed), fluctuating cold demand, and related factors [125]. These obstacles are all the more challenging for hydrogen regasification terminals due to their lower temperature of operation and hence, more R&D efforts are needed.

7. Emerging opportunities for liquid hydrogen – drivers and barriers

As discussed in Section 3.2, although liquid hydrogen as a hydrogen storage technology in the value chain has so far shown to be almost the least cost effective, there are important opportunities for the liquid hydrogen storage technology in the hydrogen economy. Because of the high energy density, liquid hydrogen fuels have been studied and practiced in the aerospace, maritime, aviation (medium- and long-haul flights), and road transport (cars, trucks and trains) sectors. Emerging opportunities also exist for liquid hydrogen, particularly in long-distance energy transmission.

Transmission of hydrogen via long-distance maritime routes is considered a primary potential application for liquid hydrogen technologies, which would connect countries and regions with low-cost renewable energy sources (RES) with locations with high industrial energy demand but limited availability of low-carbon energy sources [126]. Relevant potential market connections in many cases overlap with existing shipping corridors for fossil energy supply chains, albeit the capacity to implement such hydrogen trade requires additional development of necessary infrastructure [127].

7.1. Comparison with other liquid-phase hydrogen carriers

In addition to liquid hydrogen, LOHCs [128] and ammonia [129], as liquid-phase hydrogen carriers, are also two very promising candidates for the long-term and long-distance hydrogen storage and transmission.

Liquid organic hydrogen carriers (LOHCs) are posited as a solution that can be used not only for long-term storage under ambient conditions, but also for long-distance transmission of hydrogen and applications in maritime supply chains [130]. Compared with transmission and storage in other physical states, LOHCs are considered potentially cheap, safe, and easily managed [131], as issues related to boil off gases, high pressure, extremely cryogenic temperatures, and reversibility of hydrogen storage are greatly diminished. The basic principle of a LOHC as a medium for hydrogen storage and transmission is the coupling of two catalysed reversible reactions (i.e., hydrogenation and

Fig. 19. Current and potential applications for LNG cold energy recovery (adapted from Ref. [124]).

Table 12

Comparative technical analysis of liquid-phase hydrogen carriers (i.e., liquid hydrogen, LOHCs and ammonia).

Assessment indicators		Liquid hydrogen	LOHC (MCH)	Ammonia
Technology maturity[a]	Conversion	Hydrogen liquefaction Small scale: + Large scale:	Hydrogenation: O	Haber-Bosch process: +
	Reconversion	Liquid hydrogen regasification: +	De-hydrogenation: O	Ammonia cracking: O
	Tank storage	O→+	+	+
	Transport	Truck: + Ship: O→+	Truck: + Ship: +	Truck: + Ship: +
	Supply chain integration	O→+	O	+
Conversion and reconversion total energy consumption[b]		Current stage: 25–40% LHV_{H2} Potential: ~18% LHV_{H2}	Current stage: 35–40% LHV_{H2} Potential: 25% LHV_{H2}	Conversion: 7–18% LHV_{H2} Reconversion: <20% LHV_{H2}
Hazards[c]		Flammable; no smell and flame visibility	Toluene; flammable; moderate toxicity (other LOHC can be safer)	Flammable; acute toxicity; precursor to air pollution; corrosive
Technology improvement needs		Hydrogen liquefaction plants efficiency; boil-off management; cryogenic energy recovery during regasification	Reconversion efficiency; conversion heat recovery	Conversion efficiency; hydrogen purification after ammonia cracking;
Companies/organisations developing supply chain		HySTRA; CSIRO; Air Liquide; Fortescue Metals Group	AHEAD; Chiyoda; Hydrogenious; Framatome; Clariant	Green ammonia consortium; IHI corporation; US department of Energy

[a] +: high technology maturity (proven and commercial), O: medium technology maturity (prototype demonstrated), -: low technology maturity (validated or under development); small scale: <5 tons/day, large scale: ≥100 tons/day.

[b] Given as a percentage of the lower heating value of hydrogen (values are for high-purity hydrogen that can be used in fuel cells).

[c] Toxicity criteria are based on inhalation.

compressed hydrogen in long-distance transport.

As for LOHCs and ammonia, unpacking costs (dehydrogenation equipment and high energy requirements) are significant, but transport and storage costs are relatively minor, which suggests that these two hydrogen carriers are more suitable for very long-distance transportation. Boil-off losses of liquid hydrogen increase with distance, reducing its competitiveness with ammonia and LOHC for very long-distance shipment. It can also be seen that other factors must be considered when choosing the optimal hydrogen transport mode. For instance, if waste heat can be used at end-user side for LOHC dehydrogenation, its unpacking costs can be reduced obviously, making it much more competitive for hydrogen delivery.

7.2.2. Roadmap of liquid hydrogen in long-distance transport

The most essential way to enhance the opportunities for liquid hydrogen and to realize its cost advantage in long-distance maritime transport is to achieve high hydrogen liquefaction efficiency, low boil-off rate, suitable cryogenic energy recovery measures, and adequate scale of production. At the same time, some challenges need to be addressed first for the long-distance transport of liquid hydrogen in the future hydrogen economy.

(1) Technical challenges: Maritime transport is clearly one of the most suitable methods for realising long-distance hydrogen transport. A pilot liquid hydrogen transport vessel demonstrated by HySTRA (see Section 5.4) uses diesel as fuel. This is not an intended long-term solution and ships powered by hydrogen provides a potential solution. Internal combustion engines and fuel cells could be used, but efficiency of the former is low. In addition, hydrogen fuel cells, which have been proposed for the utilization of boil off gas from hydrogen carriers [137], have so far only been demonstrated for some small scale ships (e.g., ferry, pusher) with up to 600 kW of power. The currently reported maximum power of the fuel cells is approximately 2 MW, which is far smaller than the minimum power requirement of a commercial liquid hydrogen ship (~5 MW) [116].

(2) Regulations and standards: Hydrogen is not currently included in the IGF Code (International Code of Safety for Ships using Gases or other Low-flashpoint Fuels) or IGC Code (International Code for the Construction and Equipment of Ships Carrying Liquefied Gases in Bulk). The application process of IGC started in 2014, with the interim guidelines for carrying hydrogen being adopted in 2016 (HySTRA) [116]. 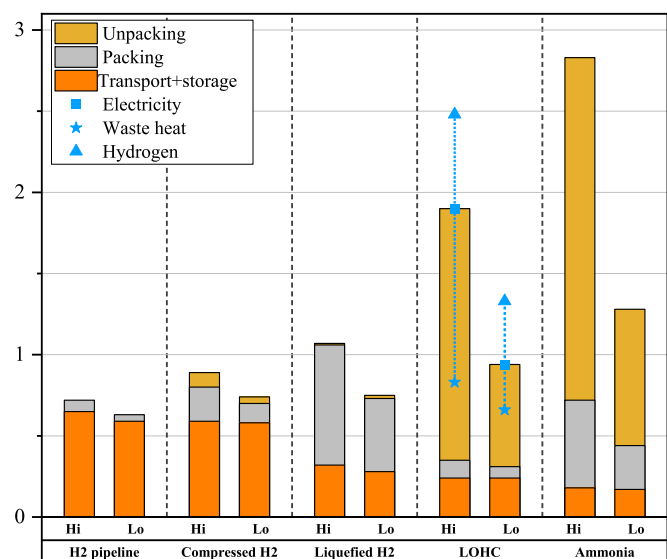As a reference, LNG took six years between the first interim guidelines and the final adoption of the IGC code. For the IGF code, Lloyd's Register has given approval for hydrogen use in fuel cells for small ships, but not for large ships to date [116]. American Bureau of Shipping (ABS) has published a Hydrogen as Marine Fuel Whitepaper, which covers further international and national standards on the design liquid hydrogen carriers [138].

(3) Infrastructure: To establish and develop the supply chain for liquid hydrogen via maritime transport, new facilities need to be developed. Large-scale hydrogen liquefaction plants will need to be constructed. Some existing LNG plants could be modified/retrofitted to save initial investment cost. In addition, bunkering facilities, and transport equipment to and from the ports, large liquid hydrogen tanks at terminals (which could also be modified/retrofitted from LNG storage tanks), other terminal components (e.g., piping manifold and the loading arms), safety measures, metrological aspects, and ships will also require significant investment and construction.
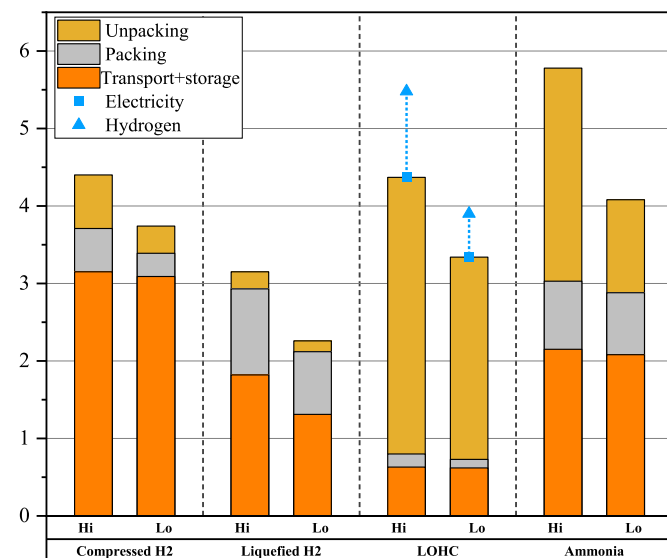
8. Summary and conclusions

This article has reviewed technological progress of the entire liquid hydrogen supply chain, including hydrogen liquefaction, liquid hydrogen storage and transmission, and liquid hydrogen regasification, from both research and industrial applications perspectives. Opportunities of liquid hydrogen in the future hydrogen economy and energy market, and the pathways to seize these opportunities are discussed based on the review. It is found that the key factor limiting the potential use of liquid hydrogen as a primary means of hydrogen storage and transmission is the very high energy penalty due to high energy consumption of hydrogen liquefaction (13.83 kWh/kg_{LH2} on average) and high hydrogen boil-off losses that occurred during storage (1–5 vol% per day). A number of technical approaches could be used to address or mitigate these challenges.

(1) High energy consumption of hydrogen liquefaction processes: The use of upgraded refrigeration/pre-cooling cycles (e.g., high-pressure Claude cycle and mixed refrigerant cycle), more suitable refrigeration media (e.g., mixed refrigerants), improved efficiency of key components (e.g., compressors), and scaling up of liquefaction plants (≥100 tons/day) are the most promising ways to bridge this gap and to reach a target of 6 kWh/kg_{LH2}.

(2) High liquid hydrogen losses during storage: Most efforts made in liquid hydrogen storage, including the optimisation of shape/

(a)

(b)

Fig. 22. Potential costs (2030–2035) of hydrogen delivery for different storage methods under (a) scenario 1 - single end user with 1 MtH$_2$ per year and (b) scenario 2 - more distributed delivery with 0.1 MtH$_2$ per year. (Cost unit: EUR/kg$_{H2}$, Hi: high electricity price, Lo: low electricity price) (data collected from Ref. [44]).

Fig. 23. Potential costs (2030–2035) of clean hydrogen delivery under different storage methods against transport distance in single end-user scenario (1 Mt/H$_2$ per year) [44].

to increase the cost of delivering liquid hydrogen, and hence cost must be balanced against performance.

With the two major issues above resolved or alleviated, liquid hydrogen will provide further opportunities as a key means of long-distance energy transmission (transport distance ≥2000–3000 km) due to cost advantages. The most obvious advantage of liquid hydrogen over other liquid-phase hydrogen carriers (i.e., LOHCs and ammonia) is its much simpler reconversion process (regasification process), which requires less investment and consumes less energy at the end-user side.

Importantly, the ultimate role that liquid hydrogen will play in the emerging hydrogen economy depends on developments throughout the entire hydrogen sociotechnical system. Countries such as Japan and South Korea with significant ship building industries will see an opportunity in developing liquid hydrogen maritime transport, and hence are expected to continue to further pursue their efforts in establishing liquid hydrogen as an important part of their hydrogen economies and in the development of hydrogen trade relations with countries such as Australia that are eager to establish hydrogen trade with Northeast Asia. As such, significant efforts are needed to address the technical challenges building and certifying hydrogen vessels (fuel cell-powered hydrogen ships), making and refining regulations and standards for the mass transportation of liquid hydrogen, and implementation of large-scale infrastructure for storage and transport across entire supply chain. Far from being a remote possibility, however, such developments are underway. Hence, the considerations outlined in this review are expected to contribute to a long-term knowledge base that will underpin the global hydrogen economy evolution.

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.

Data availability

No data was used for the research described in the article.

Acknowledgments

structure of storage tanks as well as thermal insulation systems, are aimed at reducing boil-off rates (target: below 1 vol%, or even 0.1 vol% per day). In addition to the measures taken for storage, there are also various other methods that can be implemented to mitigate boil-off problems throughout the entire liquid hydrogen supply chain, from hydrogen liquefaction (e.g., using ortho-to-para conversion catalysts), transportation (e.g., adopting additional refrigeration units, using boil-off hydrogen as fuel) to loading/unloading (e.g., adopting additional equipment to recover boil-off hydrogen). However, all these methods are likely
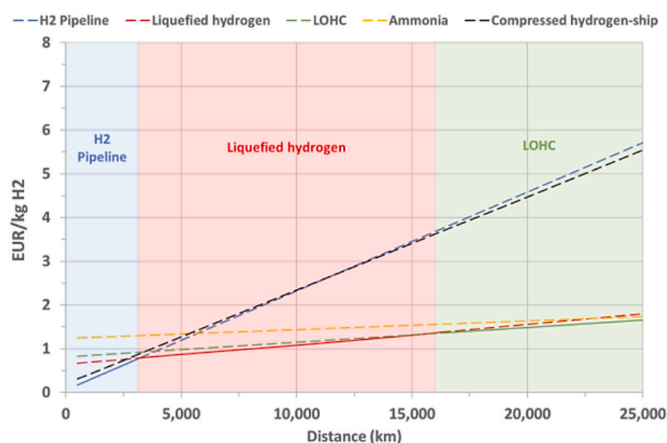
European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No. 101007976.

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