Brief Review and Technical Insight of Liquefied Hydrogen Carriers Development

Wei Song, Zilong Zhu(✉), Zhong Wan, Chan Long, and Tao Liu

Hudong-Zhonghua Shipbuilding (Group) Co., Ltd, Shanghai 200129, China
zzlong_seu@sina.com

Abstract. Countries worldwide are shifting to a hydrogen economy in response to stringent environmental regulations, and hydrogen transport between countries is expected to increase in the medium to long term. Although hydrogen is traded between countries in different forms such as ammonia, liquid hydrogen, and methanol, from the perspective of volume density and production/demand area without a separate process, the transportation of hydrogen in liquid form is the potential way for large-scale transportation of hydrogen in the future. This article aims to highlight the opportunities and challenges technical for the ocean-going liquid hydrogen carriers. An overview of development state-of-the-art and key technical challenges of liquid hydrogen carrier ships are summarized, including regulation, the cargo containment structure and insulation, boil off ratio (BOR) evaluation, boil-off gas (BOG) handling system and propulsion system. Finally, detailed technical route of the key technology required by future liquid hydrogen carrier is extrapolated, and securing a possible design through various technological alternatives.

Keywords: Liquid hydrogen carrier · Insulation · Boil off ratio · BOG handling · Propulsion system · Technical insight

1 Background and Motivation

With the strengthening of international environmental regulations and the geopolitics of the Russia-Ukraine war, the energy market will undergo rapid changes in the future. The Paris Agreement adopted on December 12, 2015 is not only a substitute for emotion in Kyoto, but also a consensus of the international community on greenhouse gases (GHGs) [1]. Most advanced and developing countries have participated in the agreement, 186 countries put forward the goal and contribution plan of reducing greenhouse gas emissions. Investment in new renewable energy has being increased in recent years, but the fluctuations in new renewable energy production and difficulties in trading renewable energy resources across countries are considered the biggest hurdles obstacles to the utilization of renewable energy. To break through the limitations of this new renewable energy, hydrogen energy has attracted attention. Hydrogen uses fuel cells to generate carbon free electric energy and heat energy that can be easily converted into renewable energy [4]. Hydrogen energy produced by water electrolysis with renewable energy is

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best solution to balance the problem of renewable energy fluctuation and imbalance in various countries. China is today the largest hydrogen consumer in the world, at about 24 MtH_2/year in 2020 [5].

Niermann et al. investigated hydrogen exports from Algeria to Hamburg [6] and analyzed the feasibility of different liquid organic hydrogen carriers (LOHC), pipeline transportation and bulk transportation of liquid hydrogen. In a similar study, chain energy efficiency and costs for ammonia and LH_2 sea transport from northern Norway to Rotterdam and global (Tokyo) were estimated by Ishimoto et al. [7]. The energy efficiency and life-cycle costs of energy transported by submarine high-voltage cables are compared with pipeline hydrogen transport, compressed hydrogen ship and liquid hydrogen with different distances in [8]. Hydrogen can be transported by ship in the form of LOHC, ammonia or liquefied hydrogen and ammonia carriers have been commercialized and widely used in industrial. For LOHC carriers, ordinary chemical carriers can be used, so it is not necessary to further develop new technologies [9]. The above three recently published overlap papers, they all agree that liquid hydrogen (LH_2) is the most promising option for long-distance seaborne hydrogen transport. Coincidentally, the liquefied hydrogen transport ship aims to the demonstration of liquefied hydrogen transport between Australia and Japan. At the end of 2019, Kawasaki Heavy Industry of Japan built a world's first liquid hydrogen carrier with two 1250 m^3 double-shell vacuum liquid hydrogen storage tank, which horizontal cylindrical pressure vessel freely enable thermal shrinkage for transporting LH_2 [10].

Several economic analyzes on the hydrogen supply chain including maritime transportation of liquefied hydrogen have been performed, but few studies has been done on the technical characteristics of liquefied hydrogen carriers. Although Japan has completed the construction of liquid hydrogen ships, this is only an experimental attempt. Therefore, this is a comprehensive and difficult task, because the volumes are vast and data sets, and the necessary methodology statement may not be fully available. A comprehensive comparison of theories and assumptions, methodological choices and levels of technical abstraction is beyond scope of this paper. Therefore, we review and discuss the most obvious differences in the existing liquid hydrogen carrier or conceptual design assumptions, and extrapolate the preliminary technical appearance of the future liquid hydrogen ship. While identifying detailed technologies necessary for the development of liquefied hydrogen carriers in the future and securing various technical alternatives, the technical feasibility of liquefied hydrogen carriers is analyzed.

2 Classification of Liquid Hydrogen Carrier Tanks

The capacity of the liquefied hydrogen carrier depends on the economics of the liquefied hydrogen supply chain which have a significant impact on the analysis. Therefore, referring to the LNG ship type, the possible capacity of each tank type of the liquefied hydrogen carrier is analyzed. As shown in Fig. 1, membrane and type B tanks are applied to large LNG carriers, and Type C tanks are mainly applied to small LNG carriers or bunkering ships.

The Type C tank has the advantage of higher design pressure and ability to store BOG in the tank due to its relatively pressure build-up (accumulation) system locking

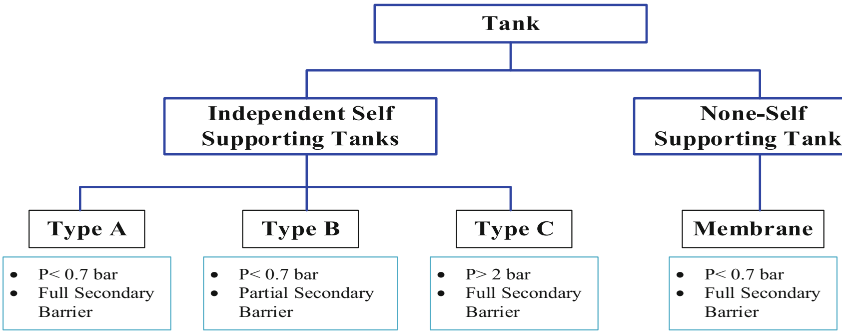

Fig. 1. Classification of cargo tanks for ships

BOG into the cargo tank, but has the disadvantage of being difficult to enlarge due to its shape and having low space efficiency. Hyundai Heavy Industries Group completed the conceptual design of a 20 K class liquid hydrogen carrier and received Approval in Principle (AIP) certification from the Korean Register of Shipping. Based on this, it is expected that it will be possible to manufacture up to 5000 ~ 6000 m^3 per type C tank, and it is judged that the capacity of the liquefied hydrogen carrier can be up to about 20 K depending on the number of tanks.

Membrane type tanks have high space efficiency and can be manufactured with a capacity of 160 K or higher based on LNG carriers. However, since this is a judgment from the viewpoint of manufacturability of the tank, it is also necessary to consider the possibility of applying an appropriate insulation system according to the capacity.

3 State of the Art and Extrapolations of Hydrogen Tanks Boil off Ratio

3.1 Type-C Tank

Liquid hydrogen storage and handling technology was first developed and applied by NASA aerospace projects [11]. The world's largest spherical LH_2 storage tank with approximately 3200 m^3 LH_2 capacity was built in the 1960s. The absolute value of the boil off gas is reported about 530 gal/day, which corresponds to boil off ratio of approximately 0.0625% per day [12]. Different types of LH_2 tanks have been designed and brought to market by Linde, the BOR of which is decided by size, shape, insulation, environment and usage pattern. For example, a cylindrical tank with a capacity of 300 m^3 has a boil off ratio of 0.3% per day, while a spherical tank with a capacity of 1100–2300 m^3 has a boil off ratio of less than 0.1% per day [13].

In the 1980s KHI built spherical LH_2 tanks with a volume of 600 m^3 and 540 m^3 LH_2 capacity which achieves a boil off ratio of 0.18% per day [14]. After 30 years of operation, no degradation in insulation performance has been detected by KHI [15]. By 2020, 1250 cubic meters of horizontal cylindrical seaborne tank have been put in operation [16], the spherical onshore terminal tank achieves thermal insulation performance with $\leq$ 0.1% per day boil off ratio [17]. Recently, a basic principle design of 11,200 m^3 volume spherical LH_2 tank with $\leq 0.1\%$ per day boil off ratio performance have been completed by KHI and certified by the classification society [18].

Most hydrogen storage vessels are double-layered with a vacuum in the middle. The space between these can also contain other materials, such as aluminum-coated polyester sheets, alternating layers of aluminum foil as well as fiberglass. The vacuum aims to reduce losses by conduction and convection, while the alternated layer aims to reduce losses by radiation [19]. Heat loss can also be reduced by reducing the ratio of the exposed surface to the volume of the tank, which is the reason why spherical tanks are often used to store the liquid hydrogen. That can be seen as a trade-off with cost. Although the surface-volume ratio of cylindrical tanks is higher than that in spherical tanks, they are easier to manufacture due to the low costs, thus making them more common. The boil-off ratio depends on the size of the tank and the intended pattern of use. For example, a 300 m^3 small tank has boil off ratio of 0.3% per day, while tanks of 1100–2300 m^3 can already achieve boil off ratio of less than 0.1% per day [20].

Based on the brief review above, it is reasonable to assume that low boil off ratio can also be achieved when LH_2 cargo tanks are sized close to the capacity of today's Liquefied Natural Gas (LNG) carriers. Since boil off ratio of 0.06–0.25% per day is already achieved for tanks between 100 and 4000 m^3, a natural question is that whether it will facilitate or impede achievable performance of low boil off ratio if further scaling-up tanks volume. Under the assumption of well insulated tank with a uniform temperature distribution inside, the heat flow into tank LH_2 will proportional to the difference between the ambient temperature and the LH_2 temperature (K), overall heat transfer coefficien t(W m^{-2} K^{-1}) and the tank surface area (m^2), as shown in Eq. (1).

$$Q_{\text{in}} = (T_{\infty,ambient} - T_{\infty,\text{LH}_2})U_{\text{overall}}A_{\text{surface}} \tag{1}$$

The boil off ratio is defined as the ratio of the amount of evaporated gas produced per unit time to the full tank inventory, but [% per day] is a more commonly used unit for cryogenic storage tanks. Qin is heat (kW), V_{tank} is volume (m^3), ρLH_2 is the density of liquid hydrogen (kg/m^3), Levap,LH_2 is the latent heat of vaporization (kJ/kg), A_{surface} is the surface area (m^2), and U_{overall} is overall heat transfer coefficient (W m^{-2} K^{-1}). The boil off ratio (% per day) can be expressed as:

$$BOR(\%/day) = \frac{Q\text{in}}{L_{\text{evap,LH}_2} \cdot \rho_{\text{LH}_2} \cdot V_{\tan \text{k}}} \cdot 24 \cdot 3600 \cdot 100\% \tag{2}$$

where the units in the equation are international standard.

From (1) and (2), it can be found that the boil off ratio is proportional to the ratio of surface area to volume, which is usually called specific surface area. For spherical and cylindrical tanks, Fig. 2 gives how the specific surface area changes with size. It can be observed that it will decline sharply with the increase of trading volume, while on the other hand, it will rise sharply for sufficiently low trading volume.

To further illustrate the effect of size on the boil off ratio at the achievable design point, Fig. 3 shows the change of daily the boil off ratio with the total heat transfer coefficient and spherical tank volume. The ambient temperature is set at 288 K. For simplicity, it is assumed that the storage tank contains 1.2 bar (a) of pure saturated liquid to hydrogen, and the air-fuel rate is 10%. The liquid density and temperature can be

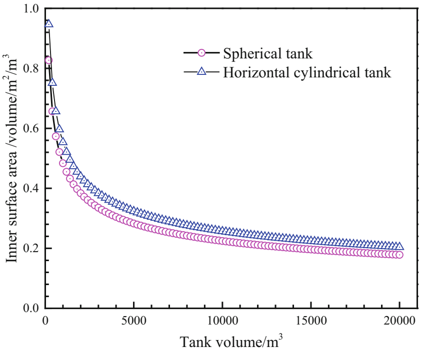

Fig. 2. Relation between internal surface area and volume for cylindrical and spherical tanks

retrieved respectively from the thermophysical properties in REFPROP [21], and the specific heat of evaporation (h_{evap}. LH_2) is 443.17 kJ/kg. For any fixed value of U, the daily the boil off ratio decreases significantly with the increase of tank size. The daily boil off ratio decreases by about 54% when the volume increases by 10 times (e.g. for 1000 m^3 relative to 100 m^3). Apart from these correlations based on equation, the rough data of three spherical tanks existed of different sizes and vintage [22], and the tank performance of other two indicators [13, 18] are also plotted in the same chart. Reference [6–8] is also included. The estimated value of the total heat transfer coefficient U of each tank can be read from the curve intersected with the data point, and it indicates that [12, 13, 17] is about 0.004 W m^{-2} K^{-1}. In theory, if the storage tank [13, 14] or [17] can be further scaled with U unchanged, the boil off ratio would be reduced to about 0.07% per day with a volume of 10,000 m^3. The corresponding figure of [22] would eventually reach about 0.17% per day.

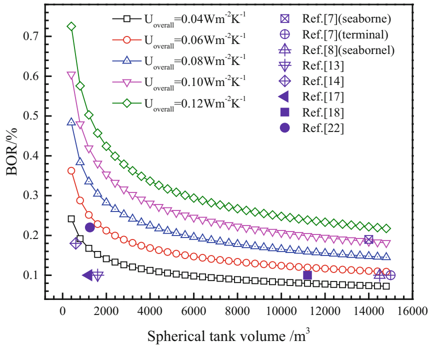

Fig. 3. Evaluation for the relation between spherical LH_2 tank volume, overall heat transfer coefficient and daily BOR.

As these illustrative examples show, due to the reduction of surface-to-volume ratio, the increase of size and diameter is usually beneficial to the low-voltage insulation system

[23]. It is still to be identified what is the best technical and economic boil off ratio of large LH_2 transport carriers in the future [24], and its size is equivalent to the current LNG tanker. This is an overall research and development task. It is necessary to weigh tank design and thermal insulation layout with a series of trade-offs, some of which may relax some restrictions, such as balancing boil off ratio and energy demand of propulsion and auxiliary systems. Multidisciplinary capabilities at different levels are required, including construction and materials technology, thermodynamics and fluid dynamics, mass and heat transfer, thermal process, naval architecture, and power and propulsion systems. In the research project, "LH_2 Pioneer—Super Insulated Marine Containment System for Global LH_2 Ship Transportation" led by SINTEF, LH_2 containment and thermal insulation, cargo loading and marine hydrogen re-liquefaction process are the key research topics [25].

3.2 Membrane Type Cargo Tank

Therefore, in order to determine the thermal insulation performance of membrane tank insulation, it is assumed well insulated membrane cargo tank with a uniform temperature distribution both inside and outside of insulation layer. Excluding insulation mounting members, piping, support members and cargo handling systems, only the thermal insulation and heat transfer around cargo tank which volume is assumed to be cubic are considered. The heat flowing into the cargo tank will be proportional to the difference between the outer surface temperature of the insulation layer and the LH_2 temperature (K), the outer surface area and the thermal conductivity, as represented in the Eq. (3).

$$Q_{\text{in}} = \lambda \cdot \frac{(T_{\infty,\text{out}} - T_{\infty,\text{LH}_2})}{D_{\text{thickness}}} \cdot A_{\text{surface}} \tag{3}$$

$$BOR(\%/day) = \frac{Q\text{in}}{L_{\text{evap,LH}_2} \cdot \rho_{\text{LH}_2} \cdot V_{\tan\text{k}}} \cdot 24 \cdot 3600 \cdot 100\% \tag{4}$$

This paper only considers heat conduction for the outer surface temperature of the insulation layer of 0 °C and the inner temperature of − 253 °C, the volume of the cargo tank varies from 1000 to 60,000 m^3, and the thickness of the insulation layer is between 0.5 and 2.5 m. Due to the different areas of the hot and cold ends, the average value of the insulation outer area is considered as the outer area of the cargo volume for the conduction heat input calculations and following calculation formula is shown in (4). $D_{\text{thickness}}$ and λ represent the thickness (m) of the insulation layer and thermal conductivity (W m^{-1} K^{-1}).

Through the summary of the above literature, it is reasonable to assume that the boil off ratio of the liquid hydrogen cargo tank which is supposed to cube cargo tank is set at 0.2% per day. Figure 4 shows the requirements for the thermal conductivity of the thermal insulation layer in liquid hydrogen ships with different capacities and thicknesses of the thermal insulation layer. As mentioned above, since this is a calculation for a fully enclosed cargo tank that does not include other heat input components, it is necessary to apply an insulation with a lower thermal conductivity than that shown in this result when other heat leaking components are considered. It is obvious from the figure that with the increase of the volume of the cargo hold, the thermal conductivity gradually increases but its increase rate gradually decreases, which means that when the volume of the cargo tank reaches a certain value, the benefits obtained by improving the thermal insulation performance of the insulation material are significantly reduced and the benefits are more pronounced with the thickness of the insulation. However, increasing the thickness of the insulation layer will affect the usable space of the cargo tank and bring about the problem of lower loading economy. Therefore, this is a pair of contradictions and need to find the best balance point.

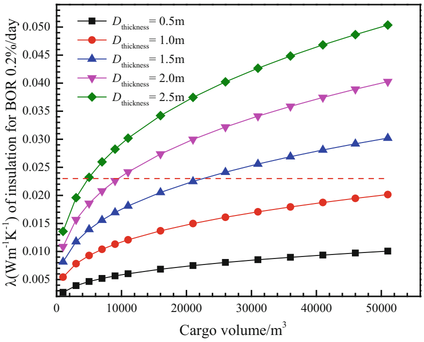

Fig. 4. Thermal conductivity of thermal insulation materials required for liquefied hydrogen tanks with different volumes

Table 1 shows the conductivities of insulation materials applicable to tanks of cryogenic cargo which are commonly used in current industry. Among the proposed materials, VIP is the only material with $\lambda < 0.01$ W m^{-1} K^{-1} and MLI exists as a better heat-insulating property than VIP. But considering the vacuum structure transformation of the membrane insulation system, it is judged to be technically difficult to apply.

Table 1. Thermal conductivity of insulation material

Insulation material	Thermal conductivity (W m^{-1} K^{-1})	Source
Mineral wool	0.035–0.045	[26]
Polyurethane	0.017–0.024	[26]
Expanded polystyrene	0.035–0.04	[26]
VIP	0.002–0.008	[26]
Glass bubble	0.047–0.2	[27]

When VIP is applied to the inter barrier space (IBS) of the liquefied hydrogen membrane tank, the temperature is lower than that of the IBS of the existing LNG cargo tank, so there is a difference in operating concept. The IBS of the existing LNG cargo tank was operated in the form of nitrogen purging, but the IBS of the hydrogen cargo hold cannot perform nitrogen purging because there is an area where the temperature is lower than the freezing point of nitrogen (– 210 °C). A method of applying helium instead of nitrogen is also possible.

It is possible to apply a vacuum to the IBS space, but it is necessary to develop and verify the technology for applying/maintaining a vacuum in a large space. In the case of an LNG membrane tank, a vacuum of – 800 mbar is applied through a global test to confirm the airtightness of the primary barrier, so it is expected that a certain level of vacuum application is structurally possible. However, in a liquefied hydrogen carrier, it is necessary to verify whether the vacuum can be maintained during the life cycle of the ship and whether freezing of nitrogen or the like occurs at the corresponding level of vacuum. When vacuum is applied to the IBS, the effects of air condensation and oxygen enrichment due to air inflow in case of vacuum loss should be considered. In addition, the correlation between the reduction in vacuum and the amount of BOG generated due to VIP aging should be considered. Figure 5 is an example of an insulation system arrangement for a membrane tank.

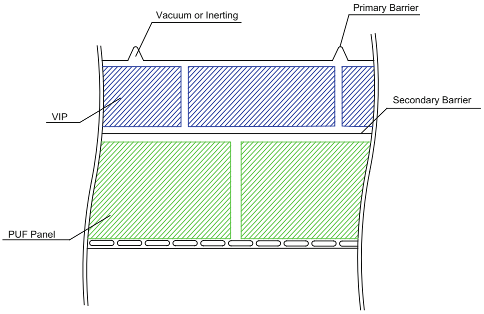

Fig. 5. Example of an insulation system for LH_2 membrane tank in the future.

4 BOG Handling System and Propulsion System

The development of BOG handling and propulsion systems needs to be considered together with the tank insulation system. In other words, it is anticipated that it will be necessary to develop an insulation system that will control BOG generation to levels required for propulsion/power generation. Furthermore, re-liquefaction of BOG and combustion via Gas Combustion Unit (GCU) is also possible. However, in the case of re-liquefaction, a large amount of energy is consumed compared with the re-liquefaction of LNG, so its application is limited and it is unrealistic to apply it in the short to medium term [28]. Example of BOG handling system and propulsion system in illustrated in Fig. 6.

LNG-based rules prohibit the release of BOG into the atmosphere except in emergency situations because air pollution is closely related to the greenhouse effect and methane emissions [29]. In the case of hydrogen, there is no risk of air pollution, so it is considered necessary to establish a certain area around the exhaust point as a safety

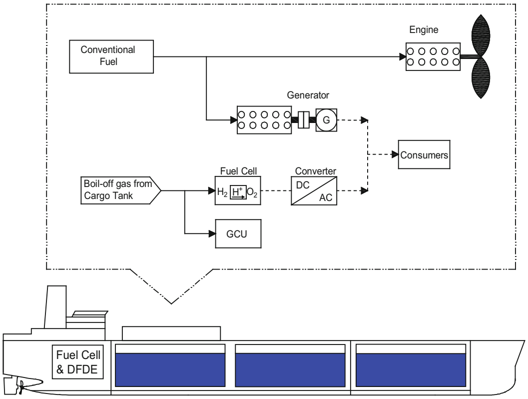

Fig. 6. Example of BOG handling system and propulsion system.

zone to eliminate the risk of fire/explosion, and to examine whether natural discharge after dilution can be performed without a separate BOG treatment facility.

Hydrogen can be usually used in the field of fuel cells (see Table 2). Due to the absence of moving parts, fuel cells have some advantages like very low noise, low vibration and low pollutant emissions. However, the tolerance to impurities are their challenges, especially the proton exchange membrane fuel cells (PEMFCs) and shock resistance. PEMFCs and solid oxide fuel cells (SOFCs) are both effective at about 60%. Besides, SOFCs has an additional advantage [30]. They can operate under high temperature (700–1000 °C), which implies that they can be used to generate steam and power needed in steam turbine. The overall system efficiency can be increased to about 80% [31]. As for the both technologies, the additional ancillary components (e.g. plant balancing) reduce the overall efficiency of a few percent. These losses become higher as long as the fuel cells become larger. The specific power (kg/kW) of SOFCs is lower than that of PEMFCs. They have high operating temperatures, long start-up times and poor tolerance to load variations. Internal combustion engines (ICEs) become more efficient owning to the larger sizes, which have a higher average power density, lower costs as well as more tolerance to load changes, and lasts longer. Some disadvantages of them can be noticed such as noise, vibration and low efficiency.

5 Conclusions

Trade of hydrogen between countries will be carried out in the form of ammonia, liquid hydrogen, LOHC, etc., taking into account the renewable energy resources of the exporting country and the hydrogen usage type and technological maturity of the importing country, and will not be traded in only one form. Therefore, in this article, the key technology or potential demand technology are listed.

(1) Development of efficient insulation system.

- For type C tank, vacuum and MLI/glass bubble insulation technology need to develop application technology.

Table. 2. Comparison between direct hydrogen use in fuel cells and ICEs

Performance	ICE	PEMFC	SOFC	GAS TURBINE
Conversion efficiency (%)	50	52	60	35
System efficiency (%)	50	56	80	58
Cost (USD/kW)	< 500	> 1500	> 4500	–
Specific power(kg/kW)	2–11	4	50	1.25–2
Partial load efficiency	High	High	High	Low
Tolerance to load variations	High	Medium	Low	High
Maturity	High	Medium	Low	High
Lifetime	High	Low	Low	–
Noise/vibration	High	Low	Low	High
NO_X and hydrocarbon emissions	Medium	Low	Low	Medium

- For Membrane tank, IBS vacuum maintenance and VIP aging technology research are required.

(2) Development of an efficient hydrogen BOG treatment system and propulsion system are needed.

  - Large-capacity fuel cell (PEM or SOFC) needs to be developed.
  - Hydrogen BOG dilution emission method needs further verification and inspection.

(3) Considering the low density of liquefied hydrogen, stability, propeller immersion, draft changes (cargo loading/unloading), etc., it is necessary to optimize the linear design of the hull.

At the beginning of the introduction of liquid hydrogen carriers, about 20,000–40,000 m^3 capacity liquid hydrogen carriers equipped with type C tanks with relatively low technical hurdles are suitable, and in the long term it is considered necessary to develop membrane-type liquid hydrogen carriers. In order to succeed in the commercialization of liquefied hydrogen carriers, the reliability and safety of the technology have to be ensured, and there are still technical challenges to be overcome. It is expected that domestic shipyards will successfully enter into the technology of liquefied hydrogen carriers by utilizing their know-how in developing LNG carriers, which is their strength, and that liquefied hydrogen will play an important role in the hydrogen supply chain.

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