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SPERA Hydrogen System^{TM} by LOHC-MCH Method for Massive Hydrogen Storage and Transportation

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Abstract: Expanding the hydrogen energy utilization is essential for decarbonization, and the commercialization of hydrogen energy carrier systems that can “store” and “transport” hydrogen in a large scale is necessary. The organic chemical hydride method incorporates hydrogen atoms into the molecular structure of a LOHC (Liquid Organic Hydrogen Carrier) to store and transport hydrogen in a liquid state under normal temperature and pressure, and is a highly safe method with low business risk. Chiyoda has been developing the technology since 2002, completed a pilot demonstration in 2014 and named it the SPERA Hydrogen^{TM} System, and successfully completed an international supply chain demonstration that transports hydrogen from Brunei to Japan in a large scale in 2020, moving to the commercialization stage. Currently, Chiyoda is conducting feasibility studies with a number of domestic and foreign companies with the aim of commercializing the system as soon as possible. In this paper, outline, features, development status and our efforts in commercialization of SPERA Hydrogen^{TM} System are introduced.

Key words: Hydrogen, carrier, LOHC, Methylcyclohexane, MCH, toluene.

1. Introduction

Since 2002, Chiyoda focused on the features of the organic chemical hydride method using a LOHC (Liquid Organic Hydrogen Carrier) as a technology for large-scale storage and transport of hydrogen. We started the development of novel dehydrogenation catalysts for MCH (Methylcyclohexane), which is the key to practical use of MCH as LOHC (LOHC-MCH). After successfully developing of a new dehydrogenation catalyst in 2011, we completed a 10,000-hour demonstration in a pilot plant to establish the process technology in 2014, and in 2015, it began participating in an international hydrogen supply chain demonstration project founded by NEDO (New Energy and Industrial Technology Development Organization, Japan), with the goal of transporting massive hydrogen from Brunei Darussalam in Southeast Asia to the Kawasaki waterfront area in Japan in 2020. The demonstration of the entire process to transport more than 100 tons of hydrogen from Brunei Darussalam to the Kawasaki waterfront was successfully completed. SPERA Hydrogen System moved to the commercialization stage. Currently, Chiyoda is conducting feasibility studies with many domestic and foreign companies with the aim of commercializing the system as soon as possible.

In this paper, outline, features, development status and our efforts in commercialization of SPERA Hydrogen System are introduced.

2. Outline of SPERA Hydrogen System

2.1 Scheme of SPERA Hydrogen System [1-5]

Fig. 1 shows the overall process of the LOHC-MCH

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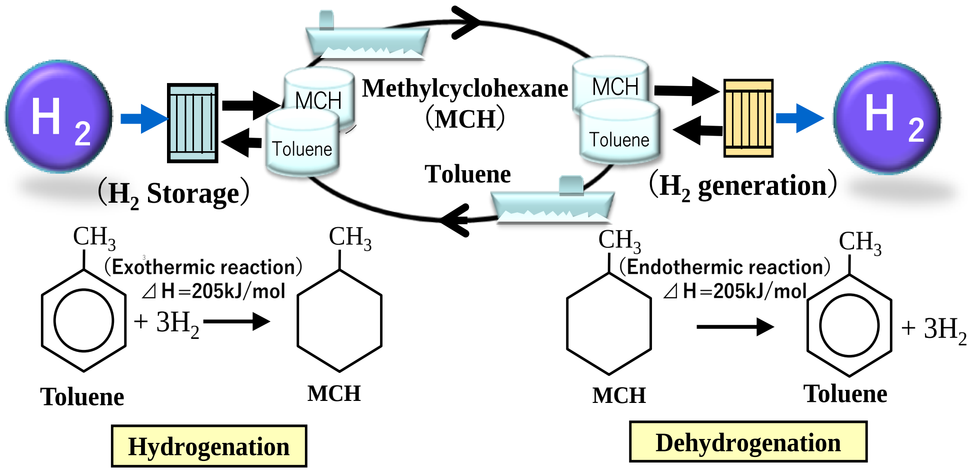

Fig. 1 Scheme of SPERA hydrogen system.

method. In this method, hydrogen atoms are stored in the molecules of MCH produced through the hydrogenation reaction of hydrogen produced from fossil resources or renewable energy with aromatics such as toluene. Hydrogen is in a gas state at ambient temperature and pressure, and its small molecular weight makes it difficult to store and transport in a large volume per weight, but by storing hydrogen atoms in the molecules of MCH through the hydrogenation process, it is possible to "store" and "transport" hydrogen as a liquid state at 1/530 of volume compared with gas volume at room temperature and pressure. In other words, it is possible to store and transport hydrogen gas as a liquid with the same volume as that of hydrogen gas compressed to 530 atm.

After transporting MCH by sea or land, the necessary amount of hydrogen is generated from MCH through dehydrogenation reaction. The toluene produced in the dehydrogenation process is returned to the hydrogen shipping place and used as a feed for the hydrogenation process repeatedly. As described above, this system consists of hydrogenation, transportation, and dehydrogenation process, and the storage and transportation can be performed in a liquid state under ambient temperature and pressure conditions.

2.2 Features [6-9]

Since hydrogen can be stored and transported in a liquid state at ambient temperature and pressure, this method has very low energy loss in long-term storage. In addition, since toluene and MCH do not chemically change under ambient conditions, this method is suitable for long-term and large-scale storage, and it is also possible to convert tanks existing at refinery etc. Furthermore, national stockpiling of hydrogen is possible by converting existing national stockpiling of oil.

Toluene and MCH are solvents used in paints and white correction pens etc., which have low toxicity and are easy to handle as clear liquids with low viscosity like water. Toluene and MCH are widely distributed as chemicals, and can be transported by sea in chemical tankers, ISO (International Organization for Standardization) tank containers (ISO standard tanks for containers that can be transshipped directly to cargo ships/trucks/railways), chemical trucks, and land transportation by rail freight. In addition to the use of the distribution infrastructure for gasoline, large tanks at refineries can also be converted for storage, thus maximizing the use of existing infrastructure and reducing capital investment costs.

Since hydrogen is an explosive gas, it is a supply chain with high potential risks in the case of large-scale storage of hydrogen under special conditions, such as liquid hydrogen, or in the case of converting hydrogen into highly toxic substances, such as ammonia. The large storage volume for power generation makes it a

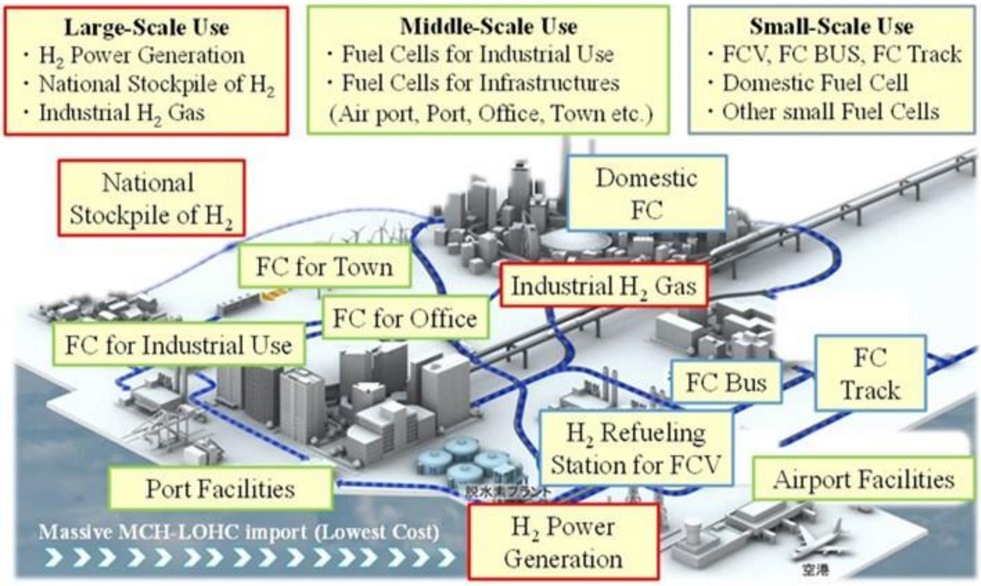

Fig. 2 Concept of integrated hydrogen supply chain.

business with high business risks such as post-accident compensation in case of accidents such as explosions or leaks caused by unexpected natural disasters such as an earthquake etc. Since energy infrastructures are lifelines, the risk assessment for the case of terrorist or missile attacks is also significant from the viewpoint of national security.

Toluene and MCH have the same hazardous material as gasoline, so they can be handled in the same way as gasoline, and they are not corrosive and have low toxicity. SPERA hydrogen system is a method that can reduce supply chain business risks and national security risks to the same level as existing gasoline. It is a highly safe system that allows hydrogen supply business with the same risk in principle as the existing petroleum business.

Since the demonstration of entire processes necessary for the commercialization was completed, the system is ready for commercialization.

2.3 Integrated Hydrogen Supply Chain [10].

Fig. 2 shows the concept of the integrated hydrogen supply chain for which we would like to realize in the future This system can be applied not only for thermal power generation, but also for national stockpiling and supply to remote islands in the same way as gasoline. MCH supplied for thermal power generation in a large scale is the lowest cost MCH due to scale merit. We consider that it will be possible to establish an integrated hydrogen supply chain by sharing low cost MCH for large-scale use as hydrogen supply to small- and medium-scale use.

3. Development

3.1 Dehydrogenation Catalyst [11-16]

Hydrogenation process was developed more than 40 years ago and many large hydrogenation plants are in operation as cyclohexane production plant through hydrogenation of benzene. MCH is produced as solvent through same hydrogenation process by using toluene as feed. Nickel catalyst is mainly used in the hydrogenation process. Since its yield is more than 99%, we had no necessity to develop a novel hydrogenation process.

On the other hand, although research of dehydrogenation catalysts for MCH was reported since the Euro-Quebec project in the 1980s, no catalyst with an industrial lifetime was developed due to significant carbon deposition by coking reaction.

We spent about 10 years since 2002 developing a novel dehydrogenation catalyst that can be used industrially.

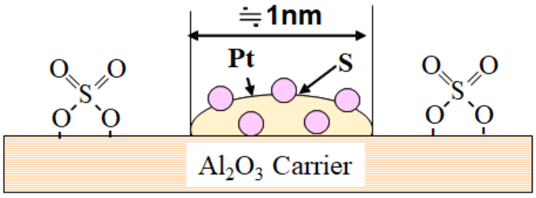

Fig. 3 Estimated catalyst surface model of developed catalyst.

Fig. 3 shows the estimated surface model of the dehydrogenation catalyst we developed. It was known that catalysts with platinum as the active metal supported on γ-alumina support.

It shows high performance in the MCH dehydrogenation reaction. We were able to develop a dehydrogenation catalyst that can be used as an industrial catalyst with a long catalyst life more than one year by reducing the size of platinum particles to about 1 nm and by partially modifying the surface of the platinum particles with sulfur.

After development in the laboratory, we established a system to produce the dehydrogenation catalyst industrially in 2011. In addition, we are currently demonstrating an improved catalyst aimed 3-4 years catalyst life.

3.2 Pilot Plant Demonstration [17-21]

In early 2012, Chiyoda decided on a pilot plant demonstration project to establish hydrogenation process and dehydrogenation process. The pilot plant was built at R&D Center of Chiyoda Corporation which is in Yokohama city in Japan in 2013. The capacity of the pilot plant is 50 Nm^3/h.

The demonstration operation was carried out for 18 months from April 2013 to October 2014, stable performance was confirmed for 18 months totally around 10,000 h. The hydrogen generated in the dehydrogenation reactor was sent to the hydrogenation reactor to react with toluene again to produce MCH. Fig. 4 shows a photograph of the pilot plant and its performance. Toluene conversion: > 98%, MCH selectivity: > 99%, and MCH yield: > 98% were confirmed as the performance after about 10,000 h operation of the hydrogenation plant. Similarly, MCH conversion: > 98%, toluene selectivity: > 99%, and toluene yield: > 98% were confirmed as results after 10,000 h operation of the dehydrogenation plant. The results of the demonstration operation showed high performance and stability as originally designed, and we were able to establish the technology for the hydrogenation and dehydrogenation processes. We named the developed system "SPERA Hydrogen" system.

Reaction Section

Storage Section

Fig. 4 Pilot plant of SPERA hydrogen system.

Location: Chiyoda Corporation R&D Center, Yokoharna, Japan; Capacity: 50 Nm^3/h; Demonstration period: 2013.4-2014.11 (≈ 10,000 h) Performance.

Hydrogenation Toluene conversion: > 99%; MCH Selectivity: 99%; MCH yield: 99%.

Dehydrogenation MCH conversion: > 98%; Toluene selectivity: 99%; Toluene yield: > 98%.

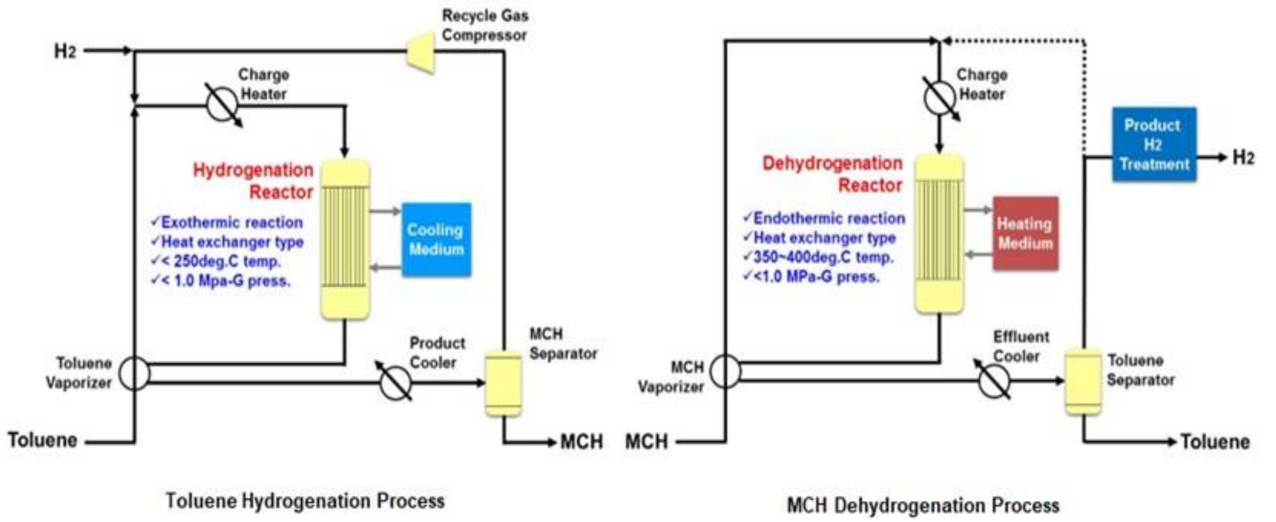

Fig. 5 Hydrogenation process and dehydrogenation process.

Fig. 5 shows an overview of the hydrogenation and dehydrogenation processes. Both reactors are heat exchanger type of tubular reactors. The reaction conditions for the hydrogenation process are temperature: < 250 ℃, pressure: < 1.0 MPaG. The outside of reaction tube charged with hydrogenation catalyst is cooled by water since hydrogenation is exothermic reaction. Hydrogenation catalyst is conventional nickel catalyst. On the other hand, the reaction conditions for the dehydrogenation process are temperature: 350-400 ℃, pressure: < 1.0 MPaG. The outside of reaction tube charged with dehydrogenation catalyst is heated by hot oil since dehydrogenation is endothermic reaction. Hot oil was heated by electrical heater. Dehydrogenation catalyst is SPERA dehydrogenation catalyst developed as mentioned above. Since both reactors are heat exchanger type, scale-up is easy by increasing the number of catalytic tubes. In addition, both toluene and MCH are not corrosive, so they can be constructed with inexpensive steel materials.

4. International Demonstration in a Large Scale [22-25]

Chiyoda participated in a national project of the international hydrogen supply chain demonstration funded by NEDO (New Energy and industrial Technology Development Organization) from FY2015. Fig. 5 shows the outline of the project. In this demonstration, hydrogen is produced in Brunei Darussalam and transported to the Kawasaki waterfront in Japan by the SPERA hydrogen system. Brunei Darussalam is in the northwestern part of Borneo Island in Southeast Asia, 5,000 km away from Japan, and is known as a natural gas exporting country to Japan.

The project was executed by AHEAD (Advanced Hydrogen Energy Chain Association for Technology Development) which is a technology research association established by Mitsubishi Corporation, and Mitsui & Co., NYK LINE and Chiyoda Corporation.

In addition to the above four companies, AHEAD has received technical cooperation from Mitsubishi Power Corporation (present Mitsubishi Heavy Industries, Ltd.) and the Development Bank of Japan, as well as from the Bruneian government and Kawasaki City.

Fig. 7 shows the photograph of hydrogenation plant in Brunei Darussalam and dehydrogenation plant in Japan. The capacity of the hydrogenation and dehydrogenation plants constructed in Brunei and Kawasaki is 300 Nm^3/h and can produce and transport up to 210 tons of hydrogen per year maximally, which is equivalent to the amount of hydrogen that can refuel 40,000 FCVs (Fuel Cell Vehicles).

Description	
Scale	210 tons/year (maximum)
Duration	In 2020
Hydrogen Supply	Brunei Darussalam (Hydrogen Production)
Hydrogen Demand	Kawasaki City, Japan (fuel for gas turbine power plant)
Transportation	ISO tank container (container ship/truck)
Business Scheme	Establishment of the Association for Technology Development. NEDO Funded Project

Fig. 6 Outline of international hydrogen supply chain demonstration project.

Hydrogenation plant in Brunei Darussalam　　Dehydrogenation plant in Japan

Fig. 7 Photograph of Hydrogenation plant in Brunei Darussalam and dehydrogenation plant in Japan.

Fig. 8 shows the scheme of the international hydrogen supply chain project. In Brunei, process gas from an adjacent LNG plant was sent to a hydrogenation plant, where hydrogen produced in a steam reforming hydrogen production unit was reacted with toluene to produce MCH. Produced MCH was loaded from tanks to ISO tank containers and transported to the port. ISO container was shipped from Brunei Darussalam by large cargo ship to the Port of Kawasaki, and then transported to the dehydrogenation plant constructed in the refinery of Toa Oil Co. Ltd. The hydrogen generated by the dehydrogenation reaction was mixed with by-product gas of the refinery supplied to the Mizue Power Plant adjacent to the refinery and used for gas turbine power generation. The toluene produced after hydrogen generation was returned to Brunei by land and sea transportation in ISO tank containers. The toluene was converted to MCH through the hydrogenation process again for reuse.

We utilized the ISO container and cargo ship for this demonstration project. Fig. 9 shows the ISO container and truck and large cargo ship. Since chemical tanker by which hydrogen can be transported in several thousand tons of hydrogen per ship project is too big for this demonstration. Thus, we were also able to demonstrate that this system can use ISO

(International organization for standardization) containers for cargo transport. Cargo transport will be convenient for transportation to small remote islands.

The above demonstration project was successfully completed in 2020 with over 100 tons of hydrogen transported from Brunei Darussalam to Japan. Chiyoda made a short movie for the introduction of the demonstration project and upload to our website.

Since the entire supply chain process including the transportation process other than the process technology of the hydrogenation and dehydrogenation processes was demonstrated in a large scale as an international supply chain, SPERA hydrogen system was moved into the commercialization phase. Currently, Chiyoda is working with a number of domestic and foreign companies to study the feasibility of commercialization of the system.

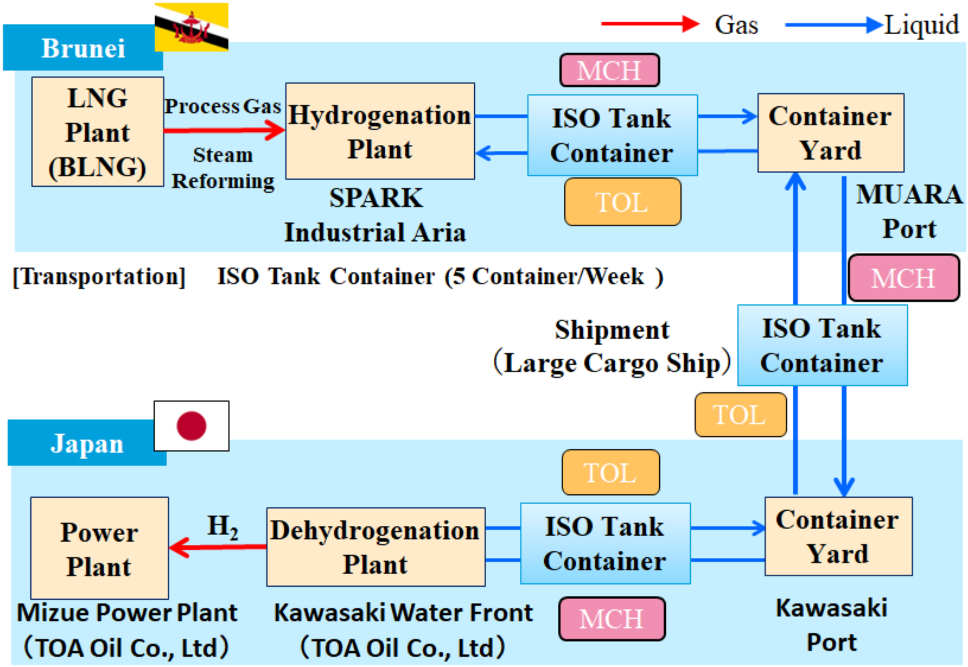

Fig. 8 Scheme of the international hydrogen supply chain project.

MCH: Methylcyclohexane, TOL: Toluene.

ISO container and track

Large cargo ship

Fig. 9 Photograph of ISO container and track and container vessel.

5. Hydrogen Supply Cost

Fig. 10 shows an example of a comparison for hydrogen supply cost estimation by European Hydrogen Backbone as an independent third party [25]. It shows a relative comparison of the overall costs of hydrogen transportation with different forms: methanol, liquefied hydrogen, liquefied ammonia and LOHC method. The liquid hydrogen method is highly expensive. The estimated cost of liquid ammonia decomposed after transportation to provide hydrogen and the LOHC method is roughly equivalent.

Chiyoda is aiming for early commercialization within several years by the SPERA hydrogen system established in the international demonstration and is conducting trial calculations of hydrogen supply costs in various cases. The hydrogen supply cost varies greatly depending on the transportation distance, availability of utilities at the hydrogenation and dehydrogenation site, and convertible existing infrastructure such as tanks and tankers.

6. Efforts for Commercialization

Toward the commercialization SPERA hydrogen system for Global Supply Chain, there are several on-going projects which are executing study or further engineering activities in Asia and Europe. In Asia, we have 2 focused H_2 demand center, one is Japan and the other is Singapore. In Singapore, 5 Singaporean companies and 2 Japanese companies have agreed in March 2020 to conduct feasibility studies for a hydrogen import value chain using SPERA hydrogen system with strong Singaporean government's support, toward the net zero in Singapore by 2050.

Chiyoda, Sembcorp Industries and Mitsubishi Corporation have signed a MOU (Memorandum of Understanding) in October 2022 [27], to start Pre-FEED (Preliminary-Front End Engineering and Design) study for low carbon hydrogen import chain from prospective hydrogen exporting countries to Singapore that aims to start operation in 2026.

Also, in Japan, Chiyoda participates in several projects and tries to establish the global supply chain to import hydrogen from overseas to Japan and is working with utilities, energy and chemical companies, equipment suppliers, industrial gas companies and government agencies [28].

In Europe, Chiyoda has concluded MOU with Port of Rotterdam and Koole Terminals, Mitsubishi Corporation in 2021 [29, 30], to do the joint study for importing hydrogen to European market, and now is under shaping the project to identify specific hydrogen off-takers in Europe and suppliers in global.

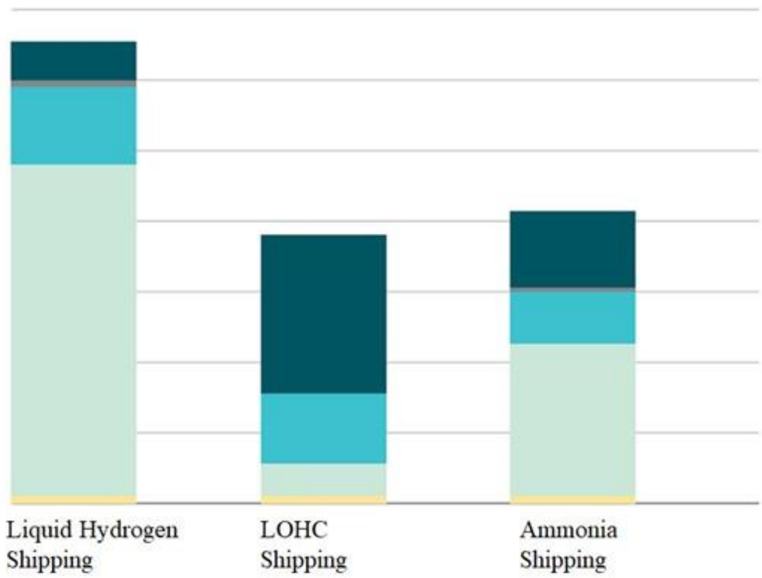

Fig. 10 Example of a relative cost comparison for hydrogen supply chain.

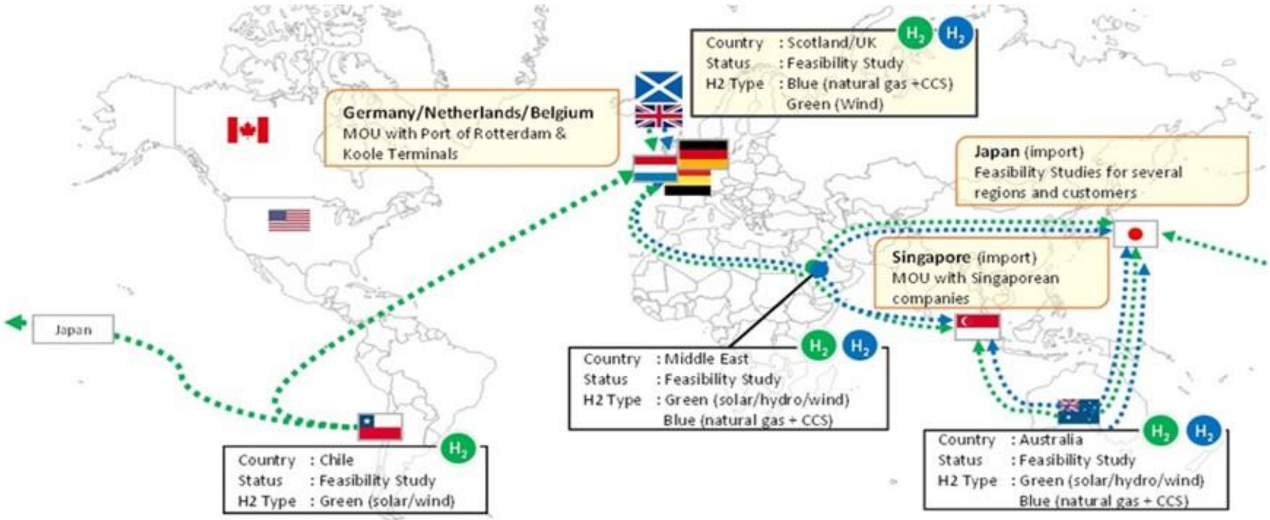

Fig. 11 Global investigation for the commercialization of SPERA hydrogen system.

Chiyoda announced to join the international consortium along with other 9 parties (government organizations, private companies in Europe) [31]. The project is called LHyTS (LOHC for Hydrogen Transport from Scotland) project, developing a pilot project as a precursor to large scale export by using MCH as a hydrogen carrier [32].

Our target schedule for commercialization of global hydrogen supply chain is to start operation by mid-2020's for pilot/semi-commercial projects and by 2030 for full-scale commercial projects, in Europe and the Asian markets.

7. Conclusion

SPERA hydrogen system is an inexpensive method because it requires no special conditions or materials at ambient temperature and pressure, and existing infrastructure such as tanks and tankers can be diverted to the maximum extent possible. The ability to provide long-term storage on a large scale is a feasible way to build a comprehensive hydrogen supply chain, from national stockpiles to small- and medium-scale operations. Since a large-scale demonstration of the entire process has been completed in the international hydrogen supply chain demonstration and moved to the commercialization stage. Chiyoda hopes to contribute to the realization of a decarbonized society through the early commercialization of SPERA hydrogen system.

Acknowledgement

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We would also like to express our gratitude to the companies that participated in AHEAD, to the companies that participated as joint research partners, and to those who cooperated with the companies.

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