

Design and analysis of a novel liquid hydrogen production system using dual mixed refrigerant-based cryogenic precooling and liquefaction processes

Ahmad K. Sleiti^{1*}, Wahib A. Al-Ammari¹

¹ Department of Mechanical & Industrial Engineering, College of Engineering, Qatar University, Doha, Qatar

* Corresponding Author: asleiti@qu.edu.qa

ABSTRACT

This study presents an innovative LH₂ production system using dual mixed refrigerant (DMR)-based cryogenic processes. New dual-refrigeration loops are developed for the precooling and liquefaction of the gaseous hydrogen with two distinguished mixed refrigerants for each process. Thorough energy, exergy, economic, and environmental (4E) analyses are conducted for the proposed LH₂ system in this study. The proposed system produces LH₂ with an energy consumption of 3.732 kWh/kg_{LH₂}, 48% lower than the single mixed refrigerant SMR-based systems and 70% lower than the current commercial plants. Furthermore, the exergy efficiency (59.65%) of the present system is enhanced by 33% compared to the SMR-based systems (44.89). In addition, the present system produces LH₂ with a levelized cost of 1.89 \$/kg_{LH₂}, which is 21% lower than that for SMR-based large-scale systems and 70% lower than that of small-scale systems. Environmentally, the proposed DMR-LH₂ system reduced CO₂ emissions by 29-69% compared to large-scale SMR-based systems. Moreover, the DMR-LH₂ system is characterized by more flexibility in the design of the process equipment and eliminates the potential for freezing problems. This study's dual mixed cryogenic refrigeration approach provides guidelines for future research to improve the technical and economic feasibility of LH₂ production, making it competitive with other energy storage and transportation options.

Keywords: liquid hydrogen, cryogenic refrigeration, mixed refrigerants, SEC, LCOH, 4E analyses.

NONMENCLATURE

Abbreviations

ACE	Annual CO ₂ emissions
COP	Coefficient of performance
CGH ₂	Compressed gaseous hydrogen

DMR	Dual mixed refrigerants
HPP	Hydrogen precooling process
HLP	Hydrogen liquefaction process
LCOH	Levelized cost of hydrogen
SEC	Specific energy consumption
SMR	Single mixed refrigerant
TAC	Total annualized cost
4E	Energy, exergy, economic, and environmental analyses

1. INTRODUCTION

Storing hydrogen in liquid form is considered a superior alternative to the current hydrogen storage and transportation methods. This is mainly due to the high energy density of liquid hydrogen (LH₂), ~120 MJ/kg, and its near-zero CO₂ emissions to the ambient (for green and blue H₂) [1]. The shift towards H₂, as future's fuel, is driven by the urgent issue of global warming, which poses a significant threat to the survival and progress of humanity [2], [3]. It is estimated that between 2021 to 2050, the utilization of H₂ could prevent the release of 80 gigatons of cumulative CO₂ emissions. Furthermore, H₂ is projected to play a key role in achieving 20% of the total emissions reduction target for 2050 [4].

From an economic perspective, LH₂ offers a more efficient and viable solution for overseas/long-distance energy transportation compared to its compressed gaseous form (CGH₂) [5], [6]. For instance, at a cryogenic temperature of 20 K (-253°C) and pressure of 1 bar, the LH₂ density is 70 kg/m³, which is 3 times higher than of CGH₂ (21 kg/m³ at 25°C and 700 bar). In addition, the average cost of CGH₂ storage is 14 \$/kWh (at 300-700 bar), which is 2.3 times higher than that of storing H₂ in liquid form (6 \$/kWh) [7]. Therefore, LH₂ is proposed to be used as an energy storage method for the renewable energy sources (wind, solar) to address their intermittency issue. This will improve their capacity by 20% to 40% [8], [9]. Furthermore, CGH₂ has a high

potential for explosion in case of storage penetration or material failure as it is stored at extremely high pressures (300-700 bar). In terms of technology readiness level (TRL), in 2019, HySTRA partners launched the first LH₂ carrier ship in the world in Japan. The prototype ship, which can carry 75 bar, was used to show off trips between Australia and Japan to launch the LH₂ economy.

Despite the advantages of LH₂ over CGH₂, it's important to note that the liquefaction process remains considerably costly (\$9,000/kgLH₂ [11]) and consumes a substantial amount of energy, ranging from 10 to 13 kWh/kg_{LH₂}[10], which is 3.5 times higher than the minimum theoretical energy needed for liquefaction (3.30 kWh/kg_{LH₂}) [12]. Therefore, several efforts were made to develop H₂ liquefaction systems (HLS) with minimal energy consumption and feasible cost.

A typical HLS consists of two processes, namely called H₂ precooling process (HPP), and H₂ liquefaction process (HLP). The HPP reduces the H₂ temperature from 25°C to less than of -190°C and the HLP reduces it more to -253°C at a nominal pressure of 21 bar. Three approaches were proposed to minimize the energy consumption of the HLS, which are (i) integrating the HPP with absorption [11], CO₂ liquefaction [12], and water splitting systems[13], (ii) Recovering the cold energy of the liquefied natural gas (LNG) (during its regasification process) in cool H₂ in the first stage of the HLP, and (iii) improving the performance of the HLS by the use of mixed refrigerants instead of pure refrigerants to accomplish the liquefaction process. The first approach increases the complexity and capital cost of the system without substantial reduction in energy consumption. The second approach is limited by the location of the LNG regasification and the available space to build LHS without safety concerns. Thus, the third approach is the most feasible way to reduce the energy consumption of LHS or other cryogenic processes [14].

The use of MR in the HLS reduces the energy consumption from 12 kWh/kg_{LH₂} (for pure refrigerants-based systems) to about 6.06 kWh/kg_{LH₂} [15] without complicating the process configuration [16], [17]. However, this energy level is still about two times higher than the minimum theoretical energy consumption for H₂ liquefaction (3.30 kWh/kg_{LH₂}). Therefore, the MR-based systems still need further improvements and investigations to reduce the specific energy consumption (SEC) to at least less than 4.00 kWh/kgLH₂, which is the main objective of this study.

To achieve this target (SEC < 4.00 kWh/kg_{LH₂}), new approach with innovative HLS is introduced in this study. First, the flowsheet of the HPP and HLP are modified to

use two refrigeration loops in each of them instead of single refrigeration loop as in conventional single mixed refrigerant (SMR)-based systems. Then, efficient, and reliable mixed refrigerants are developed for each loop. Thus, two distinguished mixed refrigerants will be used in the HPP and another two mixed refrigerants in the HLP. Therefore, the proposed system in this study is working based on dual mixed refrigerants in each process. Thus, we refer to it as a dual mixed refrigerant-hydrogen liquefaction system (DMR-HLS). With the use of DMR in the HPP, a cryogenic temperature of -195°C can be achieved without the use of very lightweight refrigerants (neon (Ne), helium (He), and hydrogen (H₂)) in the developed mixtures. This will maintain efficient compression, reduce the mass flow of the MRs, and minimize the SEC of the HPP. In addition, the use of DMR in the HLP reduces its SEC and prevents the potential of refrigerant freezing issue. To elaborate this advantage, it is noted the available studies in the literature proposed the use of a MR in a single refrigeration loop and consists of H₂, He, and Ne [18], [19]. However, at the final stage of the HLP, this MR has cryogenic temperatures less than of -253°C, which is lower than its freezing temperature (-248.6°C). Thus, using this mixture may block and freeze out the flow in the cryogenic heat exchangers of the process. This also may cause severe damage for the heat exchangers and negatively affect the efficiency of the process. Therefore, as proposed in this study, using a dual MR-based configuration will address this problem without losing the advantage of using Ne in the MR (which is the achievement of low compression power). This is done by using Ne in a MR loop with temperatures higher than -248°C and only using H₂ and He in another MR loop with temperatures less than -248°C.

To summarize, this study presents reliable, efficient, and cost-effective HLS with minimal energy consumption using dual mixed refrigerants in the HPP and HLP. The main objectives of this study are:

- Introducing an innovative, efficient, and large-scale LH₂ production system using dual mixed refrigerants-based precooling and liquefaction processes to achieve a SEC < 4.00 kWh/kg_{LH₂}.
- Developing efficient refrigerant mixtures for the new DMR-HLS that achieve superior performance without any potential for freezing problems in the process.
- Conducting energy, exergy, economic, and environmental (4E) analyses to performance of the proposed HLS.

- Evaluating the performance of the present DMR-HLS in comparison to existing conventional and SMR-based H₂ liquefaction systems.

A detailed description of the proposed system is provided in Section 2. The performance indicators are

stage intercooled compression process after leaving the cryogenic heat exchangers. In the HPP, throttling valves (TVs) are used to expand the flow from the high pressure to low pressure side of the refrigeration process. In the HLP, expanders are used for the expansion process

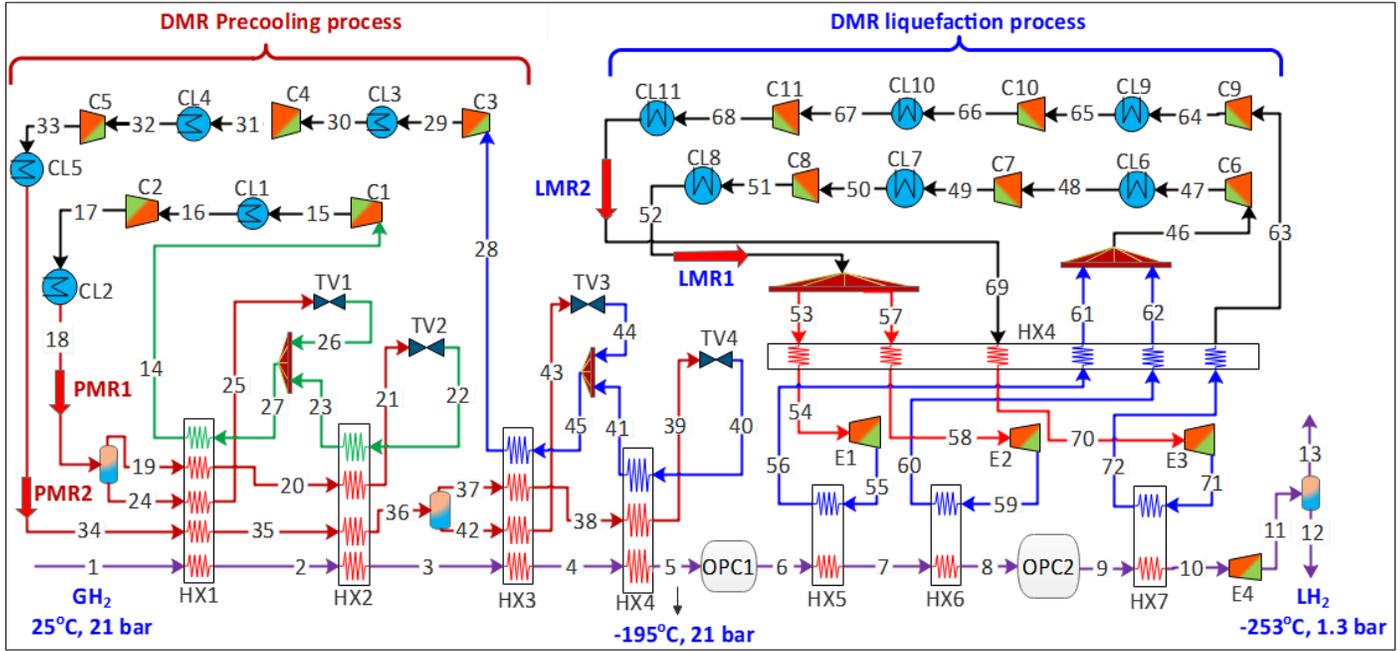


Fig. 1. Flowsheet of the new dual mixed refrigerant-based liquid hydrogen production system.

explained in Section 3. economic, the results of the energy, exergy, economic, and environmental analyses of the present system are discussed in Section 4. Finally, the main findings and conclusions of this study are summarized in the Conclusion section.

2. FLOWSHEET OF THE PROPOSED LH₂ PRODUCTION SYSTEM

Fig. 1 shows the flowsheet of the proposed LH₂ production system. It consists of two main processes, which are (i) H₂ precooling process (HPP), and (ii) H₂ liquefaction process. The HPP precools the GH₂ from 25°C (state 1) to -195°C (state 5) at 21 bar using two refrigeration loops. And the HLP reduces the temperature to the hydrogen liquefaction point (-253°C at 1.3 bar, state 12). The HPP uses two distinguished mixed refrigerants: PMR1 and PMR2. PMR1 is circulated in the first HPP refrigeration loop and responsible for the cooling duty of heat exchanger HX1 and HX2. The PMR2 is circulated in the second HPP refrigeration loop and responsible for the cooling duty of HX3 and HX4. Similarly, the HLP is driven by two refrigeration loops with the use of two distinguished refrigerants: LMR1 and LMR2. LMR1 is responsible for the cooling duty of HX5 and HX6, while LMR2 is responsible for the cooling duty of HX7. Each MR is compressed and cooled in a multi-

instead of throttling valves. Internal calculations show that the use of TVs in the HPP is more cost effective than using expanders for the throttling process. For efficient LH₂ storage after the liquefaction process, two ortho-para H₂ converters (OPC1, and OPC2) are used. These converters accelerate the conversion of ortho isomers of H₂ into ortho-isomers, which prevents the evaporation of LH₂ in the storage tanks. Further details about the OPC and their working principles are provided in [20], and [12].

3. METHODOLOGY

In this section, the definitions of the performance indicators of the HLS are introduced with further details in the economic model as the literature lacks detailed models for the MR-based HLS.

3.1 Thermodynamic performance indicators

To evaluate the thermodynamic performance of the proposed DMR-LHS, it is first simulated in the Aspen HYSYS software. Then, the material stream properties were obtained and used to analyze the performance of the system. For the simulation process, the Peng-Robinson equation of state (EOS) is used for the mixed refrigerant properties and the MBWR EOS is used for the properties of hydrogen flow streams. In addition, the

simulation process is performed under steady-state conditions and the pressure drop through the HXs and coolers is neglected.

Three thermodynamic-based indicators are used to assess the performance of the HLS. The first indicator is the specific energy consumption (SEC), which evaluates how much energy is consumed per kg of liquefied hydrogen as shown in Eq. 1.

$$SEC = \frac{\sum \dot{W}_{C,i} - \sum \dot{W}_{E,i}}{\dot{m}_{HF}} \quad (1)$$

Where $\sum \dot{W}_{C,i}$, $\sum \dot{W}_{E,i}$, and \dot{m}_{HF} are the total work rate of the compressors, total work rate of expanders, and the mass flow rate of the fed gaseous hydrogen. The second indicator is the coefficient of performance, which evaluates the effectiveness of the cryogenic heat exchangers by comparing the cooling duty $\sum Q_{HX,CD}$ to the consumed power as given in Eq. 2.

$$COP = \frac{\sum Q_{HX,CD}}{\sum \dot{W}_{C,i} - \sum \dot{W}_{E,i}} \quad (2)$$

To evaluate the overall efficiency of the energy utilization through the overall process and to assess the amount of the energy losses (destruction), the overall exergy efficiency is defined as in Eq. 3.

$$\varepsilon_{overall} = \{\dot{E}_{Product} - \dot{E}_{Feed}\} / \dot{W}_{total} \quad (3)$$

The detailed exergy models of the HLS components can be obtained from [21].

3.2 Economic assessment method

Guthrie's module costing method is adapted in this study to evaluate the economic performance of the proposed HLS. This method entails dissecting the process into smaller modules or constituents to evaluate their costs. Then, the cumulative expenses of each module are subsequently used to establish the overall cost of the HLS. Using this method, calculating CAPEX is started by the calculation of the equipment purchase cost (E_p), which is obtained from Eq. 4 [22].

$$\log_{10}(E_p) = K_1 + K_2 \log_{10}(A) + K_3 (\log_{10}(A))^2 \quad (4)$$

The component's capacity (A) and the values of the cost constants (K_1 , K_2 , and K_3) can be obtained from [21]. After calculating E_p , the bare module cost is obtained for each component using Eq. 5:

$$C_{BM} = E_p \times F_{BM} \quad (5)$$

where F_{BM} is the cost factor of the bare module, and its values for the process equipment. The constants of the above calculations are calculated using a survey of

equipment manufacturers from May to September 2001. Thus, the actual cost of equipment purchase must be updated considering the inflation rate. This is done by using the average chemical engineering plant cost index (CEPCI) of the target year (e.g. $CEPCI_{2022} = 816$) and of the reference year ($CEPCI_{2001} = 394$) as shown in Eq. 6.

$$C_{BM,real} = C_{BM} \times \frac{CEPCI_{real}}{CEPCI_{2001}} \quad (6)$$

The CAPEX also includes the cost of other equipment (e.g. mixers, separators, pipes, and valves) and installation cost. These costs are estimated as 18% of the CAPEX of the major components. Thus, the CAPEX of the plant is calculated as in Eq. 7.

$$CAPEX = 1.18 \times \sum_k^m C_{BM,real} \quad (7)$$

The OPEX of the plant includes the costs of electricity (C_{el}), feed hydrogen (C_{fH_2} , set to 1.5 \$/kg_{H2}), mixed refrigerants (C_{MRs}), labor (C_{labor}), and maintenance ($C_{maintain}$) as shown in Eq. 8. The annual electricity cost is calculated as in Eq. 9.

$$OPEX = C_{el} + C_{fH_2} + C_{MRs} + C_{labor} + C_{maintain} \quad (8)$$

$$C_{el} = c_{el} \times m_{H_2,a} \times SEC \quad (9)$$

where c_{el} is the price of electricity (set to 0.06 \$/kWh), $m_{H_2,a}$ is the total produced mass of hydrogen in a year (in kg/year), and SEC is the specific energy consumption of the process (in kWh/kg). The annual liquid hydrogen produced per year is calculated as in Eq. 10:

$$m_{H_2,a} = AR \times \dot{m}_{H_2} \left(\frac{kg}{hr} \right) \times 8760 (hrs/year) \quad (10)$$

where AF is the annual rate of the plant activity (ratio of the actual working hours of the plant to the total hours of a year). The labor and mixed refrigerant costs are taken as 0.3% of the CAPEX while the maintenance cost is set to 2% of the CAPEX. Thus, the total refrigerants, labor, and maintenance costs are calculated using Eq. 11:

$$C_{MRs} + C_{labor} + C_{maintain} = 0.023 \times CAPEX \quad (11)$$

Three economic indicators—total capital investment (TCI), total annualized cost (TAC), and levelized cost of hydrogen (LCOH)—are used to evaluate the costs of the SMR/DMR systems. These indicators are defined as in Eqs. 12 to 14.

$$TCI = 1.18 \times \sum_k^m C_{BM,k} \quad (12)$$

$$TAC = CAPEX \times \frac{i(1+i)^n}{(1+i)^n - 1} + OPEX \quad (13)$$

$$LCOH = \frac{TAC}{m_{H_2,a}} \quad (14)$$

where i is the annual-interest rate (set to 10%), and n is the lifetime of the liquefaction process (set to 20 years).

3.3 Environmental assessment method

Assuming that the HLS is driven using fossil-fuel-based energy (electricity), which is unavoidable during the transition phase to blue/green hydrogen economy, the annual CO₂ emissions (ACE) of the amount of CO₂ emissions is calculated as in Eq. 15.

$$ACE = SEC \times m_{FH,yr} \times CI \quad (15)$$

where $m_{FH,yr}$ is the amount of the produced LH2 per year and CI is the CO₂ intensity in kgCO₂/kWh.

4. RESULTS AND DISCUSSION

The results of the 4E analyses performance indicators of the proposed DMR-HLS are presented and discussed in Section 4.1 to Section 4.3. Then, to confirm the economic and environmental benefits obtained from the new DMR approach presented in this study, comparison with large-scale SMR-based system is provided in Section 4.4.

4.1 Thermodynamic performance analysis

The compositions of the developed mixed refrigerants for the HPP (PMR1 & PMR) and of HPP (LMR1 & LMR2) are presented in Table 1.

Table 1. Composition of the developed MR for the HPP and HLP (mol. %).

Components	PMR1	PMR2	LMR1	LMR2
Methane	0.00	38.00	0.00	0.00
Ethane	11.80	4.00	0.00	0.00
Propane	27.20	6.70	0.00	0.00
n-Pentane	5.00	0.00	0.00	0.00
Nitrogen	0.00	23.10	0.00	0.00
Ammonia	28.00	0.00	0.00	0.00
Ethylene	14.00	27.00	0.00	0.00
i-Butane	0.00	1.20	0.00	0.00
i-Pentane	14.00	0.00	0.00	0.00
Hydrogen	0.00	0.00	10.30	6.00
Helium	0.00	0.00	73.70	94.00
Neon	0.00	0.00	16.00	0.00

These compositions are developed based on a systematic, knowledge-based optimized, and thermodynamic approach proposed by Sleiti et al. in [23]. Unlike SMR proposed in the literature, it can be noted that the composition of PMR1 and PMR2 have zero fraction of neon, helium, and hydrogen. This minimizes the SEC of the HPP as these very lightweight

refrigerants are only needed in the liquefaction part. This also provides efficient performance for the cryogenic heat exchangers as can be concluded from the match between the hot and cold composite curves of the HPP as shown in Fig. 2(a).

Similarly, the developed refrigerants for the HLP provide sufficient match between the hot and cold composite curves of the HLP as shown in Fig. 2(b). The very small gap between the hot and cold composite curves (less than 3°C) implies that the HXs work with high effectiveness, lower refrigerant flow rate, and more compact sizes. This also reduces their capital and operational costs. In contrast, SMR-based systems still have large temperature gaps between the hot/cold composite curves (typically 5-15 °C). This negatively affects their cooling capacity, increases their required sizes, and requires higher refrigerant flow. This in turn increases the SEC as well as the liquefaction costs. Furthermore, the composition of LMR2 has zero fraction of neon, which eliminates the potential of freezing problem in the components of the HLP.

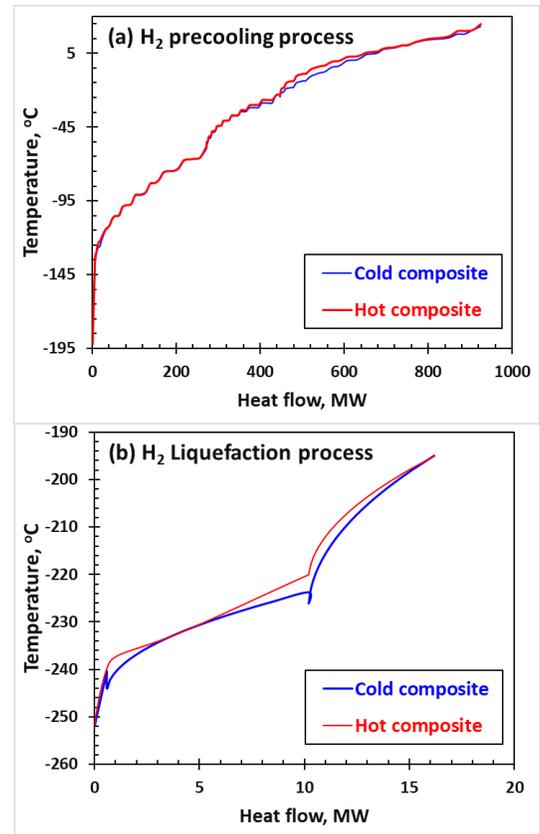


Fig. 2. Composite curves of the H₂ (a) precooling, and (b) liquefaction processes.

The main design conditions of the HPP, and HLP that are used as inputs to assess the performance of the proposed DMR-HLS are presented in Table 2. Also, Table

2 shows the SEC, COP, and exergy efficiency for each process as well as for the overall HLS. It can be noted that the overall SEC of the proposed DMR-HLS is 3.738 kWh/kg_{LH2}, which is only 13% higher than the minimum theoretical energy needed for the H₂ liquefaction (3.30 kWh/kg_{LH2}). In addition, the COP is 1.71, which is much higher than of SMR-based systems, which typically have COP less than 1. Furthermore, the exergy efficiency of the overall process is boosted to about 60% compared to 45% for SMR-based systems.

Table 2. Energy performance indicators of the proposed LH₂ production system.

Item	HPP	HLP
H ₂ mass flow rate, [kg/s]	3.45	3.45
Compressor eff., [%]	90	90
Expander efficiency, [%]	N.A	85
Inlet LH ₂ temp., [°C]	25	-195
Outlet LH ₂ temp., [°C]	-195	-253
Mass flow of MR1, [kg/s]	49.0	35.7
Mass flow of MR2, [kg/s]	35.3	9.2
High pressure of MR1, [bar]	11.9	5.6
High pressure of MR2, [bar]	39.5	3.8
HXs cooling duty, [MW]	47.7	45.8
Compression power, [MW]	10.8	39.2
Expansion power, [MW]	N.A	3.6
Total coolers' load, [MW]	21.6	38.9
SEC, [kWh/kg _{LH2}]	0.868	2.870
COP	4.420	1.283
Overall SEC	3.738	
Overall COP	1.710	
Overall exergy efficiency	59.65	

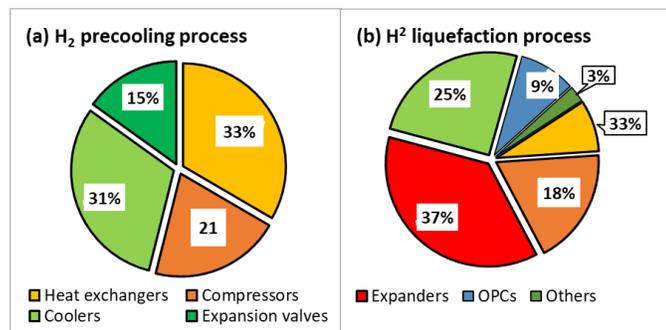


Fig. 3. Share of exergy destruction by the equipment of H₂ (a) precooling, and (b) liquefaction processes.

In terms of exergy destruction through the components of the system, it is found that the highest

exergy destruction in the HPP occurs in the cryogenic HXs (33%) followed by the coolers (31%) as shown in Fig. 3(a). In contrast, the highest exergy destruction in the HLP is caused by the expanders (37%) followed by the HXs. This implies that further improvements could be made to minimize the exergy destruction of these components and improve the energy efficiency of the whole liquefaction process.

4.2 Economic performance analysis

Fig. 4 shows the total capital investment (TCI), total annualized cost (TAC) and the levelized cost of hydrogen for the HPP, HLP, and overall DMR-HLS. It can be noted that the HLP capital cost forms about 90% of the overall cost of the HLS. This is mainly due to that the energy consumption of the HLP is about 4 times higher than of the HPP (see Table 2). In addition, the cost of the hydrogen gas fed to the process (1.50 \$/kg_{H2}) is calculated within the operational cost of the HLP. Therefore, the levelized cost of the HLP is much higher than that of the HPP. For the overall process, the levelized cost of LH₂ production using the proposed DMR_HLS is 1.89 \$/kg_{LH2}, which is 70% lower than the cost of LH₂ in current commercial plants. Also, this cost is 21% lower than of the liquefaction cost in large-scale SMR-based systems as explained in Section 4.4.

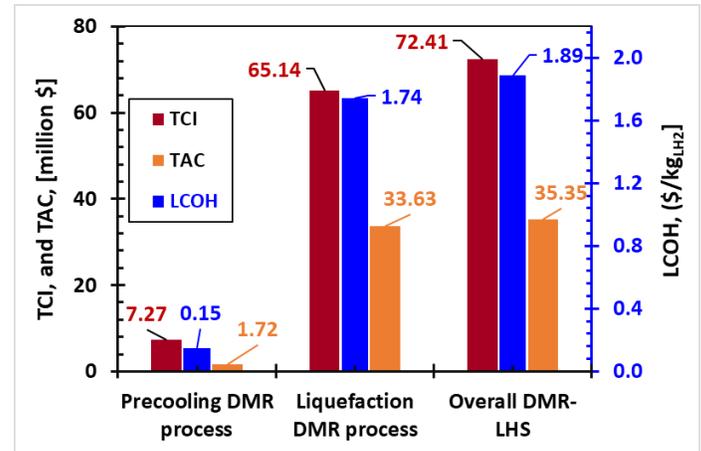


Fig. 4. Economic performance indicators of the proposed DMR-LH₂ production system.

4.3 Environmental performance analysis

The the CO₂ emissions of the proposed HLS is evaluated with other two large-scale SMR-based systems (presented by Faramarzi et al. [24], and Sadaghiani et al. [19]) as shown in Fig.5. The proposed process in [24] is driven by SMR with the use of LNG in the precooling process. The SEC of this process is 8.85 kWh.kg_{LH2} with production capacity of 369 tons per day (TPD). Also, the proposed process in [19] utilizes SMR and has SEC of 4.78

kWh/kg_{LH2} with production capacity of 300 TPD. It is assumed that the electricity is supplied from NG-based power plant with 0.0411 kgCO₂/kWh [25]. It is found that the present DMR-HLP reduces CO₂ by 29% to about 70% compared to the large-scale SMR systems (see Fig. 5).

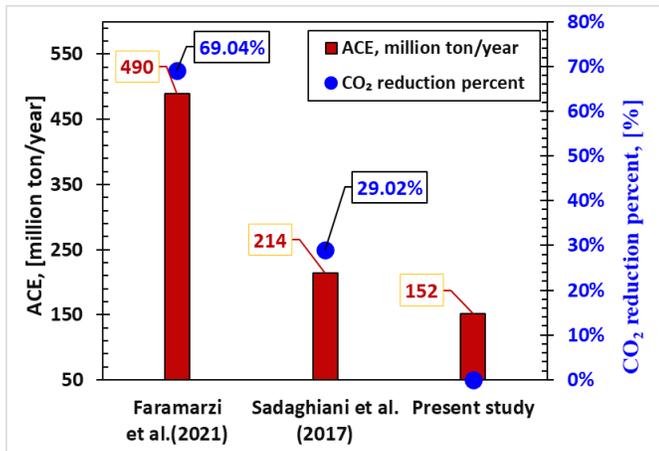


Fig. 5. Annual CO₂ emissions of the proposed DMR-HLS system compared to large-scale SMR-based systems.

4.4 Comparison with other cryogenic LH₂ systems

Limited studies in the literature have performed energy, exergy, and economic analyses for their SMR-based proposed systems. The key results are presented in Table 3. Also, the annual CO₂ emissions of these systems are calculated by the authors using Eq. 15. It can be noted that the liquefaction capacity of these systems ranges from 0.024 TPD (small-scale) to higher than 100 TPD (large-scale). On average bases, the SEC of the present DMR-HLS is 48% lower than of the large-scale SMR-based system. Also, the LCOH is 21% lower than these large-scale systems. Compared to small-scale systems, the proposed system DMR-HLS archives 37% reduction in the SEC, and 70% reduction in the LCOH. This confirms that applying the DMR approach to design and operate HLS sufficiently minimizes the energy consumption of the LH₂ production process. In addition, the economic feasibility of the liquefaction system is improved by the substantial reduction on the CAPEX and OPEX of the LH₂ production systems. This, in turns, accelerates the transition to the future liquid hydrogen-based energy storage and transportation infrastructure.

CONCLUSIONS

This work proposes a novel dual mixed-refrigerant (DMR)-based hydrogen liquefaction system (HLS) for LH₂ production. The proposed DMR-HLS is introduced to minimize the specific energy consumption (SEC) of the conventional liquefaction systems that use pure and/or

single mixed refrigerants (SMR). In addition, the proposed system addresses the potential refrigerant freezing problem noted in the SMR-based systems. Furthermore, this study performs thorough energy, exergy, economic, and environmental (4E) analyses for the proposed DMR-HLS. This fills a significant gap in the SMR-based studies that lack economic and environmental assessments to ensure their feasibility.

Table 3. Comparison of the proposed DMR-HLS with other SMR-based systems available in the literature.

Refs.	Working fluids	Capacity, [TPD]	SEC, [kWh/kg]	Ex eff., [%]	LCOH [\$/k]	ACE, [MtCO ₂ /yr]
[26]	SMR	0.024	5.90	51.4	6.32	326
[27]	LA-SMR	50	7.25	53.3	3.00	401
[22]	CO ₂ -SMR	100	7.63	31.4	2.16	422
[24]	LNG-SMR	369	8.85	47.0	2.07	490
[19]	SMR	300	4.78	47.9	2.37	214
This study	DMR	300	3.73	59.7	1.89	152

The main findings of this study can be summarized as:

- The present DMR-HLS reduces the SEC to 3.732 kWh/kg_{LH2}, 48% lower than the average SEC of the available theoretical SMR-based systems.
- The proposed DMR-HLS system increases the exergy efficiency by 33% compared to the efficiency of the SMR-based liquefaction systems.
- The levelized cost of H₂ (LCOH) is reduced to 1.89 \$/kg_{LH2}, which is 21% lower than the average cost of the large-scale SMR-based systems (2.40 \$/kg_{LH2}).
- Using dual mixed refrigerants eliminates the potential of freezing problem in the liquefaction part of the LH₂ production system.

In closing, this study's presented dual mixed refrigerant approach provides guidelines for future research to improve the efficiency of the LH₂ production system. This approach improves the technical and economic feasibility of LH₂ production, which makes it competitive with other energy storage and transportation options. As a future work, advanced exergoeconomic analyses under design and off-design

conditions for the proposed DMR-based hydrogen liquefaction system is recommended.

ACKNOWLEDGEMENT

The work presented in this publication was made possible by QUPD – CENG – 23/24 – 537 Project funded by Qatar University Office of VP for Research and NPRP 14S-0321-210080 grant from the Qatar National Research Fund (a member of Qatar Foundation). The findings herein reflect the work and are solely the responsibility of the authors.

REFERENCE

[1] M. Otto, K. L. Chagoya, R. G. Blair, S. M. Hick, and J. S. Kapat, "Optimal hydrogen carrier: Holistic evaluation of hydrogen storage and transportation concepts for power generation, aviation, and transportation," *J. Energy Storage*, vol. 55, no. PD, p. 105714, 2022, doi: 10.1016/j.est.2022.105714.

[2] J. Incer-Valverde, L. J. Patiño-Arévalo, G. Tsatsaronis, and T. Morosuk, "Hydrogen-driven Power-to-X: State of the art and multicriteria evaluation of a study case," *Energy Convers. Manag.*, vol. 266, no. March, 2022, doi: 10.1016/j.enconman.2022.115814.

[3] M. G. Rasul, M. A. Hazrat, M. A. Sattar, M. I. Jahirul, and M. J. Shearer, "The future of hydrogen: Challenges on production, storage and applications," *Energy Convers. Manag.*, vol. 272, no. September, p. 116326, 2022, doi: 10.1016/j.enconman.2022.116326.

[4] Hydrogen Council, "Hydrogen for Net-Zero. A critical cost-competitive energy vector," 2021. [Online]. Available: <https://hydrogencouncil.com/wp-content/uploads/2021/11/Hydrogen-for-Net-Zero.pdf>

[5] F. Eljack and M.-K. Kazi, "Prospects and Challenges of Green Hydrogen Economy via Multi-Sector Global Symbiosis in Qatar," *Front. Sustain.*, vol. 1, pp. 1–15, 2021, doi: 10.3389/frsus.2020.612762.

[6] M. Bailera, N. Kezibri, L. M. Romeo, S. Espatolero, P. Lisbona, and C. Bouallou, "Future applications of hydrogen production and CO₂ utilization for energy storage: Hybrid Power to Gas-Oxycombustion power plants," *Int. J. Hydrogen Energy*, vol. 42, no. 19, pp. 13625–13632, 2017, doi: 10.1016/j.ijhydene.2017.02.123.

[7] H. Nazir et al., "Is the H₂ economy realizable in the foreseeable future? Part II: H₂ storage, transportation, and distribution," *Int. J. Hydrogen Energy*, vol. 45, no. 41, pp. 20693–20708, 2020, doi: 10.1016/j.ijhydene.2020.05.241.

[8] D. Hjeij, Y. Biçer, and M. Koç, "Hydrogen strategy as an energy transition and economic transformation

avenue for natural gas exporting countries: Qatar as a case study," *Int. J. Hydrogen Energy*, vol. 47, no. 8, pp. 4977–5009, 2022, doi: 10.1016/j.ijhydene.2021.11.151.

[9] S. S. Mukherjee, H. A. Meshram, D. Rakshit, and B. B. Saha, "A comparative study of sensible energy storage and hydrogen energy storage apropos to a concentrated solar thermal power plant," *J. Energy Storage*, vol. 61, no. January, p. 106629, 2023, doi: 10.1016/j.est.2023.106629.

[10] A. K. Sleiti, W. A. Al-Ammari, F. Musharavati, and M. A. Rahman, "Techno-economic assessment of low-carbon hydrogen export from the Middle East to the Asia-Pacific, and Europe," *Energy Sources, Part B Econ. Planning, Policy*, vol. 18, no. 1, p. 2254764, 2023, doi: 10.1080/15567249.2023.2254764.

[11] A. Naquash et al., "Performance enhancement of hydrogen liquefaction process via absorption refrigeration and organic Rankine cycle-assisted liquid air energy system," *Energy Convers. Manag.*, vol. 254, no. September 2021, p. 115200, 2022, doi: 10.1016/j.enconman.2021.115200.

[12] D. Lee et al., "Integrating hydrogen liquefaction with steam methane reforming and CO₂ liquefaction processes using techno-economic perspectives," *Energy Convers. Manag.*, vol. 245, no. August, p. 114620, 2021, doi: 10.1016/j.enconman.2021.114620.

[13] A. Ebrahimi, M. H. M. Saharkhiz, and B. Ghorbani, "Thermodynamic investigation of a novel hydrogen liquefaction process using thermo-electrochemical water splitting cycle and solar collectors," *Energy Convers. Manag.*, vol. 242, p. 114318, 2021, doi: 10.1016/j.enconman.2021.114318.

[14] A. K. Sleiti, W. A. Al-Ammaria, F. Musharavati, W. A. Al-Ammari, and F. Musharavati, "Theoretical and experimental study on the selection of pure and mixed refrigerants for a power-cooling system driven by ultra-low-grade heat," *Therm. Sci. Eng. Prog.*, vol. Under Revi, p. 101962, 2023, doi: <https://doi.org/10.1016/j.tsep.2023.101962>.

[15] L. Yin and Y. Ju, "Review on the design and optimization of hydrogen liquefaction processes," *Front. Energy*, vol. 14, no. 3, pp. 530–544, 2020, doi: 10.1007/s11708-019-0657-4.

[16] B. Ghorbani, M. H. M. M.-H. Hamed, R. Shirmohammadi, M. H. M. M.-H. Hamed, and M. Mehrpooya, "Exergoeconomic analysis and multi-objective Pareto optimization of the C3MR liquefaction process," *Sustain. Energy Technol. Assessments*, vol. 17, pp. 56–67, 2016, doi: 10.1016/j.seta.2016.09.001.

[17] H. Ansarinassab and M. Mehrpooya, "Advanced exergoeconomic analysis of a novel process for

production of LNG by using a single effect absorption refrigeration cycle,” *Appl. Therm. Eng.*, vol. 114, pp. 719–732, 2017, doi: 10.1016/j.applthermaleng.2016.12.003.

[18] H. Ansarinasab, M. Mehrpooya, and M. Sadeghzadeh, “An exergy-based investigation on hydrogen liquefaction plant-exergy, exergoeconomic, and exergoenvironmental analyses,” *J. Clean. Prod.*, vol. 210, pp. 530–541, 2019, doi: 10.1016/j.jclepro.2018.11.090.

[19] M. S. Sadaghiani and M. Mehrpooya, “Introducing and energy analysis of a novel cryogenic hydrogen liquefaction process configuration,” *Int. J. Hydrogen Energy*, vol. 42, no. 9, pp. 6033–6050, 2017, doi: 10.1016/j.ijhydene.2017.01.136.

[20] G. Skaugen, D. Berstad, and Ø. Wilhelmsen, “Comparing exergy losses and evaluating the potential of catalyst-filled plate-fin and spiral-wound heat exchangers in a large-scale Claude hydrogen liquefaction process,” *Int. J. Hydrogen Energy*, vol. 45, no. 11, pp. 6663–6679, 2020, doi: 10.1016/j.ijhydene.2019.12.076.

[21] A. K. Sleiti, W. A. Al-Ammaria, and S. Ghani, “Novel dual-mixed refrigerant precooling process for high capacity hydrogen liquefaction plants with superior performance,” *J. Energy Storage*, vol. 66, no. February, p. 107471, 2023, doi: <https://doi.org/10.1016/j.est.2023.107471>.

[22] A. Naquash, M. A. Qyyum, S. Min, S. Lee, and M. Lee, “Carbon-dioxide-precooled hydrogen liquefaction process: An innovative approach for performance enhancement–Energy, exergy, and economic perspectives,” *Energy Convers. Manag.*, vol. 251, no. July 2021, p. 114947, 2022, doi: 10.1016/j.enconman.2021.114947.

[23] A. K. Sleiti and W. A. Al-Ammari, “Systematic thermodynamic approach for designing mixed refrigerants used in hydrogen precooling process,” *Int. J. Hydrogen Energy*, vol. 47, no. 48, pp. 20915–20931, 2022, doi: 10.1016/j.ijhydene.2022.04.233.

[24] S. Faramarzi, S. M. M. Nainiyan, M. Mafi, and R. Ghasemiasl, “A novel hydrogen liquefaction process based on LNG cold energy and mixed refrigerant cycle,” *Int. J. Refrig.*, vol. 131, no. March, pp. 263–274, 2021, doi: 10.1016/j.ijrefrig.2021.07.022.

[25] A. K. Sleiti, W. A. Al-ammari, R. Arshad, and T. El Mekkawy, “Energetic, economic, and environmental analysis of solid oxide fuel cell-based combined cooling, heating, and power system for cancer care hospital,” *Build. Simul.*, vol. 15, pp. 1437–1454, 2022, doi: <https://doi.org/10.1007/s12273-021-0865-9>.

[26] A. Naquash et al., “Hydrofluoroolefin-based mixed refrigerant for enhanced performance of hydrogen

liquefaction process,” *Int. J. Hydrogen Energy*, no. xxxx, pp. 1–15, 2022, doi: 10.1016/j.ijhydene.2022.02.010.

[27] Y. Yang et al., “A novel integrated system of hydrogen liquefaction process and liquid air energy storage (LAES): Energy, exergy, and economic analysis,” *Energy Convers. Manag.*, vol. 280, no. October 2022, p. 116799, 2023, doi: 10.1016/j.enconman.2023.116799.