

Conceptual design and analysis of a novel system coupling hydrogen liquefaction with multi-energy liquid air energy storage

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Abstract—This study proposed a novel coupling hydrogen liquefaction - multi-energy liquid air energy storage (M-LAES) system, aiming to reduce the energy consumption of hydrogen liquefaction while realizing the cascade utilization of cold energy in M-LAES. In the proposed coupling system, the M-LAES characterizes the delivery of the cold capacity by methanol and propane, pre-cooling hydrogen to 100 K in hydrogen liquefaction instead of conventional liquid nitrogen. A transient thermodynamic model is built to investigate the operating characteristics of the proposed system. Considering the specific energy consumption (SEC) as the objective function, the optimum flow rate and thermodynamic parameters can be determined. Compared with traditional hydrogen liquefier, the proposed system shows better performance for its lower SEC and higher exergy efficiency, about 8.745 and 32.18%, respectively. The exergy analysis shows the coupling system increases the energy efficiency of both M-LAES and hydrogen liquefaction. The proposed system outperformed the conventional LAES operation on flexibility. Energy input into M-LAES can be transformed into liquid hydrogen, instead of only electricity, opening up further possibilities for fuel cells, long-distance transport, and future clean energy management net options.

Keywords—Hydrogen liquefaction, Liquid air energy storage, System integration, Process optimization

I. INTRODUCTION

Renewable energy sources, owing the low level of greenhouse gas emissions, are rapidly developed to meet the climate protection international agreements. The contribution of renewable energy sources to the global energy supply has been emphasized significantly in the last decade throughout the world as in [1-3]. Meanwhile, with the focus on renewable energy sources, energy storage technologies are emerging to offset the intermittency and instability of

renewable energy. Among various energy storage techniques, liquid air energy storage (LAES) is supposed to play a significant role, especially in bulk energy storage, for its high energy density and free of geographical conditions.

However, the optimum generation rate in conventional LAES is only around 55% [4]. Recently, in order to achieve the delivering multiple energy vectors from LAES systems which are always overlooked in past, multi-energy liquid air energy storage (M-LAES) systems are proposed, rather than just serving as electricity generation. In M-LAES systems, the thermal capacity like cooling and heat are also regarded as the products meeting the demand of district energy systems which will improve the energy efficiency of LAES. Thus, some systems coupling with M-LAES are emerging. A system integrated the biomethane liquefaction with LAES was proposed in [5]. The cooling capacity of liquid air during gasified reduces the cooling duty of biomethane liquefaction, with a 42% improvement in the exergy efficiency. He et al. designed a system coupling LNG gasified with LAES [6]. The exergy efficiency achieved 73.92% by cascade utilizing the cooling capacity of LNG to cool the air in LAES after compressions. Andrea Vecchi quantified the techno-economic value of M-LAES under the different demands for power, heating, and cooling in [4]. The results demonstrated the availability of offering the multi-energy from M-LAES again.

Most of the research on M-LAES are focusing on compressed heat energy, while the cooling capacity of M-LAES is often ignored. Thus, considering the temperature of cooling capacity is between -95°C - 15°C [4], which just meets the need for hydrogen liquefaction pre-cooling section, a novel system coupling hydrogen liquefaction and M-LAES is proposed.

Compared with other renewable energy sources, hydrogen has its advantages of high storage capacity, long-distance transporting, and flexibility. At the same time, hydrogen contains much larger chemical energy even

compared with other hydrocarbon fuels, about 142 MJ/kg. Thus, long-term hydrogen energy development strategies and hydrogen economy have been attached by many countries. Energy storage and transportation are concerned as the keys to the utilization of hydrogen. Among the typical hydrogen storage solutions like compressed hydrogen, physisorption, and chemical storage, liquefaction hydrogen provides perfect performance on gravimetrically and volumetrically efficient. The volume density of liquid hydrogen can reach 70.8 kg/m^3 , which is even higher than that of solid hydrogen, that is, 70.6 kg/m^3 . Thus, liquid hydrogen is supposed to be the most effective medium-term to supply larger refueling stations in the absence of a pipeline network. However, hydrogen liquefaction is expensive, has low exergy efficiency, and is limited in capacity at present, which are mostly related to the process design of liquefaction. Therefore, it is necessary to pay attention to the high-efficiency hydrogen liquefaction process and the further optimization of the key parameters.

For the conversion temperature of hydrogen is lower than ambient temperature, the pre-cooling is essential to the liquefaction process. After pre-cooling, the further refrigerant cycle is developed which is always divided into Linde–Hampson cycle and the Claude cycle. In these two, the Claude is the basis of almost all the existing large-scale systems (≤ 5 TPD) for its better performance [7]. As a variation of the Claude cycle, the Brayton cycle uses a separate closed expander refrigeration cycle instead of extracting hydrogen from the feed stream. This improvement makes the choice of refrigerants more flexible rather than just hydrogen itself, attracting attention these years. For the pre-cooling, the typical hydrogen liquefaction pre-cooling is generally by liquid nitrogen for a cheap cost. However, researchers have increasingly noticed the importance of using design flexibility in pre-cooling to reduce the energy consumption of liquefaction. The IDEALHY project designs a liquefier combining the MR pre-cooling with a dual Brayton refrigeration cycle, with a capacity of 50 TPD [2]. Cardella et al. proposed a 100 TPD Claude cycle liquefier with a mixed refrigerant JT precooling. The SEC and exergy efficiency can achieve 6.2 and 43%, respectively [8]. A high-efficiency mixed refrigerant (MR) pre-cooling liquefier is proposed by Quack [9], with the exergy efficiency of about 56.8%. These conceptual designs verified the importance of the reasonable distribution of cooling capacity in pre-cooling, which can improve the low efficiency of hydrogen liquefaction. Thus, for security and cascade utilization, the cooling capacity from M-LAES will deliver to the pre-cooling of hydrogen liquefier by methanol and propane respectively in this proposed coupling system.

In this present paper, a novel system coupling M-LAES and hydrogen liquefaction using the Claude cycle with a capacity of 5 TPD is proposed. This system aims to reduce the energy consumption of hydrogen liquefaction and at the same time improve the energy efficiency of M-LAES by delivering the excess energy. And after analyzing the process, the system is simulated to determine the operating parameters with Aspen Hysys. Further, the optimization is developed to minimize the specific energy consumption (SEC) in hydrogen liquefaction. After that, the sensitivity analysis is presented to analyze the characteristics of the main parameters. Finally, the exergy analysis is described to evaluate the improvement in energy efficiency.

II. PROPOSED SYSTEM

A. Process Description

As shown in Fig. 1, the system consists of M-LAES and hydrogen liquefaction, holding the capacity of 63.26 MW and 5 TPD, respectively. The operation can be divided into charging mode, storage mode, and generation mode.

During the charging mode, the energy from the electricity sources is sent to run the compressors in M-LAES and hydrogen liquefaction. In M-LAES, the air is liquefied by a Linde-Hampson process for a mature and simple technique. The thermal media is employed to store and transfer the cold and heat energy. After cooling down the air, the thermal media will be stored in the tanks respectively. The hot thermal media working between 293-463 K is applied to store the heat energy of compressed air. The propane and methanol are employed to recover the low-temperature energy, working at 93-198 K and 183-258 K, respectively. And the excess cold methanol and propane will be sent to hydrogen liquefaction. In hydrogen liquefaction, the pre-cooling cycle, Claude cycle, and Joule-Thomson (JT) cycle consist the main part. The feed hydrogen is firstly pre-cooled by methanol in heat exchanger-1 to 185 K and then by propane in heat exchanger-2 to about 95 K. After pre-cooling, the isothermal ortho-para conversion catalyst (Co-1) is applied. A hydrogen Claude cycle generates further refrigeration to about 30 K. For the para hydrogen concentration increasing dramatically in this temperature range, two adiabatic ortho-para conversion catalysts (Co-2 and Co-3) are introduced. Finally, the liquid hydrogen of about 20 K can be obtained by Joule-Thomson expansion.

During the storage mode, the energy is stored in liquefied air and hydrogen. In M-LAES, the surplus electricity in charging mode is converted into potential electricity, and stored in liquefied air. And the compression heat and condensation heat are stored in liquefied air, diathermic oil heat storage, and methanol-propane cold storage, respectively. At the same time, in the hydrogen energy storage, the liquefied hydrogen is stored, which will be later transported to downstream of the energy consumption.

During the generation mode, the liquid air generates to provide electricity for hydrogen liquefaction. In the M-LAES, considering the loss, 99.5% of liquid air is stored reheated in different steps, and then expands through a turbine train to generate electricity. Similar to the above, the excess cooling capacity will be sent to pre-cooling hydrogen by methanol and propane. And the pre-cooled hydrogen will be liquefied by the same Claude cycle and JT cycle but powered by M-LAES.

B. Process Simulation

The transient thermodynamic model of the proposed process is built to discuss the operating characteristics. The Peng-Robinson equation of state is employed for its accuracy and simplicity in the simulation of organic and hydrogen. Several assumptions based on industrial practice are proposed as follows:

- 1) The feed hydrogen has been purified, and compressed to 2100 KPa; The product of liquid hydrogen holds the temperature and pressure at 21 K and 130 KPa, respectively; The liquid air stored in the tank is set at 86.5 K, 200 KPa;

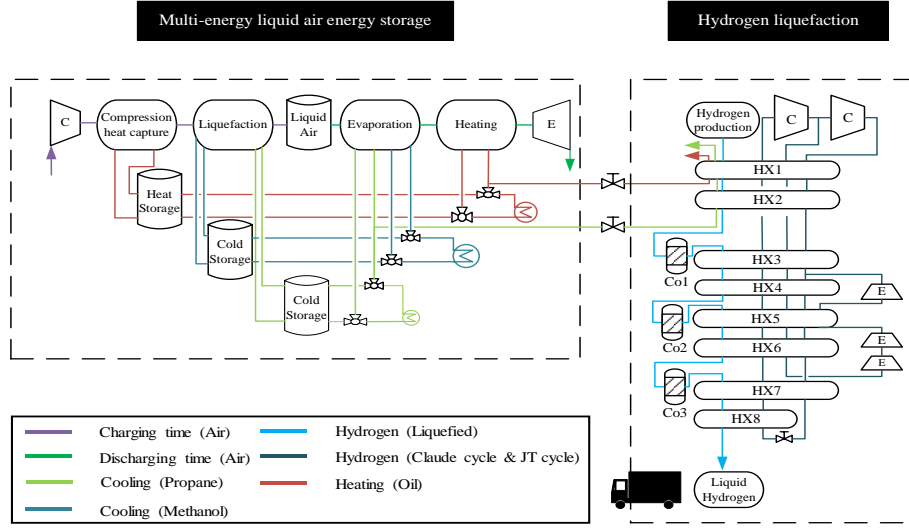
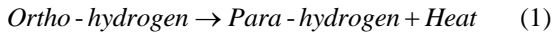


Fig. 1. Process flow diagram of the proposed system.

2) The adiabatic efficiencies of compressors and expanders in M-LAES and hydrogen liquefaction are 75%;

3) The pressure drops of the inter-cooler and heat exchangers are ignored.

The ortho-para conversion is one of the identical unique properties of hydrogen, which needs especially attention during all design about the field of hydrogen. Hydrogen is composed of para-hydrogen and ortho-hydrogen these two spin isomers, and the concentration of these two is highly related to temperature. The equilibrium hydrogen at ambient temperature is composed of 25% para-hydrogen and 75% ortho-hydrogen. However, the equilibrium liquid hydrogen at about 20 K is almost formed of para-hydrogen. This concentration change of equilibrium hydrogen in the different temperatures is called ortho-para conversion. This conversion is accompanied by heat released, which is even greater than the latent heat of vaporization, for the lower energy level of para-hydrogen. In addition, the reaction rate is quite slow to the product hydrogen has not completed this conversion. As a result, the remaining conversion reacting in the liquid hydrogen tank will extremely influence the storage. Thus, to induce the conversion, the ortho-para conversion catalytic reactor packing of $\text{Fe}(\text{OH})_3$ is carried out in three stages, one of which is kept isothermal and the remaining two are adiabatic. The ortho-para conversion can be simulated as (1):



The conversion percentage is set as (2):

$$\text{Conversion} = C_0 + C_1 * T + C_2 * T^2 \quad (2)$$

The coefficients of polynomials can be matched to meet the experimental data. Table 1 shows the concentration of

TABLE I. THE CONCENTRATION OF THE PARA-HYDROGEN IN THE STREAMS AT THE INLET AND OUTLET OF THE CATALYTIC REACTORS.

Catalytic Reactor ID	Temperature (K)	Para proportion (%)	
		Inlet	outlet
Co-1	98.15	25	39
Co-2	55.15	39	69.7
Co-3	29.95	69.7	97

the para-hydrogen in the streams at the inlet and outlet of the catalytic reactors.

III. METHODOLOGY

A. Index of Hydrogen Liquefaction

Specific energy consumption (SEC) and coefficient of performance (COP) expressed in (3) and (4) are introduced to evaluate the influence of the coupling system on the hydrogen liquefaction sub-process. SEC represents the ratio of total energy consumption of hydrogen liquefaction and the mass flow of production. This parameter shows intuitively the energy consumption of the hydrogen liquefaction.

$$\text{SEC} = W_1 / m_{\text{product}} = (\sum W_C - \sum W_E) / m_{\text{product}} \quad (3)$$

Where, W_1 , W_C , W_E is the energy consumption of the whole hydrogen liquefaction process, compressors, and expanders, respectively, kW; m_{product} is the mass flow of the liquid hydrogen, kg/h.

COP is the ratio of the cooling capacity and the total energy consumption in hydrogen liquefaction. This parameter can apparently represent the performance of a refrigeration cycle.

$$\text{COP} = m_{\text{product}} * (h_{\text{feed}} - h_{\text{LH}_2}) / W_1 \quad (4)$$

Where, h_{feed} , h_{LH_2} is the enthalpy of the feed hydrogen and liquid hydrogen, kJ/kg.

B. Exergy Analysis

Exergy is introduced to show the influence of extracting the multi-energy from M-LAES to the coupling system. Exergy is an important index to assess the quantity of energy. The exergy analysis is applied to evaluate the energy conversion in the system with various kinds of energy. Moreover, the exergy destruction is an indication to further improve the performance of the system.

The exergy of streams consists of the physical exergy, the chemical exergy, the kinetic exergy, and the potential exergy. The physical exergy of streams expressed in (5) stands for

the maximum useful work between the specific state and the reference state.

$$Ex_x^{PH} = n * ex_x^{PH} = n * [(h - h_0) - T_0(s - s_0)] \quad (5)$$

Where, h , h_0 are the enthalpy stream and its reference state, kJ/kg; s , s_0 are the entropy of stream and its reference state, kJ/kg; n is the mass of the stream, kg.

The chemical exergy, the kinetic exergy, and the potential exergy of streams are negligible. The exergy of electricity can be indicated in (6):

$$Ex_{\text{electricity}} = E_{\text{electricity}} \quad (6)$$

Exergy efficiency expressed in (7) is the ratio of exergy output from the system to the exergy input. This index shows the performance of the system about the maximum energy that can achieve.

$$\eta_{\text{ex}} = Ex_{\text{out}} / Ex_{\text{in}} \quad (7)$$

The exergy flow diagram is adopted to visualize the exergy flows, and the exergy destruction of the system is studied.

C. Optimization Methodology

The optimization is developed for better operation. For energy consumption is the most critical to hydrogen liquefaction, SEC minimization is employed to be the objective function. Meanwhile, the pressure before expansions in the Claude cycle, the mass flow of the Claude cycle, the mass flow of the JT cycle in hydrogen liquefaction, and the mass flow of methanol and propane delivered from M-LAES are the important parameters impacting the energy consumption and efficiency in this coupling system. Thus, these four parameters are selected to be the variables in optimization. Once the optimizer changes these variables, the optimization methodology will optimize the system to meet the objective function.

In order to ensure the optimization conforms to the thermodynamic theorem and the actual industrial production, the constraints are defined following:

- 1) The vapor phase fraction of the inlet streams of compressors is 100%;
- 2) The minimum temperature approaches in the heat exchangers are larger than 1K.

The constraints are applied during the optimization in terms of inequality/ equality functions. The sequential quadratic programming (SQP) method is supposed to be the most efficient and simple method for minimization with such constraints, with the ability of minimizes quadratic approximation of the Lagrangian function to meet the linear approximations of the constraints. The optimization is implemented in Aspen HYSYS.

IV. RESULTS AND DISCUSSION

A. Optimization Results

The optimum operation can be obtained from optimization. Compared with the base case, SEC in the optimized case decreases about 21.71% from 11.17 to 8.745. And the exergy efficiency increases by 28.03% to 32.18%.

These two improvements show the availability and importance of optimization. Moreover, compared with the other large-scale hydrogen liquefiers using liquid nitrogen pre-cooling like the Ingolstadt cycle, Leuna cycle, and Kuz'menko cycle, the optimal operation possesses the lowest SEC and better exergy efficiency, shown in Fig. 2. The better performance proves that the coupling system enhances the performance of hydrogen liquefaction with the cascade utilization of cold energy, rather than simple liquid nitrogen. For better comparison and description, the exergy efficiency discussed here is not that of the overall coupling system, but of the hydrogen liquefaction sub-system, which is represented by EXE. And the exergy efficiency of coupling system is studied later. At the same time, the changes of variables indicate their operation characteristics on power consumption and efficiency. The optimal variables are shown in Table 2, compared with the base case. For the compressors are the dominant power-consuming devices, the flow, and thermodynamic parameters of the Claude cycle and JT cycle passing compressors are mainly optimized.

It can be found in Table 2, that the hydrogen flow providing cooling capacity below 100 K in the Claude cycle and JT cycle is reduced, but with different reductions. That is, the distribution rate of the distributor is optimized considering the different demands of cooling capacity in 100 K-30 K and 30 K-20 K. The reduction after optimization also occurs in the pressure before expansions in hydrogen liquefaction. This reflects the consideration of both energy consumption in compressors and the requirement of cooling capacity in heat exchangers. Moreover, the increase in pre-cooling also results in better performance. And when extracting the cooling capacity from M-LAES, the performance of M-LAES has also been taken into account at the same time. The liquefaction capacity and power generation of M-LAES are not affected in the optimal case. However, the impact on M-LAES reflects on heat exchangers definitely, still ensuring the minimum temperature approaches in M-LAES larger than 3 K.

Heat exchangers are the key components of the hydrogen liquefaction process, in which the accurate matching of hot composite with cold composite can reflect the good performance of liquefaction. The heat transfer curves in optimal operation, with minimum temperature approach and logarithmic mean temperature difference (LMTD) of the main heat exchangers in hydrogen liquefaction, are shown in Fig. 3, compared with the base case. It can be found, that the optimized heat exchangers hold a better matching, resulting in a better heat exchange efficiency.

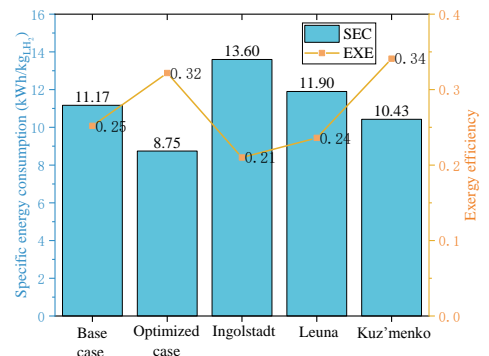


Fig. 2. Comparison of the performance of hydrogen liquefaction processes.

TABLE II. THE VALUE OF VARIABLES IN OPTIMIZED CASE AND BASE CASE

Variables	Optimized value	Base value
pressure before expansion (kPa)	2200	2083
mass flow of the Claude cycle (kg/h)	1343	1700
mass flow of the JT cycle (kg/h)	283	350
mass flow of methanol delivered from M-LAES (kg/h)	251	230
mass flow of propane delivered from M-LAES (kg/h)	3983	3000

B. Sensitivity Analysis

Considering the deviation of key parameters in different operating conditions in industrial production, the sensitivity analysis is carried out to clear the impact of key parameters. The effects of pre-expansion pressure, distribution ratio before the Claude cycle, and the mass flow of pre-cooling media on SEC, COP, and EXE are discussed. Respectively shown in Fig. 4.

As Fig. 4(a) shows, with the increase of pre-expansion pressure, SEC increases but COP and EXE decrease. This is for the power generation of expanders caused by increasing pressure is less than the increment duty of compressors. However, if the pressure is reduced to save energy consumption, the cooling capacity brought by Claude expansion will be reduced, and the cold capacity in the heat exchangers will be poor or even insufficient. Fig. 4(b) shows the influence of distributor proportion. With the decrease of the proportion to the Claude cycle, SEC decreases, and COP and EXE increase. In hydrogen liquefaction, the cooling capacity in the JT cycle is relatively fixed, so the change of proportion is often related to the flow of the Claude cycle. Thus, the power consumption of the compressors decreases more than that of the generation of expanders resulting in the SEC decreasing with the flow of the Claude cycle decreases. However, if the flow of the Claude cycle is reduced too much, the 100k-30k cooling capacity reduction definitely affects the function of the hydrogen liquefaction process. The effect of the quantity of pre-cooling media on hydrogen

liquefaction is shown in Fig. 4(c) and Fig. 4(d). With the increase of pre-cooling media, SEC decreases, and COP and EXE increase. The feed hydrogen can be perfect pre-cooling to about 100 K with sufficient pre-cooling methanol and propane, mitigating the duty of further refrigerant. Additionally, the low inlet temperature of compressors is achieved by the adequate supplies of pre-cooling capacity, reducing the total power consumption. Compared with methanol, propane has a greater impact on the system, which is related to the larger and higher quality of cooling capacity delivering in propane.

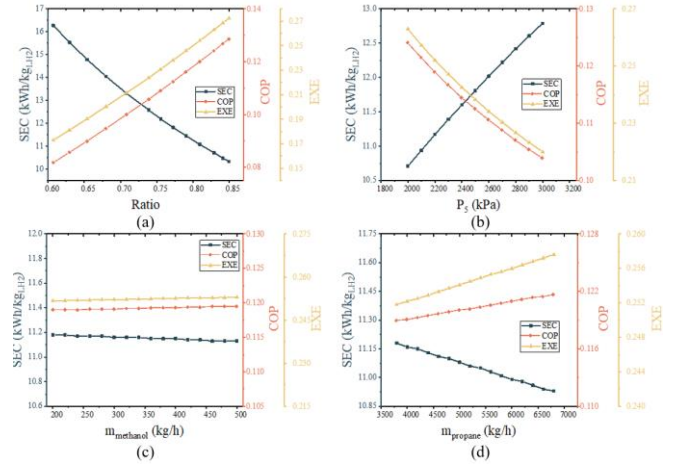


Fig. 4. Effect of parameters on hydrogen liquefaction sub-process.

C. Exergy analysis

Compared with independent LAES, the sub-process M-LAES in an integrated system with the same operation holds the exergy efficiency of 43.8%, higher than that of 43.15%. This raise proves that the excess cold energy output of M-LAES achieves the potential of multi-energy utilization in original LAES and thus improves the exergy efficiency of the energy storage system. Furthermore, with cascade utilization of this delivering cold capacity, the exergy in hydrogen liquefaction improves at the same time, as discussed in the above section.

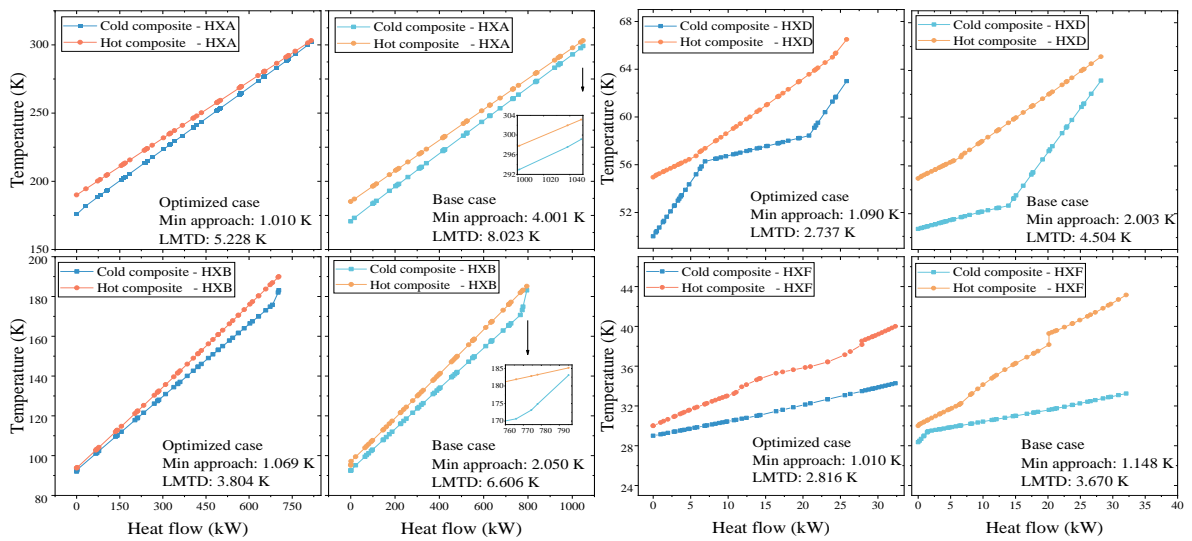


Fig. 3. Heat curves in the important heat exchangers in optimized case and base case of hydrogen liquefaction sub-process.

The energy destruction of the components in hydrogen liquefaction is shown in Fig. 5. The inter-coolers with the largest exergy destruction for the low heat transfer efficiency is the inherent problem. Following, the compressors and heat exchangers hold the relatively large energy destruction, for the friction and heat loss in compressors and the temperature difference in heat exchangers, respectively.

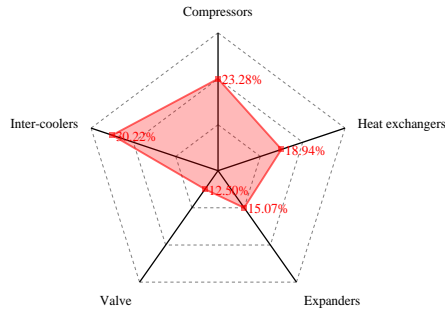


Fig. 5. Exergy destruction proportion of hydrogen liquefaction.

Fig. 6 shows the exergy destruction of some specific components, predicting further optimization. The maximum exergy destruction occurs in compressors-2, except inter-coolers. This may relate to the larger mass flow and the higher pressure ratio in compressor-2. Since the largest minimize temperature approach is about 1.572 K, heat exchanger-5 has the largest exergy loss among all the heat exchangers. And the heat exchangers in the JT cycle, like heat exchanger-7 and heat exchanger-8, perform well in exergy loss. Meanwhile, the exergy destruction in expander-3 is relatively large. Figure 7 shows the simplified version of the exergy flow diagram. The direction and quantity of energy conversion in this system can be visually found in Fig. 7. The system is divided into M-LAES liquefaction, M-LAES power generation, and hydrogen liquefaction systems. With the 63.25 MW inputting electricity, the system will output 1.01 MW of waste gas, 25.52 MW of electricity, and 0.85 MW of liquid hydrogen.

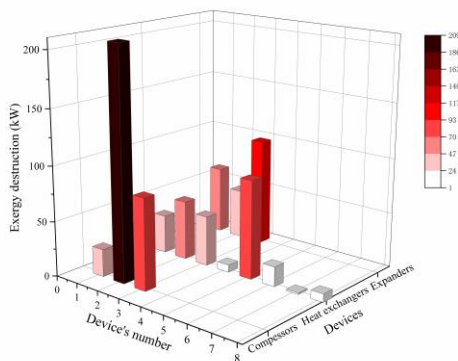


Fig. 6. Exergy destruction of dominant devices in hydrogen liquefaction.

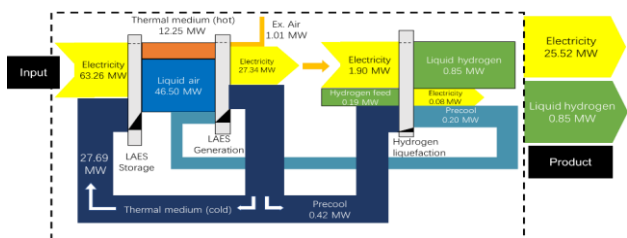


Fig. 7. Exergy flow diagram of the coupling system.

V. CONCLUSION

In this study, an integrated system coupling the M-LAES system and hydrogen liquefaction was proposed. The system is free of geographical, the capacity of the large-scale system, and advanced thermodynamic efficiency. This coupling system achieves the delivery and the cascade utilization of the excess cold energy in M-LAES. With these, the coupling system significantly improves the energy consumption in hydrogen liquefaction and the thermodynamic utilization in M-LAES.

The optimization of the coupling system was performed with the SQP method. The minimized SEC and the corresponding operation parameters were obtained with constraints. The SEC decreased from 11.17 kWh/kg to 8.745 kWh/kg and the EXE increased from 25.2% to 32.2% by optimization. Compared with other large-scale hydrogen liquefiers, the better performance in the proposed system showed its competitiveness and improvement in energy saving. Along with optimization, the sensitivity analysis of the dominant parameters was studied to illustrate the operation characteristic of the coupling system. Furthermore, a simplified version of the exergy flow diagram was presented in this paper. The exergy of this coupling system was 41.8%. The higher exergy efficiency of the two sub-process, compared with independent LAES and hydrogen liquefaction, verified the superiority of utilizing the multi-energy in LAES in the coupling system again. The exergy destructions most occurred in inter-coolers, the compressor-2, the heat exchanger-5, and expanders-3.

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