Comparative Study between precooled Claude Liquefaction Cycle and an Active Magnetic Regeneration Cycle applied to Hydrogen Liquefaction

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Abstract−The present work aims at performing a comparative study between precooled Claude cycle and an Active Magnetic Regeneration Cycle (AMR cycle) applied to hydrogen liquefaction. It deals with a comparison between performances and energy consumption evaluated for the two systems at similar operating conditions. For Claude system, energy and material balances have been performed by using Aspen Hysys simulator. Thus, liquefaction power, energy consumption and coefficient of performance (COP) have been calculated. While, the AMR system considered is constituted of 6 stages operating in cascade. Each stage contains two regenerator beds, composed of a typical magnetic material, through which a carrier fluid is forced to follow alternatively between two heat reservoirs. Thermal analysis and evaluation of performances have been performed at once by a numerical model, developed on the basis of energy equations for fluid and solid within the regenerator bed, and Aspen Hysys simulator. Using the following magnetic materials (Gd, Dy, Tb and Ho), the COP found for the AMR system is 0.096. This value is higher than Claude cycle efficiency for which a COP of 0.094 was found. In terms of energy consumption, the value found for the AMR cycle is 0.053 kW which can be neglected compared to the Claude cycle consumption (14.5 kW).

***Keywords*−Magnetic Refrigeration, Active Magnetic Refrigeration Cycle, Magnetic Materials, Hydrogen Liquefaction, Conventional Liquefaction Cycles.**

I. INTRODUCTION

Liquefaction of hydrogen has the advantage that very high hydrogen storage densities can be attained at atmospheric pressure: the density of saturated liquid hydrogen at 1 bar is 70 kg/m^3 [1]. Therefore, it is a suitable solution making possible large-scale hydrogen storage and long distance transportation. Liquid hydrogen has x characteristics such as lower weight and volume and higher energy content than the gaseous hydrogen [2]. The development of large hydrogen energy storage systems will facilitate the evolution of renewable energy sources, mitigate concerns arising from damage to the environment and eliminate the problem of energy demand fluctuations of renewable energy grids by storing the excess energy generated during times of high production to be consumed during times of low production.

Liquid hydrogen, under atmospheric pressure, can be obtained at 20.3 K. The liquefaction is carried out by extracting of 4914 kJ/kg of heat (divided between sensible heat, latent heat and conversion heat from n-H_2 to p-H_2). This liquefaction requires the use of some high level cryogenic technology whether to liquefy it or to keep it in the liquid state. In general, three processes are applied: Claude cycle, Brayton cycle and magneto-thermal cycles (i.e. magnetic refrigeration, based on the magnetocaloric effect (MCE) phenomenon) [3].

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The use of magnetic refrigeration to liquefy hydrogen is based on the magneto-caloric effect (MCE) phenomenon, which occurs in some materials when they are subjected to external magnetic field changes. The MCE is defined as the change of material temperature when applying or removing the magnetic field (magnetization /demagnetization process). In fact, if a magnetic material is placed in a magnetic field, there is usually an increase in its temperature. Conversely, demagnetization process has a cooling effect on it. Liquefaction could be carried out by cooling the gas through a thermomagnetic cycle, known as Active Magnetic Regeneration (AMR cycles) cycles. An AMR cycle consists essentially of a regenerator bed (magnetic material) which is subjected to cyclic changes in magnetic-field intensity, alternating between zero-field and the maximum field. Temperature span induced by the magnetization and demagnetization process is amplified by forcing a working fluid to move alternatively through the regenerator bed between two heat sources (hot and cold reservoirs). Thus, a large temperature span can be obtained [4].

Application of Magnetic Refrigeration to hydrogen liquefaction is performed by absorbing the liquefaction power from the gas to be liquefied via an AMR regenerator cycle. The liquefaction power absorbed is the cooling power which can be produced by the AMR cycle. This cooling power can be evaluated through modeling of magnetic refrigeration phenomena, based on characterization of heat transfer between working fluid and magnetic material and EMC calculation. In this context, several research activities related to performance investigation of AMR cycle or development of new magnetic materials have been conducted. We show here, e.g., the work of Smaili et al. [5] related to thermodynamic investigations of optimum active magnetic regenerators, Aprea et al. [6] who proposed a flexible numerical model to study an active magnetic refrigerator for near room temperature applications, Chiba et al. [7] study related to thermal investigations of an experimental active magnetic regenerative refrigerator operating near room temperature.

The interest in magnetic refrigeration for hydrogen gas liquefaction started in the early to mid-1970s by W.A. Steyert, joined by J.A. Barclay in 1977 and C.B. Zimm in 1983, at the Los Alamos National Laboratory (LANL) [8]. Later, research activities have been increased substantially and several works have been published by different researchers globally [9]- [16]. This entire work are related to exergy analysis, cooling capacity and COP investigation or design optimization of hydrogen liquefier operating between 77 K and 20K, after being precooled, in most cases by liquid nitrogen. Exception is found for Smaili et al. paper [15] where a numerical study on the first stage hydrogen magnetic liquefier operating over the temperature range: 298-233 K has been reported. The authors investigated the cooling capacity and the COP of the AMR cycle as function of mass flow rate, cycle frequency, and magnetic field.

Recently, Belkadi and Smaili [17] published in 2018 a work about thermal analysis of a multistage Active Magnetic Regenerator (AMR) cycle for hydrogen liquefaction starting from room temperature. The proposed liquefier operates with a magnetic material, assumed having a constant EMC, as refrigerant and hydrogen gaseous as carrier fluid. The number of stages, the coefficient of performance (COP) and the required volume of magnetic material has been investigated. To liquefy 1kg/h of hydrogen supplied at 298 K, a minimum required volume of 2.96 L, corresponding to COP value of 1.23 has been found for a liquefier constituted of 6 AMR cycles operating in series (cascade).

For conventional systems, liquefaction of hydrogen was first performed in 1898 by Sir James Dewar. Some years later, a pre-cooled Linde Hampson system was used as the first simple laboratory system to liquefy hydrogen [18]. Then, the process was improved by Georges Claude, the forefather of the French company ''Air Liquide'' [19].

As a review, Krasae-in *et al.* [18], published in 2010 a paper related to the development of large-scale hydrogen liquefaction processes from 1898 to 2009. This paper retraces the history of hydrogen liquefaction development starting from the first system of Sir James Dewar. A brief process description and comparison between energy consumption and overall cycle exergy efficiency were given by the authors for different liquefaction systems, including theoretical liquefaction systems (Linde-Hamson system, Helium refrigerant system and Precooled-Claude system), current plants and conceptual plants. As reported by the authors, it is found that every current plant is based on the Precooled-Claude system, which is still the same as was 50 years ago with little improvement and low exergy efficiency of just between 20–30%.

There are two main fundamental reasons as to why the liquefaction of hydrogen has low exergy efficiency and requires a substantial input of energy: (i) the extremely low boiling point of hydrogen (20 K at 1 bar) and (ii) the fact that hydrogen gas does not cool down during throttling processes (adiabatic, isenthalpic expansion) for temperatures above around 200 K (inversion temperature of hydrogen). The latter problem requires precooling in the liquefaction process, most often by the evaporation of liquid nitrogen [1].

In order to solve the problem of the low exergy efficiency of the current liquefaction plants, several research activities are oriented to find a new configuration with more efficient system. In this purpose, Krasae-in et al. [20]- [22] proposed a multi-component refrigerant (MR) refrigeration cycle for which, three studies have been published between 2010 and 2011. The first study investigates the simulation of a small-scale laboratory liquid hydrogen plant with a new innovative multi-component refrigerant (MR) refrigeration system that was capable of liquefying a feed of 2 kg/h of normal hydrogen gas at 21 bar and 298 K to normal liquid hydrogen at 2 bar and 23 K. In the second one, a test rig was constructed to verify the simulation of the proposed small-scale laboratory hydrogen liquefaction plant. The third paper presents a proposed liquid hydrogen plant using a multi-component refrigerant (MR) refrigeration system where a cycle that is capable of producing 100 tons of liquid hydrogen per day is simulated. The authors found that the overall power consumption of the proposed plant is 5.35 kWh/kgLH$_2$. The system shows very low energy consumption compared to the current plant in Ingolstadt used as a reference by the authors, which has an energy consumption of 13.58 kWh /kgLH$_2$.

In same way, a large-scale hydrogen liquefier, utilizing mixed-refrigerant (MR) pre-cooling, has been developed by Berstad *et al* [23] in 2010. The liquefier employs MR refrigerant to pre-cool hydrogen until the temperature of 75 K. Below this temperature, a reversed helium/neon Brayton cycle provides the requisite cooling. Two MR-based liquefier models have been performed. In the first, Joule–Thomson throttling valves are employed for MR expansion, while these have been replaced by liquid expanders in the second. With 21bar hydrogen feed pressure and an ambient temperature of 300 K, the resulting figures for specific liquefaction power presented by the authors for the two models are respectively 6.48 and 6.15 kWh/kgLH$_2$.

In 2017, a novel large-scale plant for hydrogen liquefying, with a production capacity of 100 tons per day, is proposed and analyzed by Aasadnia M *et al.* [24]. The liquefaction plant is proposed to provide the required LH$_2$ for a large urban area with 100 000-200 000 hydrogen vehicles supply. In the precooling section of the process, a new mixed refrigerant (MR) refrigeration cycle, combined with a Joule-Brayton refrigeration cycle, precool gaseous hydrogen feed from 298 K to the temperature of 75 K. A new refrigeration system with six simple Linde-Hampson cascade cycles cools low-temperature gaseous hydrogen from 75 K to temperature of 20 K. The process specific energy consumption is 7:69 kWh/kgLH2 as reported by the authors.

In addition, a large review and analysis of basic cycles of hydrogen liquefaction is presented by Aasadnia M et al. [2] in 2018. The review includes hybrid conceptual plants where various renewable-energy may be used in different combined configuration within hydrogen liquefaction plants. The promising role in the cost reduction and the increasing contribution of this new approach is demonstrated. As reported by the authors, results of thermodynamic analysis, of a combined refrigeration system comprising a conventional vapor-compression cycle cascaded with an absorption refrigeration cycle assisted by geothermal energy, show that the conventional cycle energy consumption is approximately 37% and 54% more than the combined system under the same operating conditions.

In the same context of assistance by geothermal energy, Gaddalla et al. [25] presented an extensive investigation for an integrated absorption cooling-hydrogen liquefaction system. The pre-cooling process of hydrogen can be achieved by using absorption system which runs on low grade heat instead of electricity.

Otherwise, enhancement of hydrogen liquefaction efficiency can also be performed by optimizing operating parameters and reducing heat losses in the main equipment of the liquefaction system. In this, approach, Skaugen et al. [26] presented a work in 2020 about exergy losses comparison and evaluation of the potential of catalyst-filled plate-fin and spiral-wound heat exchangers for use in a large-scale Claude hydrogen liquefaction process. In addition to the comparison made on characteristics, behavior and performances, maps of the local exergy destruction in the heat exchangers have also been presented. This is to reveal avenues which can be followed to further improve the process.

To reduce energy consumption, a refrigeration strategy that gives minimum entropy production/exergy destruction in a plate-fin heat exchanger that cools the hydrogen from 47.8 K to 29.3 K has been discussed in 2019 by Hande et al. [27]. Two reference cases have been studied; one where the feed stream enters at 20bar, and one where it enters at 80 bar.

In the present work, a comparative study between conventional liquefaction cycle and AMR liquefaction cycle has been performed. It deals with a comparison between performances and energy consumption evaluated for the two systems at similar operating conditions. This is to situate the AMR systems, which are now at laboratory scale, in relation to conventional systems which are already industrialized.

For the first one, precooled Claude cycle has been considered. Here, energy and material balances have been performed by use of Aspen Hysys simulator. Thus, liquefaction power, energy consumption and coefficient of performance (COP) have been calculated. While, the second one is based on a multistage AMR cycles operating in cascade. Each stage contains two regenerator beds, composed of appropriate typical magnetic material. thermal analysis and performance calculation have been performed. First, for the AMR cycle by considering energy equations for fluid and solid within the regenerator bed. A numerical model has been developed in order to determine heats exchanged between the solid (magnetic material) and the carrier fluid at each stage. Then, heats exchanged are introduced as input in Aspen Hysys simulator to perform energy and material balances and calculate performances of the liquefier (COP, work input, etc.). For this purpose, a simulation method published by Belkadi and Smaili [17] has been used to simulate the AMR liquefier with Aspen Hysys which allows rapid and rigorous calculations.

II. CONVENTIONAL BASIC CYCLES APPLIED TO HYDROGEN LIQUEFACTION

As mentioned before, every current plant is based on the Precooled-Claude system. Improvement has been conducted, for often, in the precooling section. In this section, a brief description of basic cycles of hydrogen liquefaction is presented.

A. *Basic Claude cycle*

The basic Claude cycle uses hydrogen itself as working medium after being compressed, chilled and expanded through a Joule-Thomson Valve (JT valve). A part of compressed H_2 gas is expanded in the expansion turbine to generate colder gas. This colder gas is combined with saturated vapor, coming from the flash end separator, to be used for cooling of the compressed hydrogen gas (figure 1). Energy analysis performed by Asadria et al. [2] illustrates a COP of 0.055 and a specific energy consumption of 22.1 kWh/kgLH$_2$.

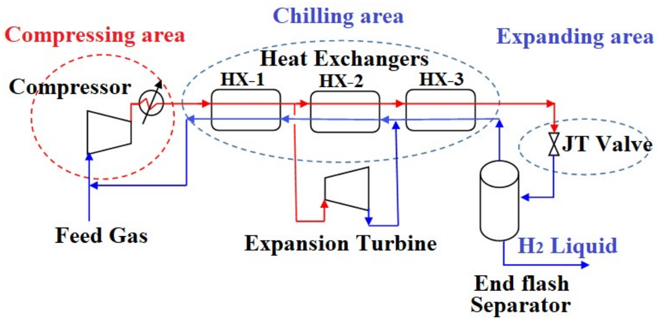

Fig. 1 Simple Claude cycle of hydrogen liquefaction.

B. *Brayton cycle.*

Brayton cycle uses helium, often mixed with neon, as an external refrigerant as illustrated in figure 2. It includes:

- A compressor to bring helium from 3 to 17 bars,
- Two parallel turbines expanding helium gas from 17 to 3 bars,
- An ambient-refrigerant heat-exchanger to cool the compressed helium.
- A cold box where hydrogen is first precooled by nitrogen and then liquefied by helium refrigerant after being expanded to 3 bars.

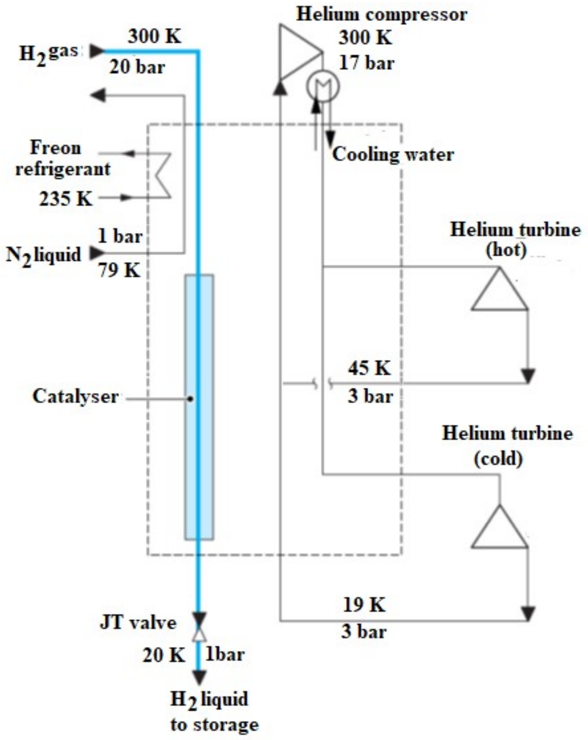

Fig.2 Brayton liquefaction cycle [adapted from 19]

C. *Liquid-nitrogen pre-cooled cycles*

As the maximum inversion temperature of hydrogen (i.e. 205 K) is below ambient, pre-cooling is an inevitable necessity of the basic simple cycle modification. Basic Claude cycle, Brayton cycle and Linde-Hampson system that utilized for air liquefying can be improved, by adding a precooling medium, e.g. liquid nitrogen bath (LN$_2$ bath), to be used for hydrogen liquefaction [2]. Fig. 3 depicts the process flow diagram (PFD) of a LN$_2$ pre-cooled Linde-Hampson cycle applied for hydrogen liquefaction.

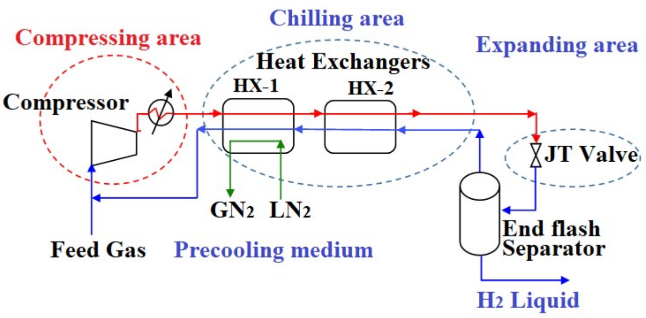

Fig. 3 Pre-cooled Linde-Hampson cycle for hydrogen liquefaction.

D. *Precooled Claude cycle*

In Precooled Claude system, liquefaction of hydrogen at 20.3 K is obtained by the combined effect of cooling and adiabatic expansion of gas after it has been previously compressed as illustrated in figure 4. Expansion can be carried out through two turbines (hot turbine and cold turbine) followed by a Joule-Thomson valve (JT valve) to avoid liquid formation at the end of the cold turbine.

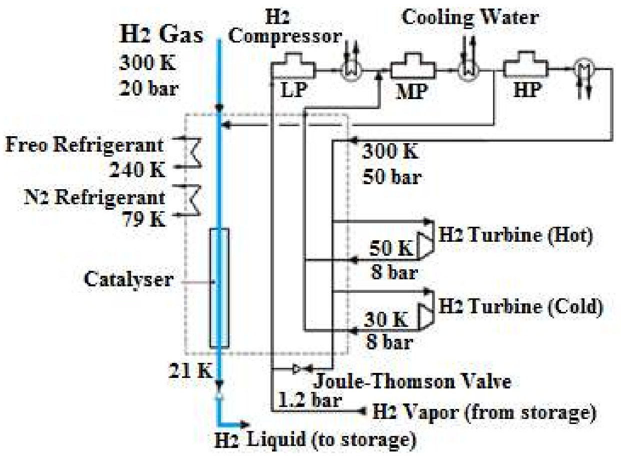

Fig. 4 Precooled Claude liquefaction Cycle [adapted from 19].

The optimum pressure for hydrogen liquefaction is generally above the critical pressure, between 15 and 25 bars. It can be supplied at the liquefaction pressure if it is produced by a hydrocarbon or methanol reforming. It should be compressed before purification and liquefaction if it is produced by an electrolyze device operating at atmospheric pressure [19].

In order to avoid hydrogen liquid losses which can be involved by the heat of conversion, a catalyzer should be provided during the liquefaction to accelerate the conversion from the normal-hydrogen, n-H_2 (25% para-hydrogen and 75% ortho-hydrogen), to para-hydrogen (p-H_2) [19].

E. *Mixed refrigerant pre-cooled cycle*

Mixed refrigerant (MR) is a mixture of hydrocarbon components (N_2, Methane, Ethane, Propane, Butane) used to precool hydrogen gas to a temperature of 75 K. Figure 5 shows the overall process diagram of a MR precooled system working according to Claude cycle.MR refrigeration is a mature technology for liquefied naturel gas (LNG) applications, but has not yet been realized commercially for hydrogen liquefaction, where open nitrogen pre-cooling is the standard [26].With MR precooling, high efficiency and minimum energy consumption can be reached.

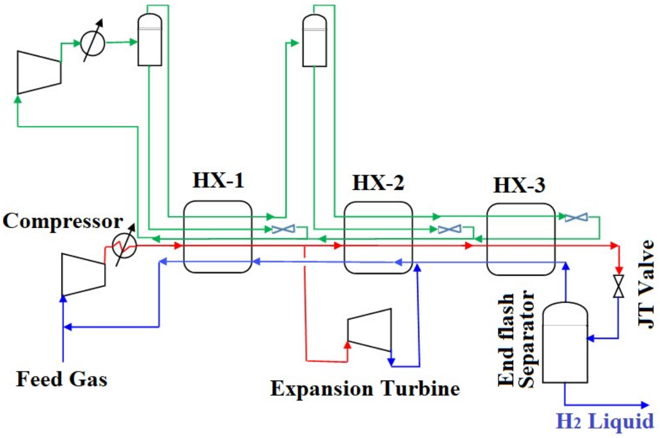

Fig. 5 Mixed refrigerant pre-cooling simple Claude cycle.

III. AMR LIQUEFACTION CYCLE

The concept of magnetic refrigeration is based on the Magnetocaloric Effect (MCE), which occurs in some materials when they are subjected to external magnetic field changes (Magnetization / Demagnetization process) through an AMR regenerator cycle. Its Application to hydrogen liquefaction consists in performing a thermal contact between the AMR regenerator cycle and the gas to be liquefied as illustrated in figure 6.Liquefaction is performed by absorbing an amount of heat from the gas (Q_L), equal to the total cooling power of the AMR liquefier, and rejecting into the atmospheric environment an amount of heat equal to Q_{Ha}.

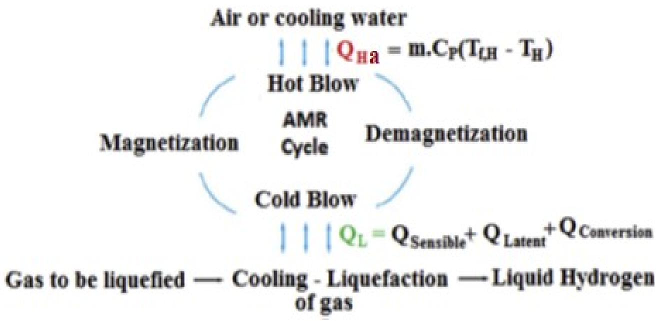

Fig. 6. Hydrogen liquefaction process via AMR cycle.

Furthermore, the use of one AMR cycle is not suitable to provide the large temperature span required for hydrogen liquefaction. Thus, applying magnetic refrigeration to the liquefaction of hydrogen is performed by assembling a number of AMR cycles operating in series (cascade cycles) as illustrated in figure 8. The cooling power produced by one stage is used to cool hydrogen entering the stage and absorbs heat rejected by the lower stage. The required cooling power of each stage is regulated by adjusting the volume of the regenerator beds (adjusting the volume of magnetic material used in each stage).

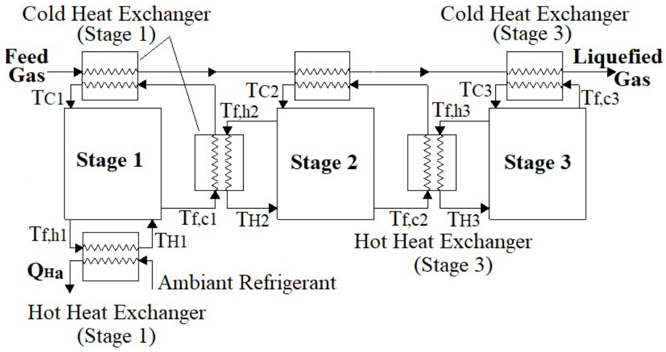

Fig. 8 Multistage AMR liquefaction system

In the present paper, the AMR cycle considered for each stage is mainly constituted, as illustrated in figure 7, of the following components [17]:

- Two Regenerator beds working alternately, AMR_A and AMR_B, (when the first regenerator bed is magnetized, the second one is demagnetized) to provide a continuous production of the cooling powerthrough the regenerator beds (Q_C).
- Reciprocating displacer which is able to move the fluid alternatively;
- Magnetic field source able to magnetize and demagnetize the regenerator beds;
- Hot heat exchanger able to reject the thermal energy (Q_H) into the hot reservoir;
- Cold heat exchanger able to receive the thermal energy from the cold reservoir (Cooling power Q_C).

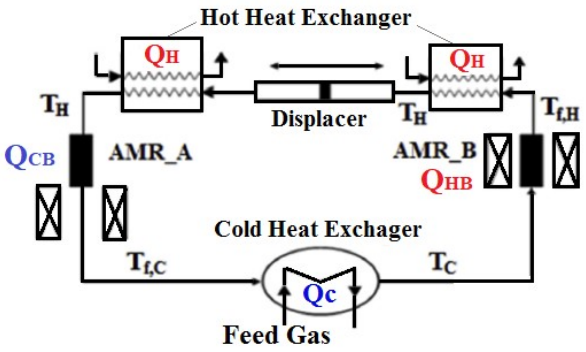

Fig.7. AMR device with two regenerator beds.

A complete AMR cycle consisting of two isentropic steps (adiabatic magnetization / demagnetization process) and two isofield steps (Cold and Hot blows) which can be described as follows) [20]:

1. Adiabatic demagnetization step:
   By reducing magnetic field from given strength B to 0 with no flow, the bed is demagnetized adiabatically.

2. Cold Blow at zero fields:
   The fluid is then forced by the displacer to move from the hot to the cold exchangers. Upon entering the demagnetized bed, the fluid temperature T_f is equal to the hot reservoir temperature T_H, exchanging heat with the bed it drops to $T_{f,C}$below the cold reservoir T_C at the cold end. Passing through the cold heat exchanger, the fluid absorbs heat from the cold reservoir (gas to be liquefied) at a rate Q_C, which represents the cooling power of the regenerator:

$$Q_C = \dot{m}_f Cp_f \left(T_C - T_{f,C}\right). \tag{1}$$

3. Adiabatic magnetization step:
   The bed is magnetized adiabatically when the magnetic field increases from 0 to B, with no flow.

4. Hot Blow at applied field:
   The fluid is then forced from the cold to the hot ends, entering the bed at the temperature equal to T_C. Since the bed temperature rises along the flow direction, the fluid temperature will also be increased, and it leaves the bed at a temperature $T_{f,H}$ higher than T_H. Passing through the hot exchanger, the fluid temperature drops to T_H, rejecting heat to the hot reservoir at a rate:

$$Q_H = \dot{m}_f Cp_f \left(T_{f,H} - T_H\right). \tag{2}$$

in addition to Q_H and Q_C given by equations 1 and 2, the exchanged heats with the regenerator beds, Q_{HB} and Q_{CB}, can be calculated according to the fluid entering temperatures in the regenerator beds T_H and T_C, respectively during the cold blow and the hot blow, and the exit temperatures $T_{f,C}$ and $T_{f,H}$.

$$Q_{CB} = \dot{m}_f Cp_f \left(T_H - T_{f,C}\right). \tag{3}$$

$$Q_{HB} = \dot{m}_f Cp_f \left(T_{f,H} - T_C\right). \tag{4}$$

Calculation of the exchanged heats by equations 1-4 involves development of a numerical model based on the fundamental equations of energy conservation considered for the fluid and the solid (magnetic material), during the cold and hot blows, and characterization of the MCE during the magnetization and demagnetization of material. For pure magnetic materials (Gd, Tb and Dy), the MCE characterization can be performed according to the Molecular Field Theory (MFT) [15, 17].

The time period of fluid flow for either blow is considered equal to τ. The adiabatic demagnetization and magnetization processes are assumed to occur instantaneously at the time τ and 2τ. This information shows the periodic operation of the AMR system; each period (2τ) constitutes a regenerative cycle where a temperature span is observed in the regenerator bed. This temperature span is amplified by alternating the carrier fluid between the two heat reservoirs, until reaching the steady state.

When the steady state is reached, the AMR cycle efficiency can be evaluated by using the coefficient of performance (COP), which is usually defined as follows:

$$COP_{AMR} = \frac{Q_C}{Q_H - Q_C + W}. \tag{5}$$

Where Q_H is the heat rejected into the hot reservoir, Q_C is the heat absorbed from the cold reservoir and W is the power input used to move the carrier fluids through the regenerator beds.

However, the overall AMR liquefier efficiency is calculated by using the overall COP as follow:

$$COP = \frac{Q_L}{Q_{Ha} - Q_L + W_{in}}. \tag{6}$$

Where Q_{Ha} is the heat rejected into the atmospheric environment at the hot heat exchanger of the first stage (stage

1), Q_L is the total heat absorbed from the gas to be liquefied (the total cooling power of the AMR liquefier) and W_{in} is the total power input used to move the carrier fluids through the regenerator beds.

IV. COMPARATIVE STUDY BETWEEN CLAUDE AND AMR HYDROGEN LIQUEFACTION CYCLES

To perform comparison between conventional and magnetic liquefaction systems, thermal analysis of the two systems has been performed by means of Aspen Hysys simulator which is a powerful software developed by AspenTech to simulate gas processing plants, oil refineries and petrochemical plants. The Simulation consists of building a Process Flow Diagram (PFD) of the liquefaction system, performing material and energy balances and calculating heat transfer and energy consumption.

A. *Claude cycle simulation*

The cycle considered in this work is based on the precooled Claude cycle where hydrogen gas is first precooled by a mechanical refrigerant until 245 K, then by nitrogen gas refrigerant until 83 K as illustrated in figure 4. The liquefaction part is carried out by cold hydrogen refrigerant after being expanded, from 50 bar to 3 bar, through two turbines. Nitrogen refrigerant loop includes two-stage compressor with intercooling, to compress nitrogen to 10 bar, and an Expander to expand it until 1.2 bar after being cooled in parallel with hydrogen in the precooling heat exchangers.

Two case studies have been performed. In the first case, hydrogen gas to be liquefied has been assumed to be supplied at liquefaction pressure. In the second case, hydrogen gas is supplied at atmospheric pressure and then compressed to the liquefaction pressure.

To calculate the cycle efficiency, we first need to determine the heat absorbed from the gas to be liquefied (Colling power Q_L) and compression consumedpower(W_C) by performing energy and material balances for the overall liquefaction cycle. This can be performed by Aspen Hysys and all operating parameters of the cycle will be determined from the simulation flow sheet.

The cycle efficiency is then computed by introducing the coefficient of performance (COP) which is defined as the ratio of heat absorbed from the gas to be liquefied (Q_L) to the net absorbed power of the cycle (Compression power W_C), as follows:

$$COP = \frac{Q_L}{W_C}. \tag{7}$$

where, Q_L is the total heat absorbed from hydrogen and W_C is the total compression power consumed.

B. *AMR cycle simulation*

Thermal analysis of the AMR cycle has been performed by Aspen Hysys simulator according to the simulation method published by Belkadi and Smaili [17]. The method consists of modeling the AMR system, once the steady state is reached, by an ordinary system where the magnetized regenerator is idealized as a heater, whereas the demagnetized one is idealized as a cooler (figure 9). The carrier fluid flows continuously through the cycle, absorbing form the heater an amount of heat equal to the heat absorbed from the regenerator bed during the hot blow (Q_{HB}). The carrier fluid rejects through the cooler an amount of heat equal to the heat exchanged with the regenerator bed during the cold blow (Q_{CB}).

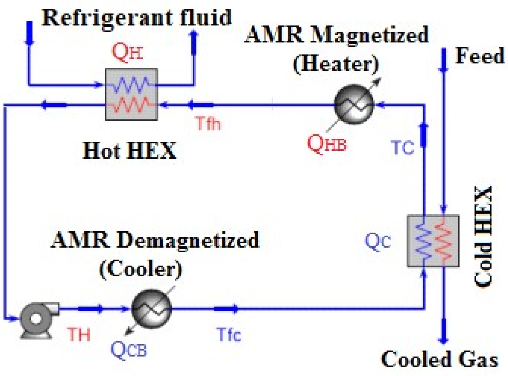

Fig. 9 AMR cycle Simulation method

The exchanged heats with the regenerator beds, Q_{HB} and Q_{CB}, should be calculated, according the equations 3 and 4, by the numerical model developed separately and introduced as input in Aspen Hysys simulation.

V. RESULTS AND DISCUSSION

As mentioned in section II, hydrogen can be supplied at the liquefaction pressure if it is produced by a hydrocarbon or methanol reforming. However, it should be compressed before purification and liquefaction if it is produced by an electrolyze device operating at atmospheric pressure [19]. In this logic, simulation of precooled Claude cycle has been performed, first, for a device where hydrogen is supplied at liquefaction pressure. Then a device where hydrogen is supplied at atmospheric pressure is considered. The simulation has been performed for different liquefaction pressures in order to determine the optimum pressure which should be considered as operating pressure. Figure 10 illustrates the evolution of the COP and power consumption (W_C) as function of liquefaction pressure (P_L) when hydrogen is supplied at liquefaction pressure. Results show the increase of the COP with the pressure. This rise becomes insignificant from the pressure of 30 bar. Thus, the optimum pressure can be taken between 15 bar and 30 bar. At 20 bar the total power required to liquefy 1kg/h is 13kW. The corresponding COP is 0.105 (10.5%). This efficiency is not so far compared with published values. Pre-cooled Claude exergy efficiency, e.g., as published by Krasae-in [18], is situated between 6.2%-8.8% (noting that for the same system, exergy efficiency is higher compared to energy efficiency).

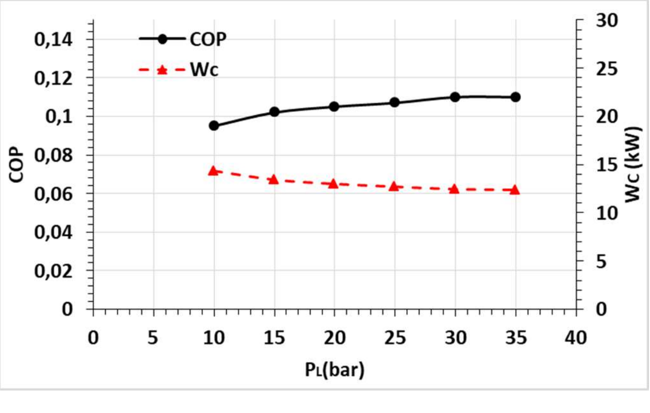

Fig.10. Evolution and COP and power required of precooled Claude cycle as function of liquefaction pressure when hydrogen is supplied at liquefaction pressure.

When hydrogen is supplied at atmospheric pressure, it should be compressed to the required liquefaction pressure. in this case the power required to compress 1kg/h from 1 bar to 20 bars is 1.5 kW. The total power required rises to 14.5 kW. Evolution of the COP of precooled Claude system and the total power required to liquefy 1kg/h of hydrogen supplied at 1 atm, as function of liquefaction pressure (P_L), is illustrated on figure 11.

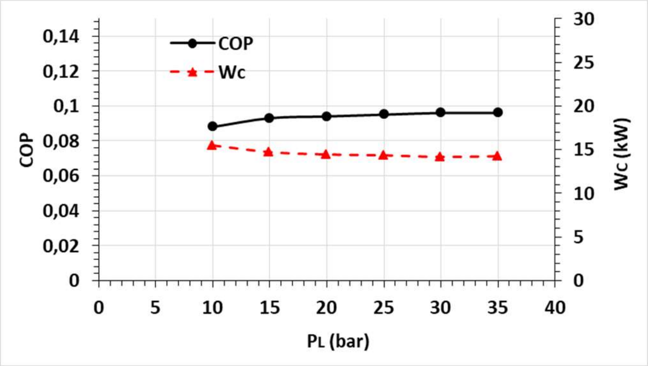

Fig. 11 Evolution of COP and power consumption of precooled Claude cycle as function of liquefaction pressure when hydrogen is supplied at atmospheric pressure.

For magnetic liquefaction, the AMR considered is composed of 6 stages operating in cascade. The hydrogen gas to be liquefied is supplied at ambient temperature under atmospheric pressure. Using Gadolinium (Gd), Terbium (Tb), Dysprosium (Dy) and Holmium (Ho) [11] as magnetic materials, thermal analysis has been performed by use of Hysys simulator as presented in section 3.2. The total volume (V) of the magnetic materials to be installed, liquefaction COP and power consumption (W_{in}) are calculated. The relation between the COP and the total volume of magnetic materials has been investigated. Results obtained are presented in figure 12. Results show that the most efficient volume is 124 liters. The corresponding COP is 0.096 (9.6%). Here, the total work input used to move the carrier fluid through the regenerator beds is insignificant (0.053 kW to liquefy 1 kg/h of H_2 supplied at atmospheric pressure).

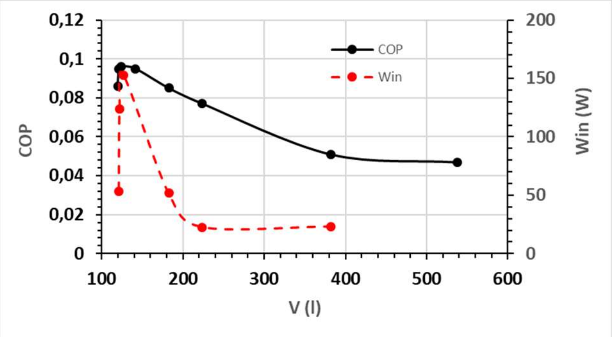

Fig.12. COP and the total work input of AMR cycle as function of magnetic material volume

To highlight the difference between the two systems, efficiency and energy consumption, calculated according to the thermal analysis procedure presented in section 3, are illustrated in table 1.

Table I
Comparison between conventional system and AMR system.

	P_L (bar)	COP	W_C (kW)	W_{in} (kW)
Claude cycle	20	0.105	13	
	20	0.094	14.5	
AMR system	1	0.096		0.053

It is found that, in terms of efficiency, the two systems have the same COP. However, the power consumption for conventional systems is very high compared to the AMR systems. These performances can be improved for the AMR liquefier by the use of other magnetic materials having large EMC.

VI. CONCLUSION

In this study, an AMR liquefaction system, constituted of 6 stages operating in cascade, has been considered to be compared with a precooled Claude system. COP and energy consumption have been calculated for the two systems at similar conditions.

In conventional systems, liquefaction can be obtained by the combined effect of cooling and adiabatic expansion of gas after it has been previously compressed. This liquefaction requires the use of large amounts of energy through cooling loops and at the recompression of gas. 13 kW is the required power found to liquefy 1kg/h supplied at 20 bar. The corresponding COP found is 0.105. When hydrogen is supplied at atmospheric pressure and compressed to be liquefied at 20 bar, the power required rises to 14.5 kW and the COP decreases to 0.094.

By use of real magnetic materials (Gadolinium Gd, Terbium Tb, Dysprosium Dy and Holmium HoN) in 6 stages AMR liquefier, the COP found is 0.096. This value is very close of the one found for precooled Claude cycle. However, the total work input used to move the carrier fluid through the regenerator beds is insignificant compared to the energy consumed in Claude system: 0.053 kW of power to liquefy 1 kg/ of H_2 supplied at atmospheric pressure against 14.5 kw for Claude system. These results can be enhanced by the use of other magnetic materials having large EMC. However, the use of a large amount of magnetic materials as solid refrigerant stills the main disadvantage of the magnetic systems: 124 litters is the volume of magnetic materials found for the AMR to be considered.

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REFERENCES

[1] J. Andersson, S. Grönkvist, Large-scale storage of hydrogen, International Journal of Hydrogen Energy 44 (2019) 11901-11919. https://doi.org/10.1016/j.ijhydene.2019.03.063
[2] M. Aasadnia, M. Mehrpooya, Large-scale liquid hydrogen production methods and approaches: A review, Applied energy 212 (2018) 57-83. https://doi.org/10.1016/j.apenergy.2017.12.033
[3] P. Malbrunot, Th. Alleau, Mémonto de l'hydrogène, Fiche 4.3 (2019).
[4] JA. Barclay, The theory of an active magnetic regenerative refrigerator, New Mexico: Los Alamos National Laboratory; 1982. LA-UR-83e1251; CONF-821237-1.
[5] A. Smaili, R. Chahine, Thermodynamic investigations of optimum active magnetic regenerators. Cryogenics 38(1998) 247-252.
[6] C. Aprea, A. Maiorino, a flexible numerical model to study an active magnetic refrigerator for near room temperature applications, Appl. Energy 87(2010) 2690-2698.
[7] Y. Chiba, A. Smaili, C. Mahmed, M. Balli, O. Sari, Thermal investigations of an experimental active magnetic regenerative refrigerator operating near room temperature, International Journal of Refrigeration 37(2014) 36-42.
[8] Jr. K.A. Gschneidner, V.K. Pecharsky, Thirty years of near room temperature magnetic cooling: where we are today and future prospects, International Journal of Refrigeration 31(2008) 945-961.

[9] L. Zhang, S.A. Sherif, T.N. Veziroglu, J.W. Sheffield, Performance analysis of reciprocating magnetic liquefiers, Int. Journal of Hydrogen Energy 19(1994) 945-956.

[10] L. Zhang, S.A. Sherif, T.N. Veziroglu, J.W. Sheffield, On exergy losses in AMR hydrogen liquefier, Int. Journal of Hydrogen Energy, 19(1994) 447-452.

[11] L. Zhang, S.A. Sherif, A.J. DeGregoria, C.B Zimm, T.N. Veziroglu, Design optimization of a 0.1- ton/day active magnetic regenerative hydrogen liquefier, Cryogenics 40 (2000) 269-278.

[12] W. Iwasaki, Magnetic refrigeration technology for an international clean energy network using hydrogen energy (WE-NET), Int. Journal Hydrogen Energy 28(2003) 559-567.

[13] T. Nakagawa, T.A. Yamamoto, T. Numazawa, Research on a magnetic refrigeration cycle for hydrogen liquefaction, Cryocoolers 14 (2007) 645-653.

[14] K. Matsumoto, T. Kondo, M. Ikeda, T. Numazawa, Numerical analysis of active magnetic regenerators for hydrogen magnetic refrigeration between 20 and 77K, Cryogenics 51 (2011) 353-357.

[15] A. Smaili, S. Aı¨t Ali, R. Chahine, Performance predictions of a first stage magnetic hydrogen liquefier, Int. Journal of Hydrogen Energy 36(2011) 4169-4177.

[16] I. Park, Y. Kim, J. Park, S. Jeong, Design method of the layered active magnetic regenerator (AMR) for hydrogen liquefaction by numerical simulation, Cryogenics 70 (2015) 57-64 https://doi.org/10.1016/j.cryogenics.2015.04.007

[17] M. Belkadi, A. Smaili, Thermal analysis of a multistage active magnetic regenerator cycle for hydrogen liquefaction, International Journal of Hydrogen Energy 43(2018) 3499-3511.

[18] S. Krasae-in, J. Stang, P. Neksa, Development of large-scale hydrogen liquefaction processes from 1898 to 2009, International Journal of Hydrogen Energy 35(2010) 4524-4533.

[19] J. Gallarda, Liquéfaction de l'hydrogène. Technique de l'Ingénieur, j3603 (2001).

[20] S. Krasae-in, J. Stang, P. Neksa, Exergy analysis on the simulation of a small-scale hydrogen liquefaction test rig with a multi-component refrigerant refrigeration system, Int Journal of Hydrogen Energy 35 (2010) 8030-8042.

[21] S. Krasae-in, A. Bredesen, J. Stang, P. Neksa, Simulation and experiment of a hydrogen liquefaction test rig using a multi-component refrigerant refrigeration system, Int. Journal of Hydrogen Energy 36 (2011) 907-919.

[22] S. Krasae-in, J. Stang, P. Neksa, Simulation on a proposed large-scale liquid hydrogen plant using a multi-component refrigerant refrigeration system, Int. Journal of Hydrogen Energy 35(2010) 12531-12544.

[23] D. O. Berstad, J.H. Stang, P. Neksa, Large-scale hydrogen liquefier utilizing mixed-refrigerant pre-cooling, International Journal of Hydrogen Energy 35(2010) 4512-4523.

[24] M. Asadnia, M. Mehrpooya, A novel hydrogen liquefaction process configuration with combined mixed refrigerant systems, International Journal of Hydrogen Energy 42(2017)15564-15585. https://doi.org/10.1016/j.ijhydene.2017.04.260

[25] M.A. Gadalla, T.A.H. Ratlamwala, I. Dincer, M. Kanoglu, Performance assessment of an integrated absorption cooling-hydrogen liquefaction system using geothermal energy, Int. J. Exergy 12(213) 205-225.

[26] G. Skaugen, D. Berstad, 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. Journal of Hydrogen Energy 45(2020) 6663-6679. https://doi.org/10.1016/j.ijhydene.2019.12.076

[27] R. Hande, Ø. Wilhelmsen, Minimum entropy generation in a heat exchanger in the cryogenic part of the hydrogen liquefaction process: On the validity of equipartition and disappearance of the highway, Int. Journal of Hydrogen Energy 44(2019) 15045-15055. https://doi.org/10.1016/j.ijhydene.2019.03.229

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