

Large-Scale H₂ Liquefaction: Ortho-Para Conversion in a Conceptual Brayton Cycle

Process Modeling, Viability, and Techno-Economic
Analysis

Energy, Flow, & Process Technology MSc Thesis
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Cover:	Personally taken from Air Products H ₂ liquefaction plant in Botlek, Rotterdam, The Netherlands.
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Abstract

The economic viability of a hydrogen economy revolves around efficient and scalable methods for its transportation and storage. Liquefaction presents a key pathway by increasing hydrogen's volumetric energy density, though current industrial plants are hampered by high energy consumption and costs. While conceptual models offer pathways to higher efficiency, they often lack the detailed technical and economic analysis needed to validate their feasibility, particularly for systems based on the Brayton cycle.

This thesis presents a comprehensive framework for the process modeling, viability assessment, and techno-economic analysis of a large-scale hydrogen liquefaction plant based on a conceptual Brayton cycle. A central contribution is the detailed modeling of the critical ortho-para hydrogen conversion, which was simulated within a catalytic Plate-Fin Heat Exchanger (PFHX) using a specialized Python-based model integrated with a full-plant Aspen HYSYS simulation. The study also addresses the effective management of excess cold hydrogen gas; an initial investigation into using an ejector for recirculation was conducted, but this approach was ultimately discarded as it did not yield economic improvements. A final, optimized hybrid Brayton-Claude cycle featuring an efficient cold gas recirculation loop was developed, enabling a plant capacity of 86 tonnes per day (TPD).

The techno-economic analysis of the final design was performed using the Aspen Process Economic Analyzer (APEA). In the baseline scenario, assuming an electricity price of 0.1 €/kWh , the plant achieved a Specific Liquefaction Cost (SLC) of $1.51\text{ €/kg}_{\text{LH}_2}$ and a Specific Energy Consumption (SEC) of $6.983\text{ kWh/kg}_{\text{LH}_2}$ in the baseline scenario.

Moreover, sensitivity analyses show a further reduction in SLC and SEC in the scenario using power recovery from the turbines: $1.49\text{ €/kg}_{\text{LH}_2}$ and $6.723\text{ kWh/kg}_{\text{LH}_2}$ respectively. Additionally, electricity price is the dominant factor influencing plant economics, with the long-term cost target of below $1.00\text{ €/kg}_{\text{LH}_2}$ being achievable at an electricity price of 0.035 €/kWh . Future projections, which account for reduced design allowances as the technology matures to a "Proven Process," suggest a potential SLC reduction of 4.64%.

In summary, this thesis establishes a robust techno-economic framework for Brayton-cycle based liquefaction, demonstrating its viability while highlighting the critical interplay between advanced process modeling, component efficiency, and energy costs in achieving a competitive large-scale liquid hydrogen supply chain.

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Nomenclature

Symbols

Roman	Description	Unit
A	Area	m^2
a	Constant in van der Waals EOS	$\text{Pa}\cdot\text{m}^6/\text{kg}^2$
b	Constant in van der Waals EOS	m^3/kg
c_p	Specific heat at constant pressure	$\text{J}/\text{kg}\cdot\text{K}$
C	Capital cost	€
C_H	Hydrogen concentration	mol/m^3
d_h	Hydraulic diameter	m
E_a	Activation energy	J/mol
H	Enthalpy	J/mol
h	Specific enthalpy	J/kg
k	Kinetic rate constant	$1/\text{s}$
k_w	Reaction constant	$\text{mol}/\text{m}^3\cdot\text{s}$
$\dot{m}$	Mass flow rate	kg/s
M	Molecular mass	g/mol
N	Number of molecules	mol
p	Pressure	Pa
p_h	Hydraulic perimeter	m
$\dot{Q}$	Heat transfer rate	W
R	Universal gas constant	$\text{J}/\text{mol}\cdot\text{K}$
$\dot{r}$	Conversion rate	$\text{m}^3/\text{mol}\cdot\text{s}$
S	Plant or equipment capacity	–
T	Temperature	K
t	Time	s
t_w	Wall thickness	m
U	Heat transfer coefficient	$\text{W}/\text{m}^2\cdot\text{K}$
V	Volume	m^3
v	Specific volume	m^3/kg
w	Specific work	J
$\dot{W}$	Power	W
x	Mole fraction	–
y	Liquid yield	–
z	Spatial coordinate	–

Greek

Symbol	Description	Unit
λ	Thermal conductivity	$\text{W}/\text{m}\cdot\text{K}$
∂	Partial Derivative	–
ε	Volume fraction	–
η	Efficiency	–
μ_{JT}	Joule-Thomson coefficient	K/Pa
μ	Viscosity	$\text{Pa}\cdot\text{s}$
ρ	Density	kg/m^3

Subscripts and Superscripts

Symbol	Description
0	Reference condition or initial state
1, 2...	State point or stage in the process
a	Annual
c	Critical point
cat	Catalyst
comp	Compressor
conv	conversion
eff	Effective coefficient
eq	Equilibrium
f	Fluid
h	Hot stream
i	Ideal
in	Inlet
inv	Inversion
is	Isentropic
LH ₂	Liquid Hydrogen
m	Mean logarithmic
max	Maximum
min	Minimum
mol	Molar-based
n	Economical value indicator
o	Ortho-hydrogen
out	Outlet
p	Para-hydrogen
trans	Transfer
turb	Turbine
turb	Turbine
w	Wall

Abbreviations

Short	Description
APEA	Aspen Process Economic Analyzer
CAPEX	Capital Expenditure
COG	Coke Oven Gas
CO ₂	Carbon Dioxide
DMR	Dual Mixed Refrigerant
DRI	Direct Reduced Iron
EOS	Equation of State
FeO _s	Framework for thermodynamic equations of state
FOM	Figure of Merit (Exergy Efficiency)
He	Helium
HP	High Pressure
HX	Heat Exchanger
H ₂	Hydrogen
IEA	International Energy Agency
IRENA	International Renewable Energy Agency
ISBL	Inside Battery Limits Investments
JT	Joule-Thomson
KPI	Key Performance Indicator
LOX	Liquid Oxygen

LHHW	Langmuir-Hinshelwood-Hougen-Watson
LH ₂	Liquid Hydrogen
LNG	Liquefied Natural Gas
LN ₂	Liquid Nitrogen
LP	Low Pressure
LR	Learning Rate
MR	Mixed Refrigerant
Ne	Neon
ODE	Ordinary Differential Equation
o-H ₂	Ortho-Hydrogen
OM	Operations and Maintenance
ORC	Organic Rankine Cycle
OSBL	Outside Battery Limits Investments
p-H ₂	Para-Hydrogen
PFHX	Plate-Fin Heat Exchanger
PR	Progress Rate
PSA	Pressure Swing Adsorption
Re	Reynolds Number
RPM	Rotations Per Minute
SAF	Sustainable Aviation Fuel
SEC	Specific Energy Consumption
SLC	Specific Liquefaction Cost
SLS	Space Launch System
TPD	Tonnes per Day
TU Delft	Delft University of Technology

1

Introduction

1.1. Overview of Hydrogen as an Energy Carrier

Hydrogen, often called the 'fuel of the future', is a simple molecule with enormous potential to power a wide variety of applications. According to the International Energy Agency (IEA), its abundance in the universe and its ability to generate carbon-free energy make it an ideal candidate for a sustainable energy future [1]. Although its presence in industry is well-established, as a feedstock in refining and chemical processes, its role in the clean energy transition is still being developed. However, the path to a hydrogen economy is far from straightforward, with significant hurdles to overcome, especially in its transportation and storage [1, 2].

Despite being the most abundant element in the universe, its low volumetric energy density poses significant logistical challenges. In its gaseous state, hydrogen occupies a significant volume, making it difficult and expensive to transport and store over large distances [3]. This is one reason why hydrogen is still relegated to niche uses despite its potential. For example, gaseous hydrogen requires significant compression to be stored efficiently. Transportation and storage alternatives present several challenges. Metal hydrides, while promising for some applications, face challenges related to weight, charging and discharging speed, and storage capacity [3]. Gaseous transport through pipelines is suitable for short distances, but becomes expensive and problematic for long ones, due to the low density of hydrogen and the risk of leaks [4]. Transport as ammonia or methanol, hydrogen derivatives that are easier to transport, requires additional conversion processes to recover pure hydrogen, resulting in efficiency losses and additional costs [5].

Furthermore, hydrogen molecules containing protium, which are those used in industrial applications, have a high specific evaporation mass but a lower volumetric heat of evaporation due to their low density [6]. This means that storing and transporting hydrogen in its gaseous form requires significant energy input and infrastructure, reinforcing the importance of liquefaction for large-scale applications.

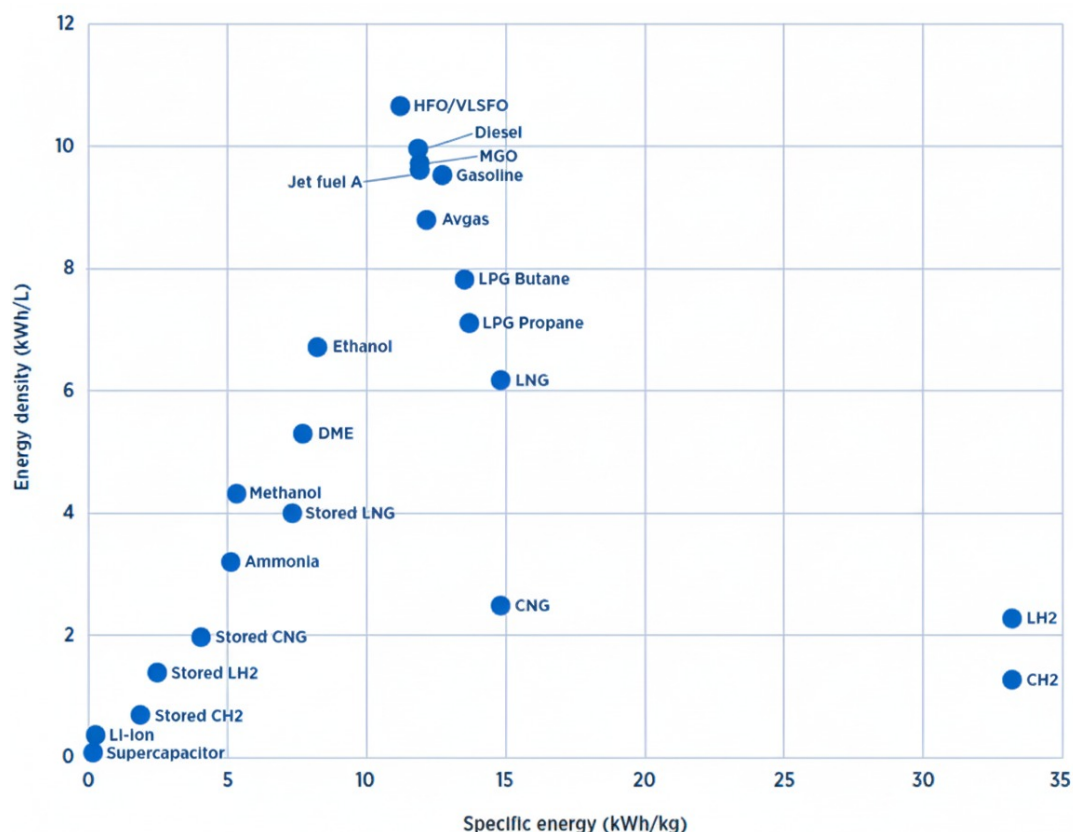


Figure 1.1: Energy density and specific energy of various fuels and energy storage systems

Liquefaction plays a critical role in the hydrogen supply chain by enabling large-scale storage and long-distance transportation, which are essential for the global adoption of hydrogen as an energy carrier. Given the low volumetric energy density of hydrogen in gaseous form, Figure 1.1 shows how hydrogen in liquid form has a significant volume requirement, allowing for more efficient and cost-effective transport through cryogenic tankers or dedicated infrastructure such as insulated pipelines, storage tanks, and loading terminals designed to handle cryogenic conditions [3]. This is particularly important for applications where pipeline transport is not feasible, such as international shipping and decentralized distribution networks.

In addition, liquid hydrogen (LH₂) offers advantages in high-purity applications, such as fuel cells and aerospace industries, where stringent quality requirements must be met. However, the energy-intensive nature of hydrogen liquefaction remains a challenge, necessitating ongoing research into improving process efficiency and reducing operational costs. Innovations such as mixed-refrigerant pre-cooling and advanced liquefaction cycles, including the Claude and Brayton cycles, are being explored to optimize energy consumption and enhance the economic viability of LH₂ production [7]. By overcoming these challenges, liquefaction can facilitate the widespread deployment of hydrogen, supporting its integration into various sectors, from mobility to industrial energy applications, and contributing to a sustainable energy future.

Applications of Liquid Hydrogen

Furthermore, liquid hydrogen is currently seen in a wide variety of industry applications such as the aviation sector, the steel industry, the maritime industry, and the freight road transport sector.

Aviation: Liquid hydrogen is increasingly being explored as a sustainable fuel for aviation (SAF) due to its high energy density and zero emission potential when used in fuel cells or combustion engines. Major aerospace companies and research institutions are actively developing hydrogen powered aircraft concepts, with Airbus, for example, aiming to introduce hydrogen fueled commercial aircraft by

2035 known as ZeroE [8]; a concept which reduces emissions by 50% and uses LH_2 as its hydrogen storage and energy carrier, a preferred approach over storing compressed hydrogen, since liquefaction allows for much higher volumetric density and lighter, more compact tanks suitable for aircraft design. However, the integration of cryogenic storage systems presents significant design challenges, as LH_2 tanks must be stored in the fuselage due to their size and insulation requirements, deviating from traditional wing-integrated fuel tanks and affecting aircraft aerodynamics, structural layout, and payload capacity.

In addition, airport infrastructure development and aircraft re-design to accommodate larger fuel tanks are still being addressed. Advances in hydrogen propulsion technologies, including direct combustion in modified gas turbines and hybrid fuel-cell power systems, are expected to drive the feasibility of LH_2 in aviation over the next decade [5, 8]. Furthermore, LH_2 has been a cornerstone of space exploration for decades, serving as a high-energy propellant in combination with liquid oxygen (LOX) in cryogenic rocket engines. Its exceptionally high specific impulse makes it ideal for deep-space missions and heavy-lift launch vehicles. The Space Launch System (SLS) from NASA, the Saturn V, and the Space Shuttle's main engines all relied on LH_2 due to its superior performance [9]. Once again, the cryogenic challenges are present, as well as boil-off losses. Despite these issues, LH_2 remains a key fuel choice for next-generation rockets, including those being designed for lunar and Mars exploration as per IRENA [5].

Steel: The steel industry is another key sector where LH_2 is gaining attention as a cleaner alternative to coal-based production methods. Traditionally, steel manufacturing relies on carbon-intensive processes such as blast furnaces that use coke as a reducing agent. The integration of hydrogen into direct reduced iron (DRI) processes offers a pathway to significantly reduce CO_2 emissions. Liquid hydrogen is particularly advantageous in locations where pipeline hydrogen infrastructure is unavailable, as it allows efficient transport and storage to supply steel plants. Countries such as Sweden and Germany are pioneering hydrogen-based steel making projects, with the aim of large-scale decarbonization of one of the most emission-intensive industries [5].

Maritime: There are now several hydrogen ships in development or in service. The first liquid hydrogen powered ferry in the world, the MF Hydra, which is equipped with hydrogen fuel cell technology, is an innovative example [10]. FPS Waal and Zulu06, two modified cargo ships that operate on hydrogen fuel cells around European rivers, are further examples [11, 12]. These vessels are setting the stage for wider use while showcasing the potential of hydrogen in transportation. The use of hydrogen in long-distance maritime transportation is growing. Because of its versatility, hydrogen fuel technology enables ships to go farther without emitting any pollution than those that run on batteries. The long-term advantages of hydrogen use are expected to make it a competitive alternative to conventional marine fuels, despite the fact that initial prices are still considerable [13]. Hydrogen will be essential to achieve the strict decarbonization goals as these advancements progress.

Freight road: LH_2 has the potential to revolutionize the freight road sector by providing a low-emission fuel alternative to high-energy diesel [14]. Unlike compressed hydrogen, which requires heavy and bulky storage tanks, LH_2 offers a more compact solution, enabling longer driving ranges and higher payload capacities. Its cryogenic nature (-253°C) presents challenges in storage and refueling, but advances in insulation technology and refueling infrastructure are gradually addressing these challenges. Major truck manufacturers, such as Nikola, Hyzon, and DAF, and logistics companies are investing in LH_2 -powered fuel cell electric trucks [15–17], leveraging its fast refueling time and ability to sustain heavy-duty transport operations with minimal downtime. Additionally, integrating LH_2 into freight logistics can significantly reduce greenhouse gas emissions, contributing to decarbonization efforts while maintaining the efficiency and reliability required for long-distance freight. However, the widespread adoption of LH_2 in the freight sector will depend on cost reductions in hydrogen production, advancements in liquefaction technology, and the development of a robust refueling network.

1.2. Production of Liquid Hydrogen, Current Trends

Most existing hydrogen liquefaction plants have capacities below 20 tons per day (TPD), with the largest reaching 32 TPD [18]. Their specific energy consumption (SEC) ranges between 10 and 20 $\text{kWh/kg}_{\text{LH}_2}$,

contributing significantly to the specific liquefaction cost (SLC) of 2.5–3 US\$/kg_{LH₂} [18]. Theoretically, the minimum energy required to liquefy hydrogen, also known as ideal SEC, from ambient conditions (300 K, 1.01 bar) is 3.9 kWh/kg_{LH₂}, including the ortho-para hydrogen conversion, suggesting room for optimization. This can be seen in the dotted line in Figure 1.2, which shows current SEC for several industrial and conceptual hydrogen liquefiers. Studies indicate that costs decrease with increasing plant capacity and estimates predicting that liquefaction costs could drop below 1 US\$/kg_{LH₂}. The Asia Pacific Energy Research Centre (APEREC) estimated that an 800 TPD liquefaction plant could achieve costs as low as 0.53–0.78 US\$/kg_{LH₂} [19]. Beyond cost reductions, the demand for larger plants is expected to grow along with the expansion of the global hydrogen market. The existing and planned hydrogen liquefaction capacity in the United States total approximately 794 TPD as of May 2024 [20]. Considering 304 TPD from 14 plants currently in operation, with capacities of 5–32 TPD, and an additional 490 TPD under construction development. For comparison, the US, in 2022, registered 241,369.86 TPD of liquefaction capacity for natural gas [21].

Several large-scale conceptual hydrogen liquefaction designs have been proposed to enhance efficiency and reduce SEC, as shown in Figure 1.2. These studies suggest that SEC levels between 6 and 8 kWh/kg_{LH₂} are feasible for large-scale plants without major technological breakthroughs [18]. However, capital and operating expenses remain crucial considerations in estimating the overall liquefaction costs and evaluating the scalability of plant components. To make large-scale hydrogen liquefaction viable, practical and cost-effective approaches must be prioritized.

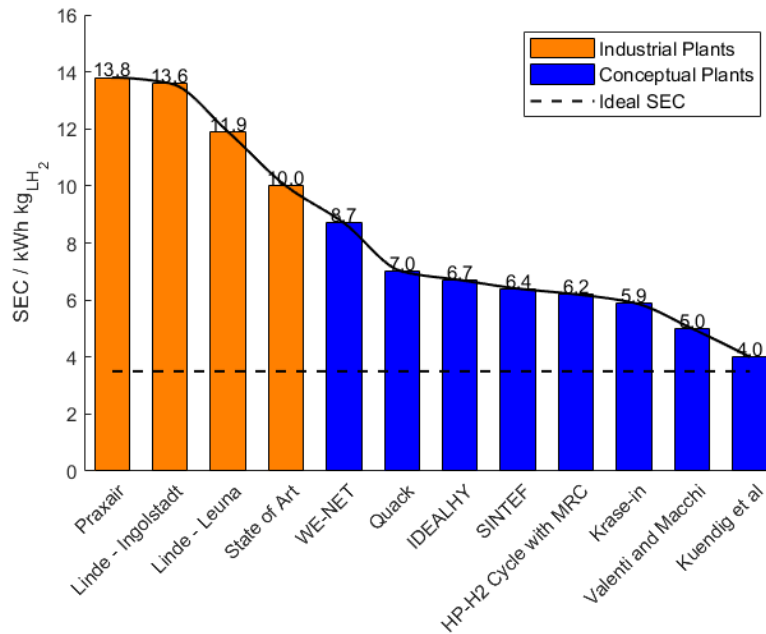


Figure 1.2: Efficiency performance of industrial and conceptual hydrogen liquefiers

This thesis focuses on hydrogen liquefaction using the Brayton cycle with an emphasis on ortho-para hydrogen conversion. By addressing key inefficiencies in existing systems, the aim is to develop a more cost-effective and energy-efficient liquefaction process. In order to achieve this, the ortho-para conversion will be modeled using an existing code by O'Neill et al. [22] to simulate the conversion occurring in the heat exchangers at cryogenic conditions, using the Helium-Neon (He–Ne) mixture as the coolant. This mixture is believed to reduce the SEC and SLC; however, there are currently few conceptual designs using this mixture, but there are many challenges such as inaccurate thermodynamic modeling due to the lack of data and precise equations of state (EOS), as well as the elevated price of He.

Next, an optimized version of the key equipment in the process flow diagram will ensure a more cost-effective simulation with the aim of reducing both capital and operating expenses; furthermore, using

a commercially available economic evaluation tool, a techno-economical analysis will be performed to have a numerical conclusion on the 2 key KPIs. In addition, sensitivity and scale-up analyzes will assess how operational parameters and increased capacity influence plant efficiency and economic feasibility. Lastly, the study projects potential learning and scaling trends for hydrogen liquefaction plants, based on cost data and forecasts from the relevant literature.

In summary, **this thesis aims to thoroughly evaluate the technical and economic feasibility of a conceptual large-scale hydrogen liquefaction process while establishing modeling and cost estimation frameworks that integrate recent technical recommendations.** As a result, the main research question of this thesis is the following:

How to optimize the process and economics of a large-scale hydrogen liquefaction plant based on a Brayton cycle?

1.3. Sub-Research Questions

To properly answer the main research question, three sub-research questions are formulated below:

1. How can a conceptual high-capacity Brayton cycle H_2 liquefaction plant be accurately modeled using a commercial process simulation tool, including ortho-para conversion reaction kinetics, while addressing equations of state limitations?
2. How can the excess H_2 gas be effectively managed and modeled in the liquefaction process?
3. How to effectively estimate the capital and operating costs of a conceptual hydrogen liquefier based on the process simulation model and preliminary equipment designs, and what are the key factors influencing the economic feasibility of the plant?

1.4. Research Plan

The literature research strategy for this thesis follows the *snowball method* and *citation searching* approach. The initial literature review is based on key publications that serve as the basis for further exploration.

1. Al Ghafri, S. Z. et al. Hydrogen liquefaction: a review of the fundamental physics, engineering practice and future opportunities, Energy & Environmental Science, 2022.
2. Keelan T. O'Neill et al. "Hydrogen ortho-para conversion: process sensitivities and optimisation". In: Chemical Engineering and Processing - Process Intensification 184 (Feb. 2023).
3. Cardella, U. F. Large-scale hydrogen liquefaction under the aspect of economic viability, Doktor-Ingenieurs, Universität München, 2018.
4. K Stolzenburg and R Mubbala. "Integrated Design for Demonstration of Efficient Liquefaction of Hydrogen (IDEALHY) Fuel Cells and Hydrogen Joint Undertaking (FCH JU) Title: Hydrogen Liquefaction Report". In: (2013).

2

Literature review

2.1. Principles of Hydrogen Liquefaction

As discussed in chapter 1, liquid hydrogen is a prospective alternative fuel in terms of its elevated specific energy, ease of transportation, storage, and better handling of safety issues. However, extensive research is needed before diving into the process modeling. This chapter serves as the backbone for the techno-economical decisions made later in the project; relying solely on arguments and assumptions found in the literature.

2.1.1. Thermodynamic Concepts for Liquefaction

Figure 2.1 shows a generic block diagram of an ideal liquefaction process. Only a synopsis of the essentials is given because hydrogen liquefaction techniques are covered in a number of recent publications [23–28].

The pre-compression stage is the initial stage of the process if the hydrogen supply is supplied at a relatively low pressure. The hydrogen then travels through an adsorption step to remove contaminants that can freeze during cryogenic liquefaction, as well as an optional pre-cooling stage to 80 K [29]. A closed-loop cryogenic refrigeration cycle is then used to cool the hydrogen to temperatures below 30 K [30].

Furthermore, ortho-hydrogen is catalytically converted to para-hydrogen in this cycle, either continuously or in batches [31]. Lastly, adiabatic expansion, which can be either turbine expansion or Joule-Thompson expansion, is usually the last stage. Liquid hydrogen is then extracted and stored at 20-21 K with an ideal para-hydrogen concentration of 99.8% [32].

Ideal Work Requirements for Gas Liquefaction: Similarly to any thermodynamic cycle, the constraints around a liquefaction system are determined by its ideal performance. An ideal liquefaction system operates through two theoretical processes derived from the Carnot cycle: a reversible isothermal compression, followed by a reversible isentropic expansion, as illustrated in Figure 2.1. By applying the principles of energy and entropy conservation to this cycle, the ideal specific work required for liquefying the gas can be determined by applying the second law of thermodynamics:

$$w_{\text{ideal}} = (h_1 - h_f) - T_o \cdot (s_1 - s_f). \quad (2.1)$$

With h_1 being the specific enthalpy of input H_2 , h_f being the specific enthalpy of liquid H_2 at 1 bar and 20 K, s_1 being the specific entropy of input H_2 , s_f being the specific entropy of liquid H_2 at 1 bar and 20 K, and T_o is ambient temperature (298.15K). As can be seen, w_{ideal} proves there is a minimum possible specific energy consumption (SEC). However, the actual SEC of liquefying a kg of hydrogen is determined by its energy consumption.

$$w = \frac{\dot{W}}{\dot{m}_f}. \quad (2.2)$$

Where w is the SEC, $\dot{W}$ is the liquefaction cycle net power consumption, and $\dot{m}$ is the liquefied hydrogen mass flow rate.

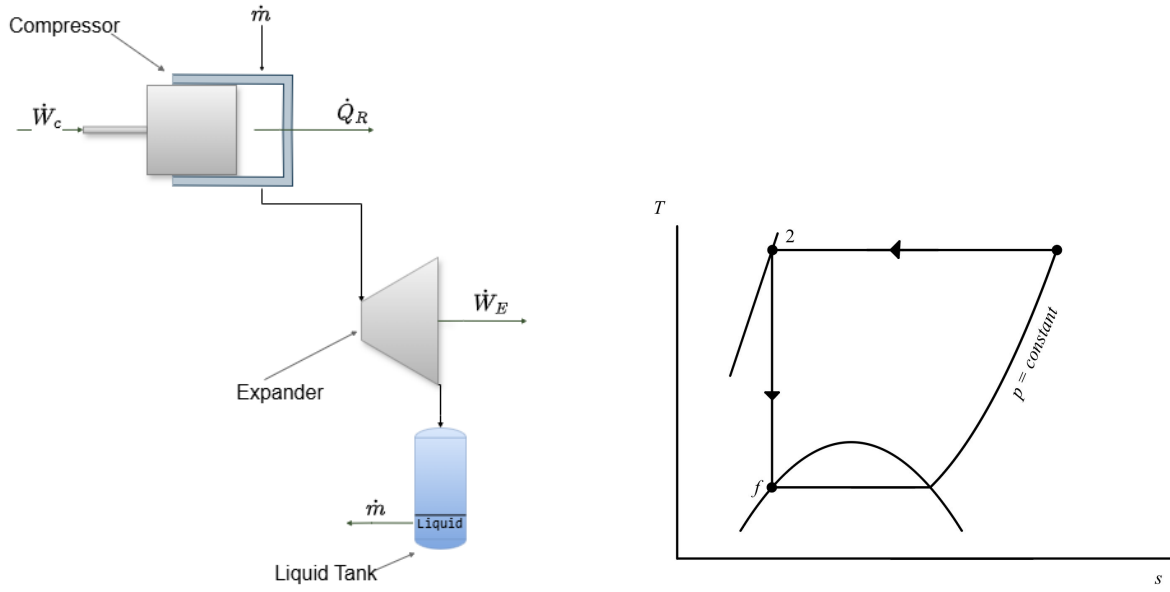


Figure 2.1: Schematic block diagram of an ideal liquefaction process and T-s thermodynamic diagram

The ideal work required for the liquefaction of various common gases, starting from an initial temperature of 300 K and a pressure of 1.013 bar, is summarized in Table 2.1, along with their boiling points and enthalpy of vaporization, respectively.

Table 2.1: Ideal-work requirements and enthalpy of vaporization for liquefaction of gases beginning at 300 K and 1.013 bar [33]

Gas	Normal Boiling Point (K)	Ideal Work of Liquefaction, $\frac{-W_i}{\dot{m}_f}$ (kJ kg ⁻¹)	Enthalpy of Vaporization (kJ kg ⁻¹)
³ He	3.19	8,178.0	20.7
⁴ He	4.21	6,819	20.9
H ₂	20.27	12,019.0	446.1
Ne	27.09	1,335.0	85.8
N ₂	77.36	768.1	199.0
Air	78.8	738.9	205.0
CO	81.6	768.6	213.0
Ar	87.28	478.6	162.0
O ₂	90.18	635.6	213.0
CH ₄	111.7	1,091.0	510.0
C ₂ H ₆	184.5	353.1	400.0
C ₃ H ₆	231.1	140.4	356.0
NH ₃	239.8	359.1	1,371.0

As indicated in the table, the work needed to liquefy hydrogen is notably higher compared to other frequently liquefied gases, including helium, despite helium having a lower boiling point than hydrogen. This suggests that the higher requirement for liquefaction work may stem from specific thermodynamic properties, such as enthalpy of vaporization, which could present additional challenges in the design of efficient hydrogen liquefaction systems [34].

Exergy efficiency, also known as figure-of-merit (FOM), is a numerical indicator of process performance,

comparing the actual specific energy consumption (SEC) with the theoretical minimum requirement.

$$\text{FOM} = \mu_{\text{ex}} = \frac{w_{\text{ideal}}}{w}. \quad (2.3)$$

An exergy efficiency value close to one indicates that the liquefier is operating close to its theoretical optimum. Henceforth, SEC and FOM are two well-used performance parameters commonly used in liquefaction systems.

Joule-Thomson Effect and Inversion Curves: The Joule-Thomson (J-T) effect describes the temperature change of a fluid when it passes through a restriction, such as an expansion valve or capillary tube, undergoing a pressure drop. This process is considered isenthalpic, meaning that there is no significant heat transfer with the surroundings and minimal changes in kinetic energy, with no work done on or by the fluid. The Joule-Thomson coefficient quantifies this temperature change with respect to pressure change at constant enthalpy. This can be seen in the equation below:

$$\mu_{\text{JT}} = \left(\frac{\partial T}{\partial p} \right)_h. \quad (2.4)$$

The value of μ_{JT} determines the outcome of an isenthalpic expansion:

- If $\mu_{\text{JT}} > 0$, the fluid cools during expansion.
- If $\mu_{\text{JT}} < 0$, the fluid heats up during expansion.
- If $\mu_{\text{JT}} = 0$, there is no temperature change during expansion.

For an ideal gas, the J-T coefficient is zero because there are no intermolecular forces; however, for real gases, the J-T coefficient can be negative, positive, or zero, depending on the initial temperature and pressure of the fluid. The J-T coefficient can be evaluated using an equation of state (EOS) for real gases, such as the van der Waals EOS. For example, the van der Waals equation gives the following expression for the J-T coefficient:

$$\mu_{\text{JT}} = \frac{(2a/RT) \cdot (1 - b/v)^2 - b}{c_p \cdot [1 - (2/vRT) \cdot (1 - b/v)^2]}. \quad (2.5)$$

In addition, the inversion curves represent the temperature and pressure points where the Joule-Thomson coefficient is zero. These curves separate the regions where expansion leads to cooling from those where expansion leads to heating. The inversion temperature, T_{inv} , is the temperature at which $\mu_{\text{JT}} = 0$ for a given pressure, and is given by the following expression for a van der Waals gas:

$$T_{\text{inv}} = \frac{2a \cdot (1 - b/v)^2}{bR}. \quad (2.6)$$

The maximum inversion temperature occurs when $b/v = 0$; therefore, $T_{\text{inv,max}} = 2a/bR$. Cooling by isenthalpic expansion is only possible when the initial temperature and pressure are inside the inversion curve, on the left side of the curve below. Figure 2.2 shows a T-p plot with isenthalpic lines; it is used to represent the inversion curve for different gases.

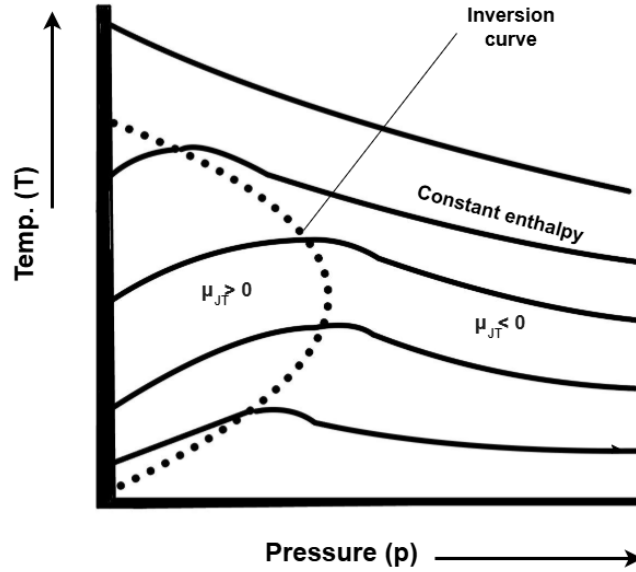


Figure 2.2: General pressure-temperature diagram of a real gas

Table 2.2: Maximum inversion temperature for several gases [35]

Gas	$T_{\text{inv,max}}$ [K]
Helium	45
Hydrogen	205
Neon	250
Carbon Monoxide	652
Nitrogen	621
Air	603
Oxygen	761
Argon	794
Methane	939

The maximum inversion temperature is different for different gases. Table 2.2 shows the maximum inversion temperature for various real gases. For gases with a maximum inversion temperature below ambient temperature, such as hydrogen, helium, and neon, the J-T expansion alone is insufficient to liquefy these gases. Therefore, pre-cooling or other processes are required to achieve liquefaction such as gas expansion via turbo-expanders, gas compressions, and heat transfer.

Gas Expansion: Gas expansion via turbo-expanders is a key process in hydrogen liquefaction, used to cool gases by converting their pressure energy into work and reducing their temperature. Turbo-expanders operate under isentropic conditions, ensuring high thermodynamic efficiency. As the gas expands and generates work, its temperature drops significantly, which aids in the cryogenic cooling process. As the isentropic efficiency needs to be taken into account, the real work output of the turbo-expander can be calculated using the following formula:

$$\dot{W}_{\text{turb}} = \eta_{\text{is}} \cdot \dot{m} \cdot (h_{\text{in}} - h_{\text{out,is}}). \quad (2.7)$$

Where the isentropic efficiency is defined as follows:

$$\eta_{\text{is}} = \frac{h_{\text{in}} - h_{\text{out}}}{h_{\text{in}} - h_{\text{out,is}}}. \quad (2.8)$$

Turbo-expanders are critical for achieving the low temperatures required in hydrogen liquefaction by efficiently extracting energy from the gas stream.

Gas Compression: Gas compression is a fundamental process in hydrogen liquefaction, where gas is pressurized to facilitate subsequent cooling and liquefaction stages. Compressors require work input to increase the gas pressure and the temperature of the gas rises as a result. The process often deviates from isentropic conditions due to irreversibility, so isentropic efficiency is used to evaluate the performance of the compressor. The actual work input to the compressor $\dot{W}_{\text{comp}}$ is related to the mass flow rate, specific enthalpies, and isentropic efficiency as follows:

$$\dot{W}_{\text{comp}} = \frac{\dot{m} \cdot (h_{\text{out}} - h_{\text{in}})}{\eta_{\text{is}}} \quad (2.9)$$

Where the compressors isentropic efficiency is expressed as follows:

$$\eta_{\text{is}} = \frac{h_{\text{in}} - h_{\text{out, is}}}{h_{\text{in}} - h_{\text{out}}} \quad (2.10)$$

Heat Transfer: Heat transfer is the process of thermal energy exchange between systems or substances due to a temperature gradient. In hydrogen liquefaction, heat exchangers facilitate the transfer of heat between hydrogen and the refrigerant. The heat transfer rate in such systems can be expressed as follows:

$$\dot{Q} = \dot{m} \cdot (h_{\text{in}} - h_{\text{out}}) \quad (2.11)$$

This equation quantifies the energy removed or added to the fluid as it flows through the heat exchanger, allowing precise control over the liquefaction process.

2.1.2. Ortho-Para Conversion: Thermodynamic Properties of Hydrogen Liquefaction and New Reaction Kinetics Experimental Validity

The unique thermochemical properties of H_2 significantly influence its liquefaction process. One critical factor is its low critical temperature (T_c) and critical pressure (p_c), which make liquefaction more challenging compared to other gases, such as nitrogen or methane. Accurate prediction of hydrogen thermodynamic properties under cryogenic conditions is essential for the design of efficient liquefaction systems. Several equations of state (EOS) are used to model hydrogen behavior, including cubic EOS like van der Waals or Peng-Robinson. However, cubic EOS are not accurate in predicting liquid molar volume [36]. Hence, more advanced models, such as the state-of-the-art EOS of Leachman et al.[37], provide precise predictions of the properties of liquid and gaseous hydrogen under cryogenic conditions.

Table 2.3: Critical-point and triple-point properties for normal hydrogen and parahydrogen used in the EOS by Leachman et al.[37].

	T (K)	p (MPa)	ρ (mol dm ⁻³)
Parahydrogen			
Critical point	32.938	1.284	15.556
Triple point	13.803	0.00700	38.215
Normal hydrogen			
Critical point	33.190	1.315	14.940
Triple point	13.952	0.00770	38.300

These models are indispensable for determining critical properties such as enthalpy, entropy, and the Joule-Thomson coefficient, which influence the performance of compressors, heat exchangers, and expanders in hydrogen liquefaction systems.

A unique event occurring in a plate-fin heat exchanger (PFHX) during cryogenic hydrogen liquefaction is known as ortho-para conversion. This is a critical aspect of hydrogen liquefaction as a result of its impact on thermal management and process efficiency. Hydrogen exists in two nuclear spin isomers: ortho-hydrogen (o- H_2), with parallel nuclear spins, and para-hydrogen (p- H_2), with antiparallel spins [28], Figure 2.3 shows a simplified visualization of the isomer spins.

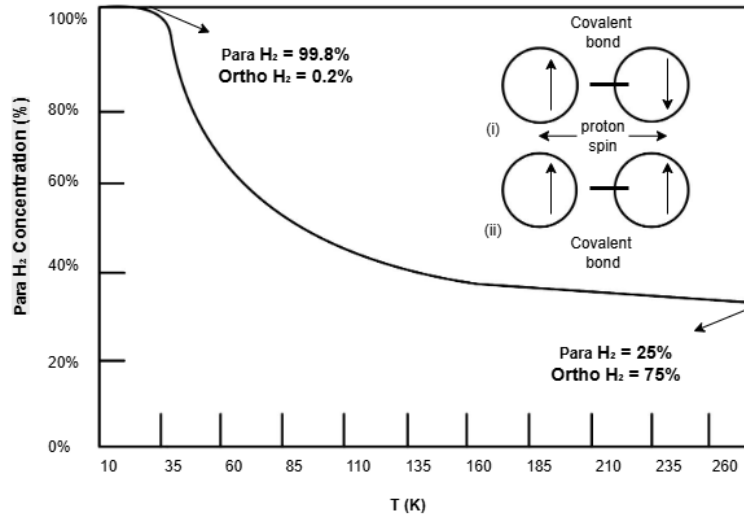


Figure 2.3: (i) para- and (ii) ortho-hydrogen spin isomers. (iii) para-Hydrogen content of hydrogen at equilibrium as a function of temperature.

Additionally, at room or higher temperatures, normal hydrogen (n-H₂) predominantly consists of 75% o-H₂ and 25% p-H₂. This is the commonly known pseudo substance that frequently provides all of the thermodynamic properties. However, at cryogenic temperatures near the boiling point of liquid hydrogen (20.38 K), the equilibrium shifts to roughly 99.8% para-hydrogen affecting overall hydrogen properties such as volumetric, optical, thermal, and magnetic [23].

Figure 2.4 shows the increasing proportion of p-H₂ as temperature decreases. This trend occurs because o-H₂ becomes thermodynamically unstable [30]. Furthermore, as the temperature lowers, the molecular energy of o-H₂ decreases. These factors drive the exothermic conversion of o-H₂ to p-H₂, a process that releases approximately 703.3 kJ kg⁻¹ of heat at the normal boiling point of hydrogen [30]. Notably, this heat of conversion is significantly higher than hydrogen's latent heat of vaporization, which is 449 kJ kg⁻¹ [37].

This exothermic reaction leads to the continuous *boil-off* of liquid hydrogen [30]. When the gaseous hydrogen with 75% ortho-hydrogen is liquefied, up to 50% stored hydrogen evaporates after 100 hours and 65% of stored hydrogen evaporates after 1000 hours [38], as seen in Figure 2.4. This is because the uncatalyzed process is a function of o-H₂ [39].

$$\frac{d[\text{o-H}_2]}{dt} = -k[\text{o-H}_2]^2. \quad (2.12)$$

Henceforth, to optimize the storage of LH₂ while reducing boil-off losses, a concentration predominantly made-up of p-H₂ is necessary in the storage tank for a successful liquefaction facility. Consequently, catalysts are implemented in the system as they aid in the acceleration of this process.

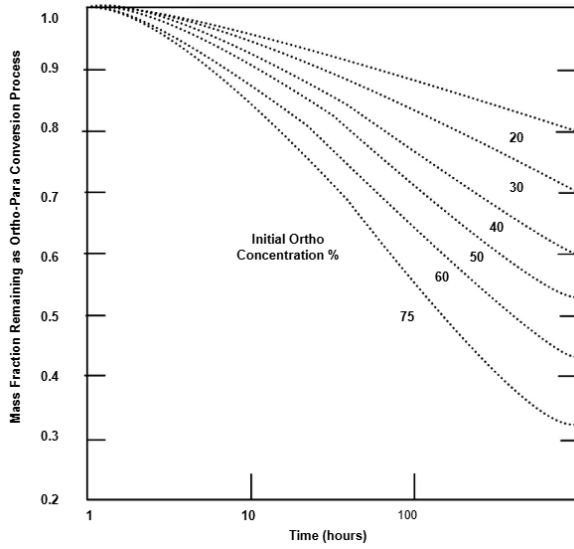


Figure 2.4: Storage Time (h)

Temperature [K]	Mole fraction of p-H ₂
20.27	0.9980
30	0.9702
40	0.8873
50	0.7796
60	0.6681
80	0.4988
100	0.3947
120	0.3296
160	0.2796
200	0.2597
300	0.2507

Table 2.4: Mole fraction of para-hydrogen (p-H₂) at different temperatures

2.1.3. Ortho-Para Conversion: Mathematical Modeling in a Catalytic Plate-Fin Heat Exchanger

Currently, commercial catalysts for hydrogen liquefaction are available as powdered or gelled paramagnetic ferric oxide, which is designed for use in packed fixed bed reactors [32]. Among these, hydrous ferric oxide is recognized as the most effective and widely used catalyst due to its high activity and low production cost. This makes it a preferred choice in the hydrogen liquefaction industry. However, solid catalysts are highly sensitive to impurities such as nitrogen, methane, carbon monoxide, and hydrogen sulfide, which can temporarily reduce activity or cause permanent damage. To mitigate this, impurities are typically removed in the early stages of the liquefaction process through adsorption, ensuring the reliability and efficiency of the catalyst [35]. Consequently, very little study has been dedicated to potential new types of conversion catalysts predicted since the late 1960s, largely due to the recognized advantages of hydrous ferric oxide [32]. When these catalysts are present, the kinetics are roughly first order, and complete conversion is accomplished in a matter of minutes [39].

O'Neill et al. Model

Researchers at the University of Western Australia developed a boundary value problem to properly model the kinetics of ortho-para conversion, heat transfer between hydrogen and the refrigerant, and pressure drop across the PFHX channels during hydrogen liquefaction using Helium (He) as refrigerant. This is found in the paper *"Hydrogen ortho-para conversion: process sensitivities and optimisation"* by O'Neill et al. [22]. State-of-the-art H₂ EOS of Leachman et al. was used to calculate the enthalpy of ortho, normal, and para hydrogen [37], while CoolProp library was used to determine the thermodynamic properties.

The overall process can be modeled as a one-dimensional reactor, where the following aspects are critical: reaction kinetics, heat transfer, and pressure drop

Reaction Kinetics: The conversion of ortho-hydrogen to para-hydrogen is represented as:



where Q_{conv} is the heat released during the conversion. Furthermore, the first order reaction rate model ($\dot{r}$) is provided by Donaubaue et al. [40] is as follows:

$$\dot{r} = k \rho_{\text{molar}} (x_{\text{p}}^{\text{eq}} - x_{\text{p}}), \quad (2.14)$$

where k is the rate constant given as:

$$k = \frac{e^{(-\frac{E_a}{RT})}}{x_{\text{p}}^{\text{eq}} (a \cdot \rho_{\text{mol}} + b)}. \quad (2.15)$$

Where the activation energy is given by E_a , and constant parameters a and b are assumed to be constant and independent of temperature, pressure, and concentration. Three sets of experimental data have been used to curve fit their values ([41–43],) of $-336.45 \text{ J mol}^{-1}$, 2.20×10^{-3} and $-35.11 \times 10^{-3} \text{ m}_{\text{cats}}^3/\text{m}_{\text{H}_2}^2$, respectively. R is the universal gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$), T is the temperature, ρ_{molar} is the molar density of hydrogen (in mol m^{-3}), x_p is the current mole fraction of p- H_2 , $x_{\text{para}}^{\text{eq}}$ is the equilibrium concentration of p- H_2 . The effects of adsorption and desorption on the catalyst surface were also added to the first-order equation previously discussed using the Langmuir-Hinshelwood technique. However, in contrast to the experimental conversion results, this adjustment did not produce any discernible improvement [22]. As a result, $x_{\text{para}}^{\text{eq}}$ is expressed as a function of temperature [22]:

$$x_p^{\text{eq}} = \frac{0.1}{\exp\left(\frac{-175}{T}\right) + 0.1} - 7.06 \cdot 10^{-9} T^3 + 3.42 \cdot 10^{-6} T^2 - 6.2 \cdot 10^{-5} T - 0.00227. \quad (2.16)$$

While the molar rate of p- H_2 production ($\partial N_p / \partial z$) is a function of the conversion rate ($\dot{r}$):

$$\frac{\partial N_p}{\partial z} = \dot{r} \cdot A_f \cdot \varepsilon_{\text{cat}}. \quad (2.17)$$

Using A_f as feed channels' cross-sectional area and ε_{cat} as the catalyst's volume fraction in the feed channels. As a result, the p- H_2 rate of change ($\partial x_p / \partial z$) is denoted by the following relation:

$$\frac{\partial x_p}{\partial z} = \frac{M_{\text{H}_2}}{\dot{m}_f} \cdot \frac{\partial N_p}{\partial z} = \frac{M_{\text{H}_2}}{\dot{m}_f} \cdot \dot{r} \cdot A_f \cdot \varepsilon_{\text{cat}}. \quad (2.18)$$

Where M_{H_2} is hydrogen's molecular mass (2.016 g mol^{-1}) and $\dot{m}_f$ is the mass flow rate of the H_2 feed stream.

In subsection 2.1.4, other kinetic rate models have been analyzed according to latest experimental studies.

Heat Transfer: The heat transfer in the PFHXs during ortho-para hydrogen conversion is influenced by the thermal properties of the feed such as the thermal conductivity, and the interaction with the solid catalyst bed. Key aspects include:

1. Heat Transfer in the Packed Bed: Calculated using the Zehner, Bauer, and Schlünder model, with heat transfer coefficients determined by packed bed Nusselt number correlations for laminar and turbulent flow [44].
2. Coolant Side Heat Transfer: Quantified using Manglik and Bergles' correlation, incorporating PFHX geometric factors such as fin thickness and height [45].
3. Effective Heat Transfer Coefficient: Incorporates extended surface efficiencies for the feed and coolant sides, calculated via Shah and Sekulić's method [46].
4. Overall Heat Transfer Coefficient (U_{eff}): Determined by summing resistances from the feed side, coolant side, and wall, using the feed side contact area (A_f) as a reference:

$$\frac{1}{U_{\text{eff}}} = \frac{d_{h,f}}{8 \cdot \lambda_{\text{eff},f}} + \frac{1}{h_{\text{eff},f}} + \frac{t_w \cdot A_f}{A_w \cdot \lambda_w} + \frac{A_w}{A_c \cdot h_{\text{eff},c}} \quad (2.19)$$

Where $d_{h,f}$ is the feed side hydraulic diameter, $\lambda_{\text{eff},f}$ is the effective thermal conductivity of the feed side [44], A_w is the average contact area from the wall, A_c is the contact area from the coolant side, and λ_w is the wall thermal conductivity [22]. Using this information, the rate of heat transfer between hot and cold side of the PFHX ($\frac{\partial Q_{\text{trans}}}{\partial z}$), and the rate of change of heat of conversion ($\frac{\partial Q_{\text{conv}}}{\partial z}$) can be calculated using the equations below. Where, $p_{h,f}$ is the hydraulic perimeter of the feed side.

$$\frac{\partial Q_{\text{trans}}}{\partial z} = U_{\text{eff}} \cdot p_{h,f} \cdot (T_f - T_c) \quad ; \quad \frac{\partial Q_{\text{conv}}}{\partial z} = (H_p - H_o) \frac{\partial N_p}{\partial z}. \quad (2.20)$$

Pressure Drop: The work of O'Neill et al. [22] demonstrates that the pressure drop in their PFHX reactor model is negligible due to the low velocity and viscosity of hydrogen and coolant. The study used Ergun's equation from 1952 [47]. The model provides a detailed understanding of how various process parameters, including inlet pressure, influence the pressure drop and ortho-para hydrogen conversion. The pressure drop is calculated using equations that consider the density, velocity and diameter of the catalyst particles [22]. The model emphasizes that the conversion kinetics is the primary limiting factor on the exit composition, rather than the pressure drop.

However, it is important to mention that the latest studies such as the one of Fu et al. [48] introduce an empirically derived and specific pressure drop equation for an experimental low-temperature catalytic hydrogen conversion device. This represents a significant contribution because, unlike the Ergun equation, commonly used to describe the flow through porous media, the proposed equation has been optimized for the specific operating conditions of the isothermal reactor used in the study, such as cryogenic temperatures and the presence of a catalytic bed of iron hydroxide. Experimental results showed that Ergun's equation did not accurately calculate the pressure drop in the porous region, especially as the flow velocity increased. Figure 2.5 shows this relation.

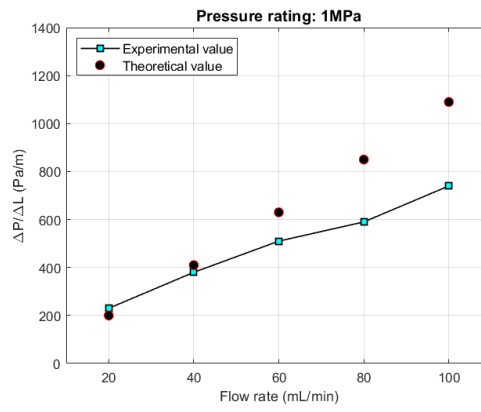


Figure 2.5: Pressure Drop and Flow Rate Curve. Experimental and Theoretical Values Comparison by Fu et al. [48].

The proposed equation instead linearly correlated the pressure drop with the flow velocity and took into account the hydrogen inlet pressure, making it more suitable for dynamic simulation and optimization of the hydrogen liquefaction process under similar conditions. The pressure drop equation used by Fu et al is the following [48]:

$$\nabla P = - \left(A \cdot p_i + \frac{\mu}{B} \right) v \quad (2.21)$$

Where ∇P is the pressure drop of the conversion column (Pa); p_i is the hydrogen inlet pressure (MPa); μ is the viscosity of hydrogen (Pa.s); v is the apparent flow rate (m s^{-1}); $A = 480.80$; $B = 8.38 \times 10^{-10}$ (m).

This improved specificity and accuracy can lead to more reliable simulations and more effective optimization of hydrogen liquefaction processes using ortho-para conversion reactors.

O'Neill et al. Boundary Value Problem: O'Neill et al. conclude the boundary value problem with a system of ODEs to monitor the following fluid properties: para-hydrogen concentration in the feed, feed pressure, feed temperature, coolant pressure, and coolant temperature. The system of ordinary differential equations (ODE) used by O'Neill et al. can be seen in Appendix A [22].

It is important to mention again that the work of O'Neill et al. used helium as a refrigerant, while CoolProp is the thermodynamic library used which utilizes the same EOS algorithms modeled by Span et al. [49]. This Master Thesis will focus modeling the same PFHX using the binary mixture Helium-Neon as the properties of Neon allow an easier compression work. This is an advantage because it reduces the compression size, and thus the total investment costs; nevertheless, careful attention must be paid to its impact on the mixture properties such as viscosity, thermal conductivity, and, therefore,

on heat transfer. Key to bring up the fact that O'Neill's code uses ideal gas transport properties for the helium-neon mixture, which decreases the validity of the results. Nonetheless, more details about how this issue will be dealt are explained in subsection 2.4.1. Tkaczuk et al. [50] EOS will be integrated into O'Neill et al. code [22].

In summary, ortho-para conversion is an essential aspect of hydrogen liquefaction that requires careful consideration of reaction kinetics, heat transfer, and fluid properties. The integration of catalysts within PFHXs and the accurate modeling of these processes are critical to the overall efficiency and economic viability of large-scale hydrogen liquefaction plants. Numerical models help to understand the influence of key process variables on reactor performance and optimization of the entire liquefaction process.

2.1.4. Latest Experimental Kinetic Models Validation

In November 2024, Wijnans et al. [51] published their research paper which examined the kinetics of conversion of ortho-hydrogen to para-hydrogen using the Ionex Type OP catalyst and compared the experimental results with published kinetic models. The importance of spin conversion is crucial in the production of liquid hydrogen, since the para-H₂ form is more stable at cryogenic temperatures.

Experimental data were obtained using a custom-built cryostat and a Raman spectrometer to quantify the ortho- and para-hydrogen content [51]. The experimental results were compared with the kinetic models of Wilhelmsen, Donaubauer, and Zhuzhgov/Buyanov ([52], [40], [53]), developed by adapting experimental data produced in the 1950s and 1960s. The study found that the Wilhelmsen model provided the best overall accuracy in predicting para-H₂ concentration, particularly after re-parameterizing the model on Weitzel et al.'s data [41].

In summary, this study evaluated existing kinetic models for the conversion of ortho-hydrogen to para-hydrogen, while generating new experimental data using a commercial catalyst. The results indicate that the Wilhelmsen kinetic model, with modified parameters, offers predictive capabilities relevant to the current process design and will be tested out in O'Neill's PFHX code to simulate ortho-para conversion. Wilhelmsen et al. kinetic rate model is expressed as follows [52]:

$$\frac{dx}{dt} = \frac{k_w}{C_H} \ln \left[\left(\frac{x}{x^{eq}} \right)^a \frac{1 - x^{eq}}{1 - x} \right] \quad (2.22)$$

Where C_H is the total hydrogen concentration in mol m^{-3} , and the reaction constant (k_w) is temperature $T[\text{K}]$ and pressure $P[\text{bar}]$ dependent according to the following equation:

$$k_w = b + c \frac{T}{T_c} + d \frac{P}{P_c} \quad (2.23)$$

Table 2.5 shows the fitted parameters and their corresponding units and values used by Wijnans et al. [51] in the model of Wilhelmsen et al. [52].

Table 2.5: Parameters of kinetic equation fitted by Wilhelmsen et al. in Wijnans et al. Supplementary Data [51]

Parameter	T_c	P_c	a	b	c	d
Unit	K	bar	–	$\text{mol/m}^3/\text{s}$	$\text{mol/m}^3/\text{s}$	$\text{mol/m}^3/\text{s}$
Value	32.937	12.8377	1.0924	59.7	-253.9	-11.6

2.2. Existing & Conceptual Liquefaction Technologies

This section will focus on the existing hydrogen liquefaction plants Linde-Leuna, Praxair, and Kawasaki Heavy Industries (KHI). Most of them make use of the **Claude** refrigeration cycle. Consequently, careful attention will be given to several conceptual hydrogen liquefaction systems which over the years have adopted the **Brayton** refrigeration cycle. As a result, introductory segments will be provided to first explain in details the key distinctions between these two refrigeration cycles. Lastly, relevant technical and economical statistics are presented such that a comparative analysis can be made between them.

2.2.1. The Claude Cycle

The Claude cycle is a refrigeration process known for its efficiency in cryogenic applications, including hydrogen liquefaction. By combining Joule-Thomson (JT) expansion with work-producing turbo-expanders, it achieves the extreme temperature reductions needed to liquefy hydrogen. According to the study by Yang et al. [54], using HYSYS, the specific energy consumption (SEC) for a Claude cycle is 5.62 kWh/kg_{LH₂} and for a Brayton cycle is 5.87 kWh/kg_{LH₂}. Figure 2.6 shows the high pressure H₂ Claude cycle by Cardella et al. [18].

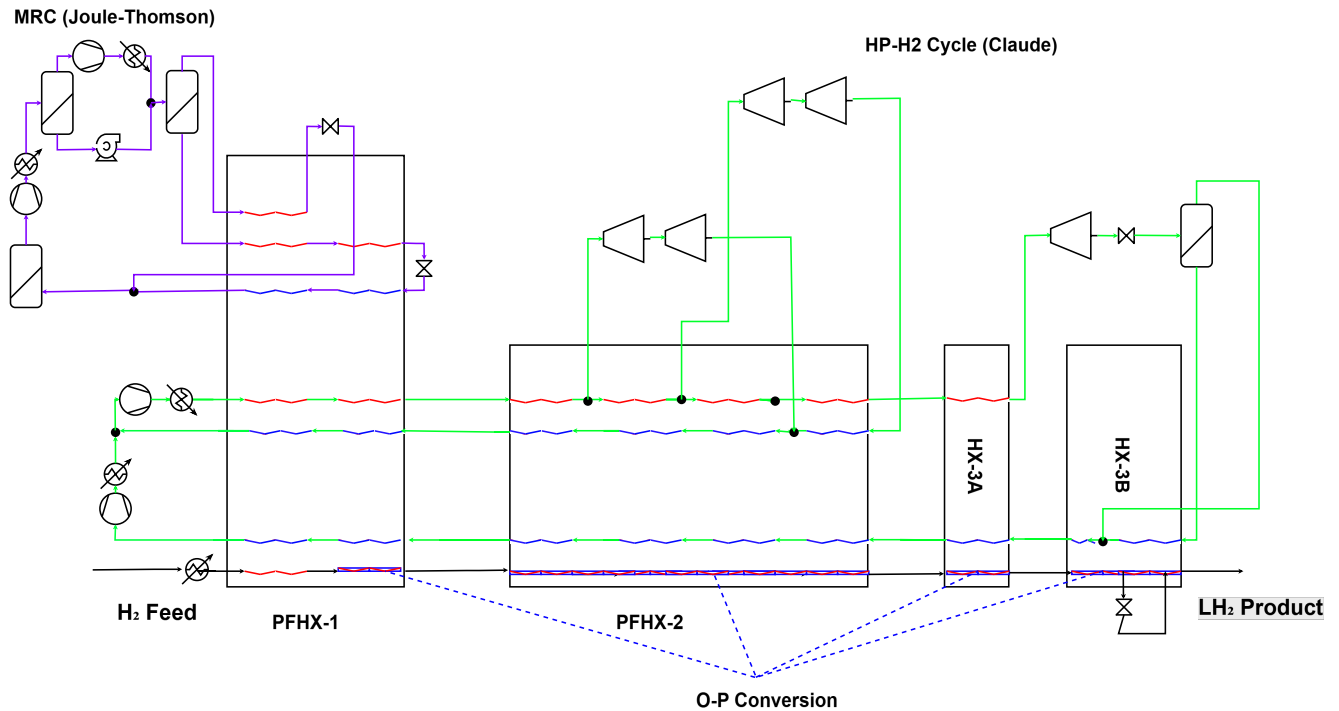


Figure 2.6: Claude Cycle: Schematic diagram for high pressure H₂ by Cardella et al [18]

Core Components and Working Principle The Claude cycle involves compression, heat exchange, and expansion to achieve cryogenic temperatures. High-pressure H₂ is pre-cooled through plate-fin heat exchangers (PFHX), which maximize efficiency. Turbo-expanders then reduce the gas temperature while recovering work, followed by JT expansion to reach the final liquefaction temperature. This integration of turbo-expanders reduces energy consumption, making the cycle ideal for hydrogen liquefaction where efficiency is critical.

Application in Hydrogen Liquefaction The Claude cycle typically includes multiple refrigeration loops with mixed refrigerants to enhance performance. Feed hydrogen is compressed to 20-80 bar and pre-cooled to around 80 K, incorporating catalytic ortho-para hydrogen conversion to prevent boil-off losses. The cryogenic section uses turbo-expanders and JT valves for further cooling. Turbo-expanders provide temperature reductions and recover energy, while JT expansion ensures liquefaction at 20 K and 1-2 bar.

Advantages and Challenges The Claude cycle offers greater thermodynamic efficiency than simpler cycles like Linde-Hampson [55], making it ideal for large-scale plants. Its flexibility allows integration with mixed refrigerants and advanced heat exchangers. Challenges include maintaining cryogenic components and managing precise turbo-expander operations. Catalytic ortho-para conversion must be carefully controlled to avoid disrupting the cryogenic process.

2.2.2. Industrial Hydrogen Liquefaction Plants

Linde-Leuna. The Linde-Leuna hydrogen liquefaction plant in Leuna, Germany was originally built in 2007 with a capacity of 5 tonnes per day (TPD), and has commissioned a second plant in early 2021

[56]. This plant has a specific energy consumption (SEC) of 11.9 kWh/kg_{LH₂} and an exergy efficiency of 23.6% [23]. The plant uses a modified Claude refrigeration cycle, and its refrigerant cycle is made of liquid nitrogen (LN₂) at 80K, reaching a >95% p-H₂ concentration [18].

Praxair. Praxair has five hydrogen liquefaction plants in the United States, with capacities ranging from 20 to 36 TPD. Their largest plant is a double-train liquefier [18]. These plants use a pre-cooled modified Claude cycle, with three heat exchangers and continuous conversion from ortho to para-hydrogen. Nitrogen gas and refrigerant from an external cycle are used as the main refrigerants in the first heat exchanger. In the second heat exchanger, liquid nitrogen and hydrogen recycle flow are used, while in the third heat exchanger only hydrogen flow is used. Praxair systems typically have a SEC between 12.5 and 15 kWh/kg_{LH₂}, with an exergy efficiency (FOM) between 19 and 24% [18].

Kawasaki Heavy Industries Plants. KHI has built the first commercial-scale hydrogen liquefaction plant in Japan. This represents a significant milestone in the Japanese context and demonstrates KHI's commitment to the hydrogen sector. Kawasaki Heavy Industries' (KHI) hydrogen liquefaction system demonstrates an innovative solutions in the industry with an established 5 TPD capacity. Their liquefaction system also uses the Claude refrigeration cycle as well as a combination of established and new technologies to improve efficiency, with a significant focus on reducing energy consumption and operating costs. Their turbo-expanders reach up to 100,000 RPM [18]. Nonetheless, SLC, SEC, and exergy efficiency data are not available.

SK E&S. The construction of South Korean SK E&S plant in Incheon gets the title of the world's largest hydrogen liquefaction plant, with a total capacity of 600 TPD [57]. The facility purifies gaseous by-product hydrogen from a nearby petrochemical facility into high-purity hydrogen and liquefies it by cooling to cryogenic temperatures of 20 K. The plant began full operations in 2024. Nonetheless, technical details have not been made public by the company.

Kimitsu Japan Hydrogen Liquefaction. The Kimitsu Japan Hydrogen Liquefaction plant, owned by Nippon Steel Corporation (NSC), is a demonstration plant for the production of liquid hydrogen for use as a fuel for fuel cell vehicles. It was the first plant in the world to separate hydrogen from coke oven gas (COG), a by-product of steel mills, and liquefy it. The plant has a capacity of 0.2 TPD and uses a **helium Brayton cycle** for the hydrogen liquefaction process. The plant uses the pressure swing adsorption (PSA) method to adsorb, separate and refine hydrogen, which accounts for approximately 55% of coke oven gas (COG) [58]. This process eliminates the generation of off-gases and waste heat that would normally result from chemical reactions such as reforming. The residual gas is fed back into the COG or used as fuel for the steel making process.

2.2.3. The Brayton Cycle

The Brayton cycle is a robust refrigeration process widely applied in hydrogen liquefaction, particularly for its ability to achieve cryogenic temperatures efficiently. By utilizing closed-loop refrigeration with gases such as helium or neon, the cycle ensures precise temperature control for hydrogen liquefaction at 20 K. Figure 2.7 shows a schematic diagram of the Brayton cycle integrated into a conceptual hydrogen liquefaction plant developed by Berstad et al. [59]

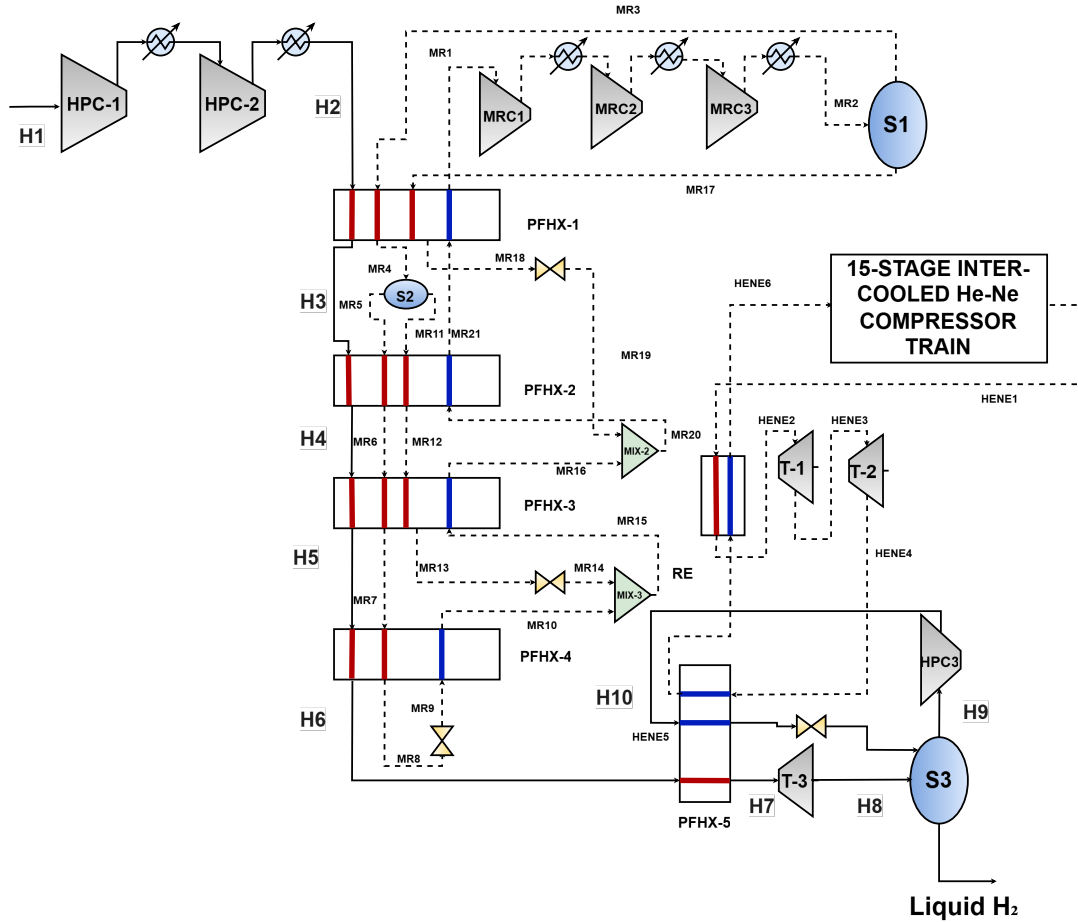


Figure 2.7: Brayton Cycle: Schematic diagram for H_2 liquefier developed by Berstad et al. [59]

Core Components and Working Principles The Brayton cycle relies on compression, heat exchange, and expansion within a closed-loop system. The gaseous refrigerant is compressed, cooled through heat exchangers, and then expanded in turbo-expanders to achieve significant temperature drops. This refrigerant can be Helium, Hydrogen, or a mixture such as Helium-Neon [7]. Furthermore, this cooling is transferred to the hydrogen stream through plate-fin heat exchangers, enabling efficient heat exchange and minimizing energy losses. The closed-loop design and use of turbo-expanders ensure consistent performance and high thermodynamic efficiency [7].

Application in Hydrogen Liquefaction In the Brayton cycle, hydrogen is first compressed and pre-cooled to around 80 K using mixed refrigerants [28]. The Brayton cycle then provides cryogenic cooling, with turbo-expanders reducing the refrigerant temperature to cryogenic levels. This approach is particularly suited for modular and scalable hydrogen liquefaction systems, which ensures flexibility and operational efficiency.

Advantages and Challenges The Brayton cycle offers significant energy savings due to its closed-loop operation and effective use of turbo-expanders. It is highly adaptable, as mixed refrigerants allow their composition to be adjusted to adapt the evaporation curve to the needs of hydrogen cooling, optimizing thermodynamic efficiency, and removing the need for liquid nitrogen [7]. This makes it ideal for small scale hydrogen liquefaction facilities of max 2.5 TPD. However, in recent years, interest in the Brayton cycle has also increased for large-capacity hydrogen liquefaction plants. Nonetheless, the thermodynamic modeling of mixtures can be challenging, especially for cryogenic liquefaction cycles [18]. An accurate EOS is needed to predict mixture behavior and optimize process design with the highest possible outlet p- H_2 concentration as target to reduce boil-off losses.

Table 2.6 shows an easy comparison between the two refrigeration cycles which make up the past, present, and future of cryogenic hydrogen liquefaction systems.

Table 2.6: Comparison of Claude cycle (Linde-Leuna) and Brayton cycle (IDEALHY) hydrogen liquefaction processes, *The study did not provide data for this value, [18]

Project	Linde – Leuna (Established)	IDEALHY, 2013 (Conceptual)
Liquefaction cycle		
Type	LN ₂ pre-cooled Claude	MR pre-cooled Brayton
Capacity [TPD]	5	50
SEC [kWh/kgLH ₂]	11.9	6.7
Exergy efficiency	23.6%	32%
Hydrogen feed		
Pressure [MPa]	2.4	2
Temperature [K]	*	293
para-H ₂ fraction	*	25%
Pre-cooling		
Refrigerant	Nitrogen	Multi-component
Refrigerant cycle	LN ₂ evaporation	MR closed loop
Temperature [K]	80	130
Cryogenic cooling		
Refrigerant	Hydrogen	Helium/Neon
Refrigeration cycle	Claude	Brayton
Temperature [K]	*	26.8
Compressor type	Piston	Turbo
Compressor stages	2 × 2	15
Expander type	Turbo	Turbo
No. of expanders	2	5
Expander efficiency	>85%	80%
O-P conversion	Continuous	Batch and continuous
Expansion		
Expansion device	J–T	Expander
Isentropic efficiency	N/A	80%
Liquid H₂ product		
Pressure [MPa]	0.13	0.2
Temperature [K]	21	22.8
para-H ₂ fraction	>95%	>98.5%

2.2.4. Conceptual Hydrogen Liquefaction Systems

Over the past decades, a lot of effort has been dedicated towards improving efficiency and costs for hydrogen liquefaction systems [18, 23, 24, 60]. The majority of these conceptual designs have been done following simulations using commercial software reaching extraordinary large scales ranging from 50 to 600 TPD [18, 23, 24, 60].

IDEALHY Project. The IDEALHY project presents a 50 TPD high-efficiency hydrogen liquefaction process explicitly designed to halve the specific energy consumption compared to conventional methods [7]. The process employs a multi-stage Brayton refrigeration cycle integrated with pre-cooling and cryo-

genic cooling stages. This system includes compression to 80 bar, mixed refrigerants for pre-cooling, and overlapping Brayton cycles for cryogenic cooling, using Nelium 25 (75% Helium and 25% Neon) as refrigerant; see (Figure 2.7). This process achieves liquefaction at 22.8 K and 2 bar. Its specific energy consumption is approximately 6.7 kWh/kg_{LH₂}, and its exergy efficiency is 32%, with further reductions possible by utilizing waste cold energy from LNG regasification [18].

Berstad et al. Berstad's system is a large-scale hydrogen liquefaction design based on a nine-component mixed refrigerants pre-cooling stage combined with a Brayton cycle; featuring a fifteen-stage compressor, reaching a total of 86 TPD [59]. Moreover, it uses Nelium for the cryogenic liquefaction of hydrogen. The process achieves a specific energy consumption of 6.15–6.516 kWh/kg_{LH₂}, and its exergy efficiency is 44.7–47.1%. A unique feature is the elimination of liquid nitrogen pre-cooling, replaced with a tailored refrigerant mixture, enhancing thermodynamic efficiency by minimizing heat transfer irreversibilities. This system also incorporates catalytic plate-fin heat exchangers optimized for ortho-para hydrogen conversion reaching >98.5% of p-H₂ [59], ensuring effective energy utilization.

Krasae-in. Krasae-in's hydrogen liquefaction concept employs a cascaded mixed refrigerant Brayton cycle for pre-cooling and cryogenic cooling stages. This approach allows significant operational flexibility, with the refrigerant composition adjustable to closely match the cooling curve of hydrogen. The system's modularity and efficiency make it particularly suitable for mid-to-large-scale applications, reaching a potential of 100 TPD [61]. The integration of advanced heat exchangers reduces entropy generation, ensuring an efficient cooling process. Krasae's conceptual system achieves a SEC of a 5.91 kWh/kg_{LH₂}, which represents a 50% reduction compared with the current plant in Ingolstadt with an energy consumption of 13.58 kWh/kg_{LH₂} [61].

QUACK. The QUACK hydrogen liquefier is designed for high efficiency, emphasizing advanced Brayton refrigeration techniques. It employs helium-neon mixtures as refrigerants for cryogenic cooling stages, leveraging their favorable thermodynamic properties. This 170 TPD system achieves remarkable energy efficiencies and incorporates extensive catalytic ortho-para conversion units to handle the exothermic reaction heat effectively. It is characterized by a high exergy efficiency of 56.8%, reducing both operational costs and energy consumption. This results in an overall power requirement of 6.93 kWh/kg_{LH₂}, depending on the pressure of the feed and the temperature of the product [18, 62].

These systems exemplify state-of-the-art engineering approaches to hydrogen liquefaction, leveraging the Brayton cycle's flexibility and integration with modern refrigeration strategies. Their innovations opened up the way for scalable, more efficient, and potentially economically viable hydrogen liquefaction solutions.

2.2.5. Recent Studies on Different Cycles

Tamarona et al. The work done by Tamarona et al. [63] introduces a framework to model a large-scale hydrogen liquefaction process and assess its technical and economic feasibility, focusing on the 125 TPD high pressure hydrogen (HP-H₂) Claude cycle concept, integrated with a Joule-Thomson (JT) mixed refrigerant (MR) pre-cooling cycle [63].

This approach, considered one of the most promising for implementation in the near future, is distinguished by the use of a MR pre-cooling system with a specific composition, which eliminates the need for a liquid nitrogen (LN₂) pre-cooling system, typically responsible for a considerable portion of the total liquefaction energy. The integration and preliminary sizing of key process components, such as plate and fin heat exchangers (PFHX), turbomachinery, and adsorption systems, aim to optimize process efficiency and reduce specific energy consumption (SEC), with the aim of achieving a lower specific liquefaction cost (SLC) than current plants.

The results for the base 125 TPD system show an SEC of 7,241 kWh/kg_{LH₂}, which drops to 6,666 kWh/kg_{LH₂} considering the energy recovery from the turbines. The exergy efficiency is 39.45%, increasing to 42.85% with energy recovery [63]. The estimated specific liquefaction cost (SLC) for the baseline scenario is €1.55/kg_{LH₂} (at an electricity cost of €0.1/kWh), with a significant reduction potential to around €1.0/kg_{LH₂} at lower electricity costs (around €0.05/kWh).

This demonstrates the potential economic viability of integrating large-scale hydrogen liquefaction plants into sustainable energy infrastructures, offering a substantial cost reduction compared to current indus-

trial liquefiers, although cost estimates have inherent uncertainty (Class 4, $\pm 30\%$) and there is room for further improvement through design optimization and refinement.

This study will be the baseline for comparison between a conceptual large-scale hydrogen liquefaction plant using the Claude cycle, and a conceptual one which uses the Brayton cycle.

Yang et al. Another recent study which is interesting to look at is the publication by Yang et al. [54], which introduces a novel integrated approach for hydrogen liquefaction, combining the process with liquefied natural gas (LNG) gasification and an organic Rankine cycle (ORC) [54]. This integration represents a significant advancement over traditional liquefaction systems, as it allows to exploit the cold energy of the LNG, otherwise wasted during gasification, to feed an ORC that in turn contributes to the pre-cooling of the hydrogen. The use of a binary working fluid (ethylene and propane) in the ORC is a further innovation, optimized to maximize the efficiency of heat exchange with LNG and achieve a 2.2% improvement in the use of cold energy compared to the use of a pure refrigerant [54].

This integrated approach not only reduces the specific energy consumption of the liquefaction process, but also promotes greater sustainability through the recovery and valorization of otherwise wasted energy, opening new perspectives for the design of more efficient and environmentally friendly hydrogen liquefaction plants. The exergy analysis performed by Yang et al. also highlights that the proposed integration can lead to a specific energy consumption of $6.59 \text{ kWh/kg}_{\text{LH}_2}$ with an exergy efficiency of 47.0%.

2.3. Equipment for Hydrogen Liquefaction: Latest Conceptual Improvements

This section will cover crucial industrial components which facilitate the hydrogen liquefaction process. These components are:

- Compressors
- Heat Exchangers
- Turbo-expanders

These key components for the equipment design focus in this thesis are the critical parts of the hydrogen liquefaction system, and the most careful considerations will be provided. In addition, other crucial components such as the cryogenic cold box and the Joule-Thomson (JT) valve will also be addressed. Other standard equipment, typical in industrial process plants such as phase separators, pipelines, valves, pumps, coolers, chillers, instrumentation, and measuring devices, while integral to the overall operation, will not be analyzed in depth within the scope of this thesis.

2.3.1. Heat Exchangers

Heat exchangers are essential components in low temperature liquefaction systems. Their main function is to facilitate the exchange of energy between fluids with temperature gradients. Aluminum plate-fin heat exchangers (PFHX) are extensively employed in industrial hydrogen liquefiers, due to their adaptability in accommodating various process channels and capacities. These exchangers offer a substantial heat transfer surface area ($> 2000 \text{ m}^2 \text{ m}^{-3}$ possible [64]), enabling efficient thermal exchange with minimal temperature differences and pressure losses. Additionally, they are space-efficient and cost-effective [65], making them an ideal choice for cryogenic applications [28, 29, 66]. Furthermore, they are designed to sustain pressure of up to 130 bar [18].

The heat exchange rate $\dot{Q}$ is determined by the heat exchange surface area A , the heat transfer coefficient U , and the logarithmic temperature difference between the fluids T_m [40]; these parameters are connected with the following equation for heat exchangers [67]:

$$\dot{Q} = U \cdot A \cdot \Delta T_m \quad (2.24)$$

During the process simulation, the first law of energy balances. Henceforth, the logarithmic temperature difference is calculated from the composite curves of both the hot and cold streams. Consequently, the heat exchange surface area A and the overall heat transfer coefficient U are determined.

In PFHX, hot and cold streams flow through stacked passages, reaching up to 200 passages [29]. As discussed in subsection 2.1.2, hydrous ferric oxide catalysts are present to facilitate the continuous rate kinetics of the ortho-para conversion in the main stream of hydrogen.

Previous works implemented the kinetic PFHX model by Donaubaer et al. [40]. This model uses a one-dimensional steady-state pseudo-homogeneous continuum reactor model of the counterflow-cooled catalytic PFHX, using a combination of mass, heat and momentum transfer, and kinetics; furthermore, it uses Leachman et al. EOS [37]. However, in 2022 O'Neill et al. further expanded the kinetic modeling of PFHX from Donaubaer et al. with various differences: 1) Higher analysis accuracy on the catalyst fraction; 2) Higher level of understanding for the reactor design, analyzing factors such as mixed refrigerants like Helium-Neon by using Tkaczuk et al. equation of state [50]; 3) Comparison of several modeling parameters with the scope to determine the relative influence of heat transfer versus conversion kinetics [22]. Furthermore, the a and b parameters from Equation 2.15 have been modified by taking into account the catalyst fraction and the space-velocity. The results obtained were the following:

O'Neill et al. found that the p-H₂ fraction at the outlet is dependent on the PFHX size. The longer the PFHX, the greater the residence time for both heat exchange and ortho-para conversion. Using pure helium as coolant, a 4 m long PFHX is sufficient to achieve a 99.8% p-H₂ fraction [22]; however when integrating the helium-neon mixture, the sizing of the PFHX might change. Hence, a larger PFHX results in higher CAPEX and OPEX, as well as cold box adjustments. According to Linde, the maximum size for LH₂ industrial heat exchangers is roughly 8.2 m × 3.4 m × 1.5 m, with a core volume ranging from 15 to 30 m³ [64].

2.3.2. Compressors

Compressors are vital components in the hydrogen liquefaction process, serving to increase the pressure of hydrogen and refrigerant streams to facilitate effective cooling and liquefaction. Their role is particularly significant in overcoming the thermodynamic challenges associated with hydrogen's low boiling point and the need for high-pressure refrigeration cycles. The efficiency, reliability, and design of compressors directly impact the exergy efficiency and energy consumption of the liquefaction plant [29]. In a hydrogen liquefaction system, compressors can consume up to 90% of the overall power consumption [18, 29, 59]. In hydrogen liquefaction, compressors are employed in several stages:

- **Feed Hydrogen Compression:** Hydrogen feedstock is compressed to pressures of up to 80 bar before entering the pre-cooling and cryogenic refrigeration stages [18]. This compression reduces the work required in subsequent cooling stages. For the purpose of this thesis, the pressure range goes from 2 bar up to 75 bar.
- **Refrigerant Compression:** Compressors are used in the refrigeration loops, such as those involving mixed refrigerants like helium-neon, to maintain the necessary pressure levels for effective heat exchange and cooling in cryogenic heat exchangers [18]

An elevated compression efficiency reduces energy demands and enhances the liquefaction process's economic feasibility, making compressor selection and optimization a critical aspect of plant design.

Several types of compressor are commonly used in hydrogen liquefaction plants, each suited to specific operational needs. The most common are reciprocating piston compressors, rotary screw compressors, and turbo-compressor [29, 68].

Reciprocating compressors employ pistons within cylinders to compress gas in discrete stages. They are capable of reaching high efficiencies ($\eta_{is} = 0.75 - 0.92$) and precise flow control [29]. However, they are specifically installed thanks to their high pressure ratios per stage; with a range per stage of 1.6-2.7 [69]. In installations, 4 multi-stage compressors are implemented in the liquefaction system. However, they require a higher maintenance level and most of them cannot provide uninterrupted performance for two years [29].

On the other hand, **rotary screw compressors** have a cheaper cost, thus lowering the CAPEX; nonetheless, they have much lower efficiency ($\eta_{is} = 0.65 - 0.75$) [18, 68], leading them to be a suitable option for small scale liquefaction plants (< 3 TPD [70]). As a result, rotary screw compressors will not be considered further.

Turbo compressors are a crucial technology for large-scale hydrogen liquefaction, offering greater efficiency and capacity than reciprocating compressors [71]. They are suitable for large scale applications due to their ability to handle large flow volumes [7, 71]. However, hydrogen compression presents challenges due to its low molar mass, often requiring a large number of stages to reach the desired pressures. Turbo compressors can be classified into centrifugal (or radial) and axial. Axial compressors show greater efficiency in managing large volumes, while centrifugal compressors are more reliable, less subject to fouling and with greater operational flexibility [29]. Despite their advantages, hydrogen turbo compressors must overcome limitations related to the mechanical resistance of the impeller and hydrogen embrittlement, which occurs when ferrous materials lose ductility [72]. Currently, these compressors are not yet widely used in industrial hydrogen liquefaction plants [63], but research is focusing on the development of high-speed centrifugal compressors with more resistant materials to reduce the number of stages required and increase compression efficiency. As a result, this type of compressors will be used in the process modeling.

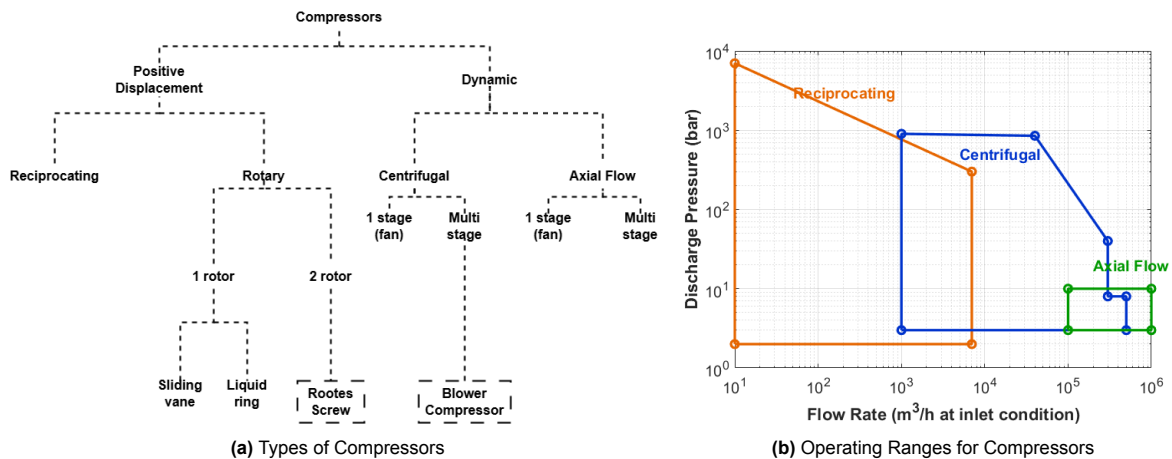


Figure 2.8: Common types of compressors found in process engineering and their operating ranges

However, compressors operating at cryogenic conditions must be designed to handle extreme temperatures and pressures, increasing capital and maintenance costs [63]. Additionally, a hydrogen liquefaction plant needs special attention when using Helium-Neon as mixed refrigerant. Although it is found that increasing the molecular weight of the refrigerant (by adding Neon) can significantly reduce the size of the refrigerant compressor, and as a result the CAPEX [73]; mixtures can also lead to the potential formation of solids, also known as the *freeze-out* process. As a result, compressors using refrigerant blends therefore require specific consideration of the refrigerant composition [18]. Nonetheless, for the purpose of this master thesis, freeze-out will not be considered.

Henceforth, modern compressors, particularly turbo centrifugal and axial compressors, offer high isentropic efficiencies (up to 85%), minimizing energy losses [18]. Furthermore, compressors can handle a wide range of flow rates and pressures, making them adaptable to various plant capacities [18]. Lastly, with an appropriate regular maintenance, compressors will provide a reliable and consistent performance due to their elevated lifespan [18].

2.3.3. Turboexpanders

Turbo expanders or turbines are crucial components in hydrogen liquefaction, playing a central role in refrigeration cycles by enabling efficient cooling at cryogenic temperatures. By converting high-pressure gas into a low-pressure, low-temperature stream, turbo expanders significantly contribute to the reduction of both hydrogen's and the coolant's temperature, allowing for its liquefaction. Their performance and proper selection are critical for the overall efficiency and reliability of the liquefaction process [18].

Turbo expanders operate based on the principle of isentropic expansion, where a high-pressure gas undergoes expansion in a turbine to produce work. This process reduces the gas's temperature and

pressure simultaneously, making turbo expanders ideal for cryogenic refrigeration systems. Their primary roles in hydrogen liquefaction include:

Cryogenic cooling: turbo expanders provide the deep cooling required to bring hydrogen gas from pre-cooled states (around 80–100 K) to cryogenic temperatures below 30 K [18]. **Energy recovery:** the work produced during expansion is often used to drive compressors or generate electricity, improving the plant's overall energy efficiency [63]. According to Berstad et al. by recovering 80% of the power from turbines can reduce the SEC by 7% [59]. Nonetheless, because of their high CAPEX, the energy recovery only becomes an attractive addition for large scale liquefaction plants with turbines > 100 kW. **Ortho-Para conversion support:** Turbo expanders are typically integrated with catalytic heat exchangers where ortho-para conversion occurs. The cooling achieved through expansion helps maintain the necessary temperature profile for effective conversion [18, 22].

Selecting the appropriate turbo expander for hydrogen liquefaction involves careful consideration of operating conditions, efficiency, material selection, dynamic balance, and integration with other process equipment. Turbo expanders must handle specific temperature, pressure, and flow rate ranges of hydrogen and refrigerant streams, with typical inlet conditions involving pressures between 20 and 80 bar and temperatures from 80 K to 300 K [18]. High isentropic efficiencies, often exceeding 85% are essential to minimize energy loss and maximize cooling capacity [18, 63]. Material selection is also critical, as expanders must maintain structural integrity and thermal performance at cryogenic temperatures, often using high-strength alloys such as stainless steel and aluminum [18]. Common turbo expanders are radial and axial flow turbines on the refrigerant side. Their difference lies under efficiency and costs; axial turbines are more expensive but also more efficient at $Re > 10^6$ [74]. On the other hand, the benefit of radial turbines is that the work recovered from one stage is equal to that of two or more axial turbine stages [74]. Even though the state-of-the-art turbines for hydrogen liquefaction systems are oil-bearing and gas-bearing [29, 70], common conceptual design based on the brayton cycle such as IDEALHY, Krasae-in, and Berstad et al. [7, 59, 61] utilize a turbine expander on the H_2 and refrigerant side; but no specific information is given towards which type of turbo expander is used [18].

Performance metrics for turbo expanders include isentropic efficiency, expansion ratio, specific cooling power, and reliability. Isentropic efficiency measures how closely the expander's performance approximates ideal isentropic conditions, with higher efficiencies reducing energy consumption and enhancing cooling. Common isentropic efficiencies are found to be in the range of 80-90% [18]. The expansion ratio—defined as the ratio of inlet to outlet pressure, determines the temperature drop achieved, with higher ratios providing greater cooling but potentially increasing mechanical stress if not optimized [18]. Specific cooling power, indicating the cooling achieved per unit mass flow of gas, directly impacts the plant's liquefaction capacity [18]. Reliability and maintenance are also paramount, as expanders must operate continuously in extreme conditions, often requiring advanced bearings and optimized lubrication systems to minimize wear [18, 63].

Latest Sensitivity Analysis

Furthermore, the study conducted by Torres-Gomez et al. [75], proposes different expansion configurations to improve the efficiency of hydrogen liquefaction, focusing on the implementation of single-stage turbo expanders in the final expansion. The comparative analysis is based on three specific cases, each with a different approach to the expansion phase.

Case A: Joule-Thomson (JT) Valve This is the reference case, using a standard JT valve for isenthalpic expansion. Hydrogen is expanded from a temperature of about 31 K to the saturation temperature at 1.05 bar (20.49 K). During this process, the isenthalpic expansion maintains a nearly constant temperature until reaching the saturation curve, which leads to a significant increase in entropy and low yield, since a larger amount of hydrogen must be recirculated in the cycle [75]. This type of expansion is common in existing liquefaction plants, but has limitations in terms of efficiency.

Case B: Fully wet turbo expander In this configuration, the hydrogen is cooled to a lower temperature (21-22 K) while maintaining the same inlet pressure, so the expansion ends on the liquid saturation curve at the outlet pressure. This guarantees a 100% yield. Furthermore, the increase in entropy dur-

ing the expansion is significantly smaller than in Case A [75]. However, this approach requires a higher input of work from the auxiliary helium cycle to further cool the hydrogen by about 10 K.

Case C: Single-phase turbo expander and Joule-Thomson valve in series This case represents a compromise between the two previous ones. The hydrogen is initially cooled to about 31 K, as in Case A. The expansion is performed first by a turbo expander in the single-phase region to an intermediate pressure, followed by a JT valve which further expands the hydrogen to the desired pressure. The entropy increase is lower than in Case A, leading to higher yield and efficiency [75]. This configuration takes advantage of the advantages of both components, reducing losses associated with isenthalpic expansion and minimizing the need for excessive cooling.

These configurations were analyzed using the WE-NET cycles [76] and Berstad et al. [59] as baseline. For the WE-NET cycle, Case C showed a 13.3% yield increase and a 4.6% exergy efficiency improvement compared to Case A. For the Berstad et al. cycle, Case C showed an exergy efficiency improvement of 1.6% at 50 bar and 2.7% at 80 bar. The study concludes that option C offers an improvement over the sole use of JT valves, with the added benefit of not operating in two-phase conditions, avoiding cavitation problems [75].

2.3.4. Other Components

Cryogenic adsorbers and cold box are other crucial components that make up the liquefaction system. Cryogenic adsorbers play a crucial role in hydrogen liquefaction by removing trace impurities such as water, oxygen, nitrogen, and hydrocarbons that could freeze at cryogenic temperatures and cause blockages in process equipment. These adsorbers operate at very low temperatures, typically within the range of 80–120 K, ensuring that even the smallest contaminants are effectively captured before hydrogen enters the deeper cooling stages. The adsorption process relies on materials like activated carbon or molecular sieves, which exhibit high selectivity for specific impurities under cryogenic conditions. The efficiency of these adsorbers directly impacts the reliability and longevity of heat exchangers and turbines in the liquefaction cycle, minimizing maintenance downtime and optimizing process stability [54].

The cold box is a fundamental structural component in hydrogen liquefaction plants, housing key cryogenic equipment such as heat exchangers, expanders, and phase separators. It provides thermal insulation and structural integrity, ensuring that the internal components operate at the required low temperatures with minimal heat ingress from the external environment. Cold boxes are usually constructed from stainless steel and insulated with multilayer vacuum insulation or perlite, the cold box maintains temperatures as low as 20 K while withstanding mechanical stresses caused by thermal expansion and contraction [77]. Its design must accommodate the integration of multiple cryogenic streams while minimizing pressure drops and ensuring efficient heat exchange. Given its critical role, optimizing the cold box layout is essential to achieving high liquefaction efficiency and reducing energy consumption [7].

2.4. Cryogenic Mixed Refrigerants for Liquefaction

Cryogenic mixture refrigerants (MR) are increasingly utilized in hydrogen liquefaction processes due to their thermodynamic efficiency and ability to closely match the cooling curve of hydrogen gas. Unlike single-component refrigerants such as nitrogen, mixed refrigerants offer improved heat transfer properties, reduced energy consumption, and better adaptability to different temperature ranges. This section will discuss the advantages of MR, their typical compositions, and their role in hydrogen liquefaction cycles. The following subsections will further analyze specific properties of helium-neon mixtures, the application of mixed refrigerants in pre-cooling cycles, and the associated design challenges and advantages.

2.4.1. Transport Properties of Helium-Neon (Helium) Mixtures Using Entropy-Scaling Method

Helium-neon mixtures are of particular interest in cryogenic applications due to their low boiling temperatures and unique thermodynamic properties. Helium, with a boiling temperature of about 4.2 K, is often used as a low-temperature refrigerant [50]. The addition of neon, which has a boiling point of

about 27 K, allows the properties of the mixture to be modulated for applications over a wider temperature range [50]. Helium-neon mixtures are used in Brayton refrigeration cycles, providing cooling at various intermediate temperatures. The IDEALHY is a conceptual design of a 50 TPD liquefier based on an MR pre-cooled 'nelium' (75% helium, 25% neon) mixed refrigerant in a dual Brayton refrigeration cycle [7, 18]. Accurate thermodynamic modeling of these mixtures is critical to optimize the performance of liquefaction processes. Tkaczuk et al. equation of state (EOS) is used in the O'Neill et al. PFHX code to describe the thermodynamic properties of helium-neon mixtures, [22, 50].

However, it is important to highlight that this model used ideal gas transport properties, which compromised the validity of precise results. As a result, in collaboration with the University of Stuttgart, specifically within the Institute of Thermodynamics and Thermal Process Engineering, the entropy scaling method will be used to accurately estimate these transport properties such as viscosity and thermal conductivity.

The entropy scaling method is applied to estimate the transport properties of helium-neon (nelium) mixtures at cryogenic temperatures, with the aim of overcoming the limitations of the ideal gas approximation currently used in heat exchanger calculations. As highlighted by Schmitt et al. and Lötgering-Lin et al. [78, 79], entropy scaling is based on the observation that transport properties, such as viscosity and thermal conductivity, can be related to the configurational entropy of the fluid. Instead of modeling transport properties as a direct function of temperature and pressure, entropy scaling expresses them as a function of configurational entropy, which greatly simplifies modeling and allows capturing the complex behavior of the fluid over a wide range of conditions [79]. This approach, combined with a framework for thermodynamic equations of state (FeO_s), allows reliable extrapolations even in regions of state where experimental data are scarce. For mixtures, entropy scaling requires a mixing model to calculate the configurational entropy of the mixture from the properties of the pure components, providing an accurate prediction without compromising generality [79]. Additionally, the use of accurate EOS, such as molecular ones, is crucial to describe configurational entropy and predict transport properties in unexplored regions [78].

2.4.2. Use of Mixed Refrigerants in Pre-Cooling Cycles

In hydrogen pre-cooling cycles, the use of mixed refrigerants (MR) offers significant advantages over pure refrigerants. Common MR compositions include hydrogen, nitrogen, neon, helium, and hydrocarbons ranging from methane to butane [18]. A key advantage of MRs is the ability to modify the mixture composition to match the evaporation curve to the hydrogen cooling curve, reducing temperature differences and improving overall thermodynamic efficiency [18]. Blends may also exhibit improved heat transfer properties and compression characteristics compared to pure refrigerants [18]. Conceptual processes have explored the use of MR in pre-cooling cycles at different temperatures, such as 130 K, with gradual removal of components based on temperature [18, 59]. Mixed refrigerants are more advantageous than nitrogen because of the large temperature difference between the cooling and heating curves which limits heat recovery [18, 29, 59]. Further details about the MR in the pre-cooling stage will be given in ??.

2.4.3. Design Challenges and Advantages

Designing hydrogen liquefaction systems with MR presents several challenges and advantages. One of the main challenges is the selection and optimization of the refrigerant composition, which must balance the requirements of different cooling stages and the thermodynamic properties of the components [18]. Additionally, the possibility of freezing of the heavier refrigerant components at cryogenic temperatures requires careful design of heat exchangers to prevent blockages [18]. Despite these challenges, the benefits of using MR are substantial. Improved thermodynamic efficiency and the potential for lower energy consumption are the main advantages of MR systems, leading to reduced operating costs [18]. Furthermore, MR offer greater flexibility in the design of liquefaction systems, allowing for more precise tailoring to specific cooling requirements and increased efficiency [18]. Using a dual mixed refrigerant (DMR) system, as proposed by some studies, can eliminate the potential for freezing problems and improve overall process efficiency [80]. A DMR system uses two separate refrigeration cycles, each with distinct MR compositions that maximize heat recovery and reduce energy consumption.

2.5. Process Economics of Hydrogen Liquefaction

The economic evaluation of hydrogen liquefaction is crucial in determining the feasibility and competitiveness of liquid hydrogen production. The process economics consists of capital expenditure (CAPEX), operating expenditure (OPEX), and specific liquefaction cost (SLC), all of which are influenced by economies of scale, equipment efficiency, and energy consumption.

Hydrogen liquefaction is inherently expensive as a result of the energy-intensive nature of cooling hydrogen to cryogenic temperatures. The CAPEX includes costs associated with planning, construction, and equipment installation, while the OPEX includes variable and fixed costs such as energy consumption, maintenance, and operational labor [29, 63].

A key parameter in process economics is the Specific Liquefaction Cost (SLC), which is defined as:

$$SLC = \frac{CAPEX + OPEX + OM}{\dot{m}_{LH_2}} \quad (2.25)$$

OM refers to operations and maintenance costs, nonetheless they will be taken into account towards the final SLC value. Recent studies indicate that large-scale hydrogen liquefiers can achieve an SLC of less than \$1 per kg_{LH₂}, a significant improvement over the typical range of \$2.5 - \$3.0 per kg_{LH₂} in current industrial plants [18, 29]. This reduction is largely attributed to process optimization, high-efficiency turbo-expanders, and economies of scale. Furthermore, sensitivity analyses show that the cost of electricity has a substantial impact on hydrogen liquefaction economics, as well as feed pressure. Scaling up to 250–500 TPD can further reduce costs due to increased efficiency in component utilization and operational integration [63].

Hence, the next sections will dive deeper into both CAPEX and OPEX, while integrating common cost estimation models for liquefaction found in the literature.

2.5.1. Capital Expenditures

Capital expenditure (CAPEX) for hydrogen liquefaction includes the initial investment required for plant construction, including acquisition of cryogenic equipment, refrigeration cycles, compressors, expanders, storage tanks, and auxiliary infrastructure. Due to the complexity of liquefying hydrogen to 20.38 K, CAPEX is a significant factor that affects the economic viability of large-scale hydrogen liquefaction plants. According to the Association for the Advancement of Cost Estimating International (AACE International), the preliminary cost estimates "Class 4" has the goal to compare different designs, relying on limited cost information and preliminary design results, with an estimated accuracy of ±30% [81]. Several scientific papers provide correlations and models to better estimate "Class 4" capital expenditures [81–83].

According to Towler & Sinnott [81], CAPEX includes the following costs:

- **The inside battery limits (ISBL) investments** - This represents the total cost of the plant, covering both direct and indirect field expenditures.
 - **Direct field costs:**
 1. Expenses related to key process equipment such as reactors, compressors, and heat exchangers.
 2. Costs of bulk materials like piping, catalysts, and instrumentation.
 3. Civil engineering works.
 4. Labor and supervision for installation.
 - **Indirect field costs**
 1. Construction-related expenses, including temporary structures, site utilities, and water supply.
 2. Costs associated with field operations and services.
 3. Insurance coverage.
 4. Labor benefits and associated expenses.

5. Miscellaneous overhead costs, including legal fees, agent charges, import duties, and administrative expenses.
- **Outside Battery Limits (OSBL) Investment** – These costs involve infrastructure upgrades or additions required to integrate the new plant into the existing site. This may include setting up power generation systems, cooling towers, and maintenance facilities.
 - **Engineering Costs** – Also known as contractor charges or home office expenses, these cover all costs related to engineering and design necessary for project execution. They include:
 1. Detailed design and engineering for process equipment and piping systems.
 2. Procurement of bulk materials and key plant components.
 3. Supervision of construction activities.
 4. Administrative costs for tasks like project management, quality inspections, and compliance oversight.
 5. Bonding costs.
 6. Contractor's profit margin.
 - **Contingency Charges** - These are additional funds allocated in the project budget to account for any unexpected expenses or deviations from the estimated costs.

Offsite costs are generally estimated as a percentage of ISBL costs during the initial design phase, typically ranging from 10% to 100%, depending on project scope and the extent of its impact on existing infrastructure. In petrochemical projects, these costs generally range between 20% and 50% of ISBL costs, with a default estimate of 40% when specific details are not available [81]. Established locations, such as "brownfield" projects with existing infrastructure, tend to have lower offsite costs. In contrast, projects developed in entirely new locations, like "greenfield" projects, generally incur higher offsite costs.

A common guideline for estimating engineering costs is 30% of ISBL plus OSBL costs for smaller projects, while for larger projects, this value is around 10% of ISBL plus OSBL costs. Furthermore, contingency costs should be at least 10% of ISBL plus OSBL costs for any project. If the project involves low technological readiness, higher contingency allowances are necessary. In such cases, contingency costs can reach up to 50% of ISBL plus OSBL costs [81].

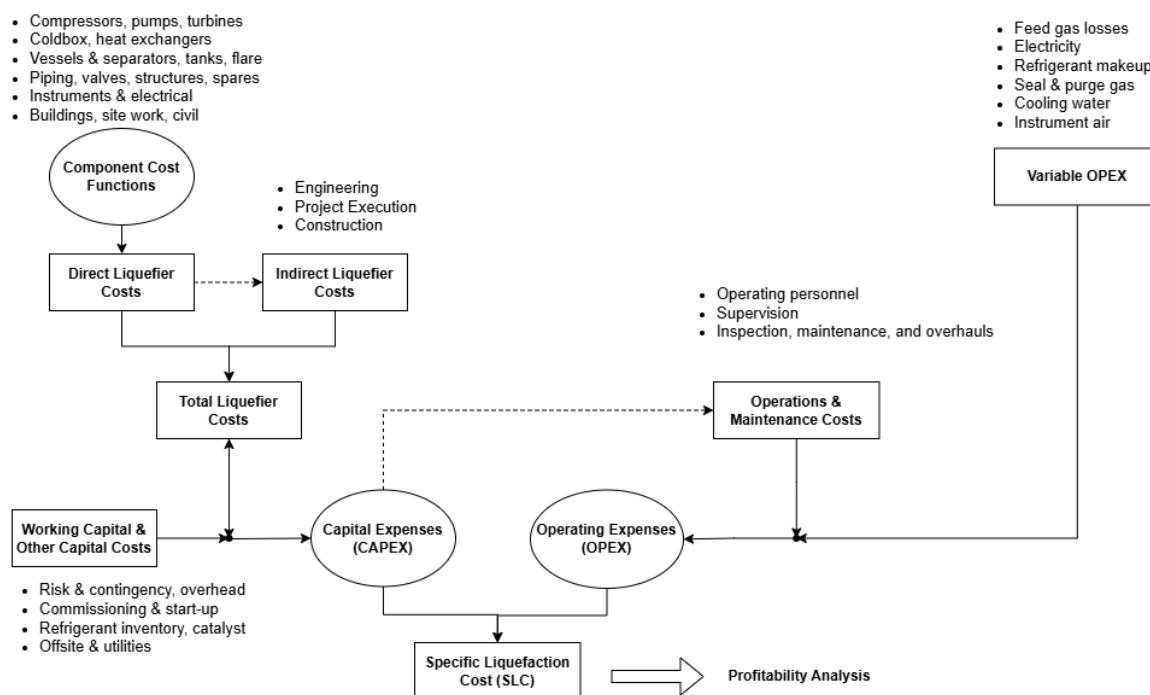


Figure 2.9: Cardella et al. proposed step-by-step methodology for estimating the SLC of the liquefier [84]

Syed et al. and Cardella et al. [84, 85] conducted capital cost evaluations specifically for hydrogen liquefaction systems. Both studies utilized a cost estimation model based on process equipment cost functions incorporating adjusted factors from existing literature to estimate ISBL investment. However, Cardella et al. developed cost correlations tailored specifically for hydrogen liquefaction equipment, validating their model using cost data from industrial manufacturers. Nonetheless, the fitted correlations derived from their analysis remain unpublished. Figure 2.9 illustrates the schematic approach proposed by Cardella et al. for assessing the economic feasibility of hydrogen liquefaction plants [84].

Currently, there are multiple cost estimation softwares. The Aspen Process Economic Analyzer (APEA) from Aspen Technologies will be used as cost estimating software. This program achieves high-level cost estimations based on Aspen ICARUS Technology algorithm. APEA is directly included into Aspen ONE package.

Cost Escalation: Material and labor costs fluctuate due to inflation. To ensure accurate projections, outdated cost data must be adjusted using publicly available cost indices. The equation below allows for the adjustment of equipment and process plant costs over time [81]:

$$\text{Cost in year A} = \text{Cost in year B} \times \frac{\text{Cost index in year A}}{\text{Cost index in year B}} \quad (2.26)$$

For precise estimates, costs should be broken down into labor and materials. However, using composite indices from trade journals is often more practical. The Chemical Engineering Plant Cost Index (CEPCI), published monthly in *Chemical Engineering*, is the most widely used in the US process industry. CEPCI includes sub-indices for equipment, construction labor, buildings, and engineering supervision. Other journals, such as the *Oil and Gas Journal*, *Engineering News Record*, and *Process Engineering*, also publish cost indices regularly [81].

2.5.2. Operating Expenditures

Operating expenditure (OPEX), also known as the cash cost of production (CCOP), represents the cost of running a process plant, including packaging, distribution, and overhead expenses [83]. OPEX

is categorized into variable and fixed production costs [81]. Variable costs depend on the plant's production level and include raw materials, utilities such as fuel, steam, and cooling water, electricity, and consumables such as solvents, acids, and catalysts that require replenishment. In chemical plants, raw materials account for 80-90% of OPEX [81], utilities for 5-10% [81], and consumables for less than 3% [81].

In hydrogen liquefaction processes, raw material costs, hydrogen gas in this case, are typically excluded from cost assessments, except for losses, as liquefaction does not increase hydrogen's intrinsic value, but rather enables its storage and transportation over long distances. Consequently, utility costs, particularly electricity, dominate operating expenses, significantly impacting overall liquefaction costs. Cardella et al. [29, 84] identified key variable costs in large-scale hydrogen liquefaction, including electricity, hydrogen feed gas losses, liquid nitrogen, refrigerant make-up, cooling water make-up, and seal and purge gas. The specific cost of these elements varies across cost estimation models as they are often based on assumptions and can differ significantly depending on source and operational conditions [18].

On the other hand, fixed costs remain unchanged regardless of production output, and according to Towler & Sinnott [86] include:

- Operating labor: Based on shift schedules and operator salaries.
- Supervision: Typically 25% of operating labor costs.
- Direct salary overhead: 40-60% of labor and supervision expenses.
- Maintenance: Estimated at 3-5% of ISBL investment, depending on plant reliability.
- Property taxes & insurance: 1-2% of ISBL fixed capital.
- Rent of land & buildings: 1-2% of ISBL + OSBL costs.
- General plant overhead: Covers HR, R&D, IT, and finance.
- Environmental charges: Typically 1-2% of ISBL + OSBL costs, allocated for regulatory compliance.
- Licensing fees & royalties: Expenses related to intellectual property or proprietary technologies.
- Capital charges: Interest on project loans, excluding equity returns.
- Sales & marketing costs: Minimal for raw commodities, but can be significant for branded products.

OPEX plays a crucial role in determining a plant's long-term profitability, making cost optimization essential for economic sustainability. Furthermore, it is imperative for these costs to be analyzed at the very early stages of the design. Although Towler & Sinnott [86] cost estimate for the OPEX is available, the previously mentioned Aspen Process Economic Analyzer (APEA) will be used in this thesis work, as it has built-in features for investment analysis.

2.5.3. Economy of Scale

Economy of scale plays a crucial role in reducing the costs of hydrogen liquefaction. Increasing the capacity of liquefaction plants leads to a decrease in specific capital costs per unit of product [63]. This principle, resulting from the division of labor, is evident in the process plant industry, where larger plants have lower construction costs per unit of product [87, 88]. Compressors, which account for a significant share of investment costs, particularly benefit from this dynamic, with specific costs decreasing as capacity increases. Increasing plant capacity allows for a better distribution of fixed costs, such as engineering, infrastructure and labor, over a larger volume of liquefied hydrogen, resulting in a reduction in unit cost.

The cost curve method can be used to show the economy of scale of industrial chemical plants. By doing this, engineers can make an order of magnitude estimate for the capital cost of the plant [86]. This is achieved using the following equation [67].

$$C_2 = \frac{C_1}{S_1^n} \times S_2^n = a S_2^n. \quad (2.27)$$

Hence, from Equation 2.27, the capital cost per unit of production is calculated using the following equation:

$$\frac{C_2}{S_2} = aS_2^{n-1}. \quad (2.28)$$

Amos used Equation 2.27 in 1998 to find the base cost/size and the value of the constant n using published hydrogen storage and transport cost data. The IDEALHY project used Equation 2.27 to deduce the capital cost of a 40 TPD hydrogen liquefaction plant from a base reference of 50 TPD plant. Tamarona et al. used 2/3 for the constant n , and 7.37 M€ for a at 50 TPD and zero for a 0 TPD plant [63].

The value of n is almost always less than 1 [67]; as a result, the value of $n - 1$ will be negative. This means that as S_2 increases, the cost of unit production C_2/S_2 , decreases. Hence, a lower capital cost per unit of production enables owners of larger plants to offer more competitive pricing while still recovering their initial investment. This incentivizes chemical companies to construct larger-scale facilities. In short, to achieve competitive costs in hydrogen liquefaction, it is essential to exploit economies of scale by building plants with greater production capacity. This phenomenon is also known as *learning curves*, and it generally represents the correlation between a technology's specific costs (adjusted for inflation) as the dependent variable and its cumulative capacity or experience as the independent variable [89]. The logarithmic linear function is the most commonly used model for depicting this learning trend, as it offers simplicity and a strong fit to observed data. The log-linear equation for the experience curve is given as follows [90]:

$$c = c_0 \left(\frac{q}{q_0} \right)^{-b} = a \cdot q^{-b} \quad (2.29)$$

Here, the main parameter is exponent b , also known as the learning coefficient [90]. This term is used to calculate both the progress ratio (PR) and the learning rate (LR), using the following equations:

$$\text{PR} = 2^{-b} \quad \text{and} \quad \text{LR} = (1 - \text{PR}) \quad (2.30)$$

The learning rate refers to the rate at which production costs decrease as cumulative production or operational experience increases [91]. For example, a learning rate of 20% means that every time cumulative production doubles, the unit cost decreases by 20%. The progress ratio is used to describe how production costs decrease as cumulative output increases [91]. It represents the fraction of cost retained when cumulative production doubles. A lower PR means the plant is efficiently optimizing processes.

The concept of learning curves has been a valuable tool for modeling technological advancements, shaping policy decisions, and predicting future costs associated with energy technologies over the past two decades. Figure 2.10 illustrates the learning curves of various electricity generation technologies [92]. By making assumptions about future deployment levels, this relationship can help forecast future cost trends in electricity generation, for example, by assuming that past learning rates will continue unchanged in the future. Focusing on this aspect is hugely important for the 2050 global hydrogen demand ([1, 2, 93, 94]) considering its current elevated prices, which rise doubts for potential investors.

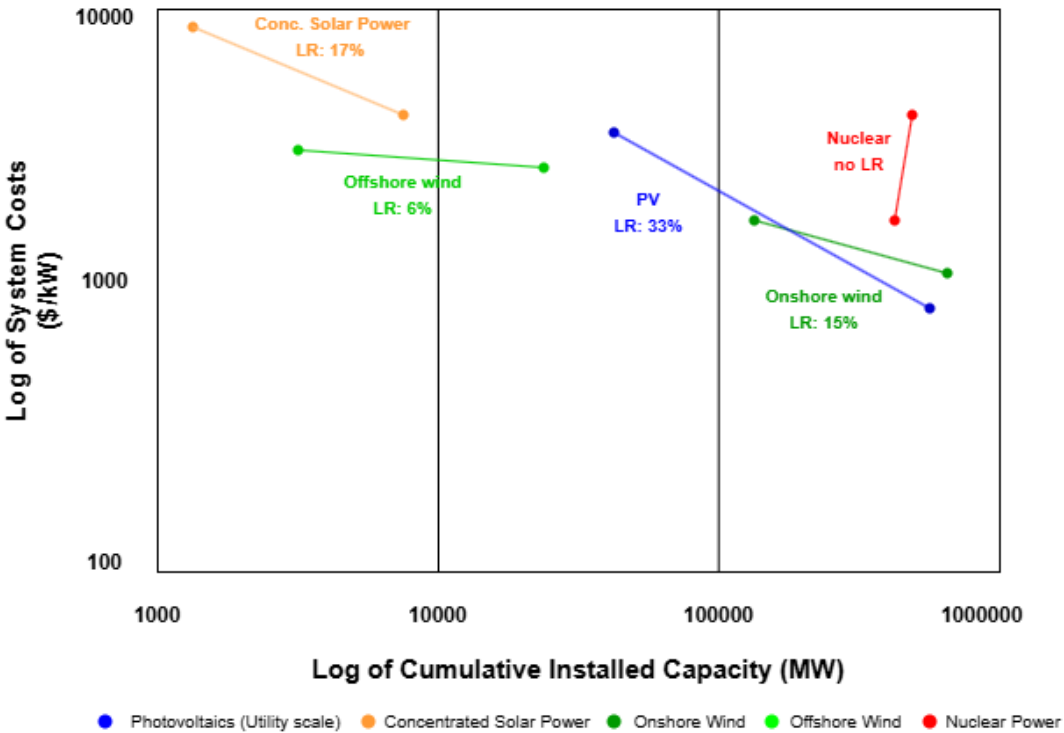


Figure 2.10: Learning curves of multiple power generation sources [95]

3

Methodology

This chapter outlines the methodological approach followed in conducting the techno-economic analysis of hydrogen liquefaction. The study focuses on two primary domains, distinguished by the software environment employed: Python (via PyCharm) and Aspen HYSYS V14. Together, these tools enable a comprehensive approach to process modeling, system integration, and cost estimation.

Within the Python environment, the open-source GitHub repository by O'Neill et al. [96] was employed to simulate the final catalytic plate-fin heat exchanger (PFHX), responsible for achieving ortho-to-para hydrogen conversion. The original codebase was extended by introducing a helium-neon (He-Ne) mixture as the refrigerant, replacing pure helium. This adjustment better reflects realistic cryogenic cycle conditions and offers potential reductions in compressor work.

Complementing the Python-based work, process optimization was performed on an existing Aspen HYSYS model adapted from the thesis by Kerkar et al. [97]. Various process configurations were explored to reduce specific energy consumption and associated costs. For instance, the integration of an ejector in the cryogenic loop was assessed for its potential to reduce CAPEX, though it was ultimately rejected due to significant OPEX penalties; nonetheless, insights around how secondary inlet pressure and outlet mass flow of the excess cold H_2 gas have been analyzed and provided. The finalized process incorporates a hybrid Claude-Brayton cycle, which recirculates cold hydrogen streams to enhance thermodynamic efficiency and reduce overall expenditure.

3.1. Plate-Fin Heat Exchanger

This section presents the modeling and analysis of the catalytic plate-fin heat exchanger (PFHX), a key component responsible for the final stage of ortho-to-para hydrogen conversion. The simulation framework is based on the GitHub repository developed by O'Neill et al. [96], which couples heat transfer, pressure drop, and spin-isomer kinetics within a single computational toolchain. The purpose is to simulate o-p conversion and size the dimensions of the final PFHX present in the simulation process

3.1.1. Helium-Neon Incorporation

Background and Justification: To enhance the thermodynamic performance of the PFHX model, a helium-neon (He-Ne) binary mixture was selected as the refrigerant. The addition of neon increases the molar heat capacity and reduces the volumetric flow rate of the coolant stream, leading to lower compression work and improved efficiency within the cryogenic cycle. This mixture provides a more realistic representation of advanced cooling systems compared to the baseline use of pure helium.

Since the original simulation framework was configured for single-component fluids, its extension to handle binary mixtures required modification at both the fluid property and solver levels. To enable this, the thermophysical behavior of helium and neon was defined using accurate Helmholtz energy-based equations of state. The necessary data were sourced from the supplementary dataset by Tkaczuk et al. [50], which provides high-resolution parameterizations for noble gas mixtures under cryogenic

conditions. These were encoded in JSON format and integrated into the modeling environment using the CoolProp library.

The custom fluid configuration is fully compatible with the CoolProp thermophysical property library and is accessed within the Python model using a dedicated *HeliumNeon* class, ensuring compatibility with all heat and mass transfer calculations in the PFHX boundary value problem (BVP) model. This implementation dynamically updates key properties, including density, viscosity, and thermal conductivity, along the exchanger length, allowing precise simulation of heat transfer and pressure drop under realistic process conditions.

JSON-Based Fluid Property Definition: The integration of the helium-neon mixture into the simulation environment required custom fluid definitions to be embedded into the CoolProp thermodynamic property engine. This was achieved using JSON-formatted data files containing high-accuracy Helmholtz energy EOS parameters for both helium and neon, as published in the supplementary material by Tkaczuk et al. [50]. These files enable CoolProp to internally generate the required thermodynamic surfaces, including pressure, enthalpy, entropy, and derivatives needed for property calculations across the full cryogenic temperature range.

The JSON files, *Helium.json* and *Neon.json*, were placed within the project directory and loaded using the function `setup_CoolProp()` defined in the module *helium_neon.py*. This function registers each fluid with CoolProp's HEOS backend, enabling its use within any *AbstractState* object. The relevant configuration commands are shown below:

```
1 CP.set_config_bool(CP.OVERWRITE_FLUIDS, True)
2 CP.set_config_bool(CP.OVERWRITE_BINARY_INTERACTION, True)
3 CP.add_fluids_as_JSON('HEOS', json.dumps([contents]))
```

The above snippet is found at line 19 from the *helium_neon.py* from O'Neill et al.'s *pfx* folder [22]. To model mixture behavior, the code further defines a `departure_JSON` block which implements Gaussian + Exponential departure functions for helium-neon interactions. These functions are based on fitted coefficients for residual Helmholtz energy terms, allowing accurate modeling of mixture non-idealities. Additionally, the standard Lorentz-Berthelot mixing rules were applied to define binary interaction parameters (BIPs), ensuring thermodynamic consistency.

The departure function for the Helmholtz residual term is structured as follows:

$$\alpha^{\text{departure}} = \sum_i n_i \cdot \delta^{d_i} \cdot \tau^{t_i} \cdot \exp \left(-\eta_i \cdot \delta - \beta_i \cdot \tau - \gamma_i \cdot (\delta - 1)^2 - \epsilon_i \cdot (\tau - 1)^2 \right) \quad (3.1)$$

where $\delta = \rho/\rho_{\text{ref}}$ and $\tau = T_{\text{ref}}/T$, with n_i , d_i , t_i , η_i , β_i , γ_i , and ϵ_i being empirical coefficients derived from the Tkaczuk dataset [50].

Once the JSON files are registered, the helium-neon mixture can be instantiated via the *HeliumNeon* class, which acts as a wrapper around a *CoolProp.AbstractState* object. Mole fractions are set explicitly in the constructor, allowing flexibility for sensitivity studies on composition effects. This setup ensures that all downstream property calls (e.g., for viscosity, enthalpy, or thermal conductivity) automatically access the correct mixture properties during PFHX simulation.

Fluid Initialization and Interaction: The helium-neon mixture is initialized within the PFHX simulation via the `model()` function in *model.py*. The function begins by reading a configuration dictionary, which includes the coolant type, its composition, and process conditions. If the coolant is specified as "HeliumNeon", the simulation first calls the `setup_CoolProp()` function from *helium_neon.py* to register helium and neon as custom fluids in CoolProp. This step also loads the custom binary interaction parameters and departure function coefficients defined in the JSON files.

Listing 3.1: Coolant setup in *model.py*

```
1 if coolant_name == 'HeliumNeon':
2     helium_neon.setup_CoolProp(['Helium', 'Neon', 'Argon'])
3     helium_fraction = configuration['coolant']['helium_fraction']
4     coolant = fluids.HeliumNeon(coolant_mass_flowrate_kps, helium_fraction)
```

The `setup_CoolProp()` function in `helium_neon.py` activates CoolProp's custom fluid registration system using:

Listing 3.2: Custom fluid registration in `helium_neon.py`

```
1 CP.set_config_bool(CP.OVERWRITE_FLUIDS, True)
2 CP.set_config_bool(CP.OVERWRITE_BINARY_INTERACTION, True)
3 ...
4 CP.add_fluids_as_JSON('HEOS', json.dumps([contents]))
```

Once registered, the `HeliumNeon` object is created using the provided helium mole fraction. This class, defined in `fluids.py`, wraps a `CoolProp.AbstractState` for the "HEOS" backend, allowing property evaluation for the specified binary mixture. Internally, the constructor computes the neon mole fraction as the complement of helium and sets the composition via `set_mole_fractions()`:

Listing 3.3: Binary composition setting in `fluids.py`

```
1 self.AS = CP.AbstractState("HEOS", "Helium&Neon")
2 self.AS.set_mole_fractions([helium_fraction, 1 - helium_fraction])
```

This object is passed as the `coolant` parameter into the BVP solver. During each iteration in the `bvp_function()` in `bvp_model.py`, the coolant's pressure and temperature are updated based on the current node's process conditions. Then, `coolant.set_properties()` is called to refresh all relevant thermophysical properties:

Listing 3.4: Coolant update in `bvp_model.py`

```
1 coolant.update_conditions(T_coolant, P_coolant)
2 coolant.set_properties()
```

The `set_properties()` method queries CoolProp for viscosity, density, enthalpy, specific heat, and thermal conductivity, making these properties available for heat transfer and momentum balance calculations within the exchanger.

This dynamic updating approach ensures that coolant behavior evolves consistently with changing thermal gradients across the PFHX length. Furthermore, the mole fraction of helium is a configurable input parameter, enabling composition-based sensitivity analyses without requiring structural changes to the codebase. Nonetheless, the mixture will be kept at 80%-20% helium-neon respectively to enhance efficiency with the thermodynamical properties data from University of Stuttgart.

Thermophysical Properties Calculation: The helium-neon mixture's transport properties, viscosity and thermal conductivity, are essential for accurate modeling of heat transfer and pressure drop in the PFHX. These are computed dynamically during the simulation using CoolProp, based on the local pressure and temperature at each spatial node.

Within the `HeliumNeon` class in `fluids.py`, the method `set_properties()` calls CoolProp to update the properties of the binary mixture:

Listing 3.5: Transport property update in `fluids.py`

```
1 self.viscosity = self.AS.viscosity()
2 self.thermal_conductivity = self.AS.conductivity()
```

These methods rely on the Helmholtz energy formalism and the departure function defined by Tkaczuk et al. in the JSON mixture data [50]. The underlying correlations are parameterized as follows.

Viscosity:

$$\mu(T) = \mu_0 \cdot \exp \left(\sum_{i=0}^n b_i \cdot \log^i \left(\frac{T}{T_0} \right) \right) \quad (3.2)$$

Thermal Conductivity:

$$\lambda(T) = \lambda_0 \cdot \exp \left(\sum_{i=0}^n a_i \cdot \log^i \left(\frac{T}{T_0} \right) \right) \quad (3.3)$$

Here, T_0 is a reference temperature, and a_i, b_i are fitting coefficients obtained from experimental data. These expressions are embedded within CoolProp's backend and evaluated via the `AbstractState` object.

The transport properties are recomputed at every iteration step in the BVP solver, ensuring they are consistent with the evolving temperature and pressure conditions of the coolant stream. This approach provides physically meaningful coupling between thermodynamic state and heat exchanger performance.

Integration into the Simulation Loop: The helium-neon coolant is fully integrated into the PFHX simulation loop through its coupling with the BVP solver. At each iteration node, the model updates local temperature and pressure for the coolant, triggering a call to `set_properties()`. This ensures that heat transfer and pressure drop calculations are based on real-time conditions.

The implementation ensures thermodynamic consistency throughout the exchanger length. All relevant coolant-side properties, density, enthalpy, specific heat, viscosity, and thermal conductivity, are dynamically updated, enabling accurate performance prediction under varying load and boundary conditions.

Output Validation: The successful integration of the helium-neon mixture was validated through para-hydrogen conversion results. Simulation outputs show that conversion exceeds 99.0% at the PFHX outlet under nominal operating conditions. This indicates that the thermophysical behavior of the coolant was accurately captured and that the coupled solver performed as expected. The inlet conditions for both the coolant and the reactant H_2 gas are set equal to the working ASPEN HYSYS V14 process.

The result aligns with literature benchmarks and confirms the model's ability to predict spin-isomer conversion under realistic cryogenic conditions, further supporting its suitability for sizing and economic evaluation. Chapter 6 explains in more detail the output results of the PFHX simulation.

3.2. ASPEN HYSYS Process Simulation

This section outlines the methodological steps taken to improve the hydrogen liquefaction process through thermodynamic and process-level optimization. Building on an existing Aspen HYSYS model developed by Kerkar et al. [97], the objective was to address specific inefficiencies in the Brayton-based cryogenic cooling segment, where most energy consumption occurs. The study focuses on evaluating design alternatives to reduce specific energy consumption (SEC) and specific liquefaction cost (SLC), considering both capital and operational impacts. The final optimized design is benchmarked against the Claude cycle developed by Tamarona et al. [63], enabling a comparison between the two leading cryogenic configurations.

Starting Framework: The starting point for this analysis was the process simulation developed by Kerkar et al. [97], which implements a H_2 liquefaction plant in Aspen HYSYS V12. Notice Kerkar et al. used an older version of Aspen HYSYS, the reason why this thesis used the V14 version is explained in the Chapter 6. His work focused on a Brayton cycle-based configuration and included a preliminary techno-economic evaluation. However, the model exhibited various inefficiencies, particularly in the cryogenic cooling loop, which limited its overall performance and economic viability.

The baseline performance indicators from Kerkar et al. [97] Brayton cycle model are as follows:

- **Specific Energy Consumption (SEC):**
 - Without power recovery: 7.3187 kWh/kg_{LH₂}
 - With power recovery: 6.927 kWh/kg_{LH₂}
- **Specific Liquefaction Cost (SLC):**
 - With power recovery: €1.57/kg_{LH₂}

These values provide the baseline for evaluating techno-economical improvements, and they will serve as reference points when compared with the results of this thesis.

Objective of Optimization: The main goal of the process modifications is to improve the energy and economic performance of the H₂ liquefaction cycle. This is achieved by identifying inefficiencies in the baseline process, particularly within the Brayton cryogenic cooling segment, and introducing targeted modifications aimed at lowering CAPEX, OPEX, or both. All design changes are evaluated on the basis of their quantitative impact on SEC and SLC, ensuring that performance improvements are both thermodynamically and economically justified.

The diagram illustrates a hydrogen liquefaction process. It begins with a feed stream H1 entering a series of heat exchangers (HPC-1, HPC-2, HPC-3). The process then involves a series of heat exchangers (PFHX-1 to PFHX-5) and a reboiler (RE). The streams are then compressed (C-1 to C-5) and cooled (T-1, T-2, T-3) before being stored in tanks S1, S2, and S3. The final product is liquid H2, which is fed into FC1. The diagram is labeled with H1 through H10, MR1 through MR21, HENE1 through HENE6, and S1 through S3.

The hydrogen compression train includes three stages with intercooling and is followed by a multi-stage mixed refrigerant (MR) cycle for pre-cooling down to 78 K. Below this, the process relies on an open Brayton cycle operating with helium-neon for further cooling, with the final stage carried out via expansion and subcooling at cryogenic temperatures. In Kerkar et al. thesis, ortho-para conversion is not taken into account [97].

Tables 3.1 to 3.3 present the operating conditions of all major process streams in Kerkar et al. [97] H_2 liquefaction flowsheet. Each table lists the stream name, temperature, pressure, mass flow rate, and vapor quality at specific locations throughout the process.

Table 3.1 summarizes the hydrogen stream state points, covering the compression train, heat exchanger network, and final liquefaction stages. Table 3.2 provides the thermodynamic states of the helium-neon Brayton cycle refrigerant. Finally, Table 3.3 details the MR streams used for pre-cooling, including pressure staging and flash expansion segments. These data serve as a baseline for comparison with the optimized process configuration developed in this thesis.

Table 3.1: Hydrogen stream state points in Kerkar et al. baseline model [97].

Stream	Temp. [K]	Press. [bar]	Flowrate [kg/s]	Vapour Quality
H1	310	20	1	1
H2	310	80	1	1
H3	245	77.5	1	1
H4	145	76	1	1
H5	86	75.5	1	1
H6	38	75	1	1
H78	26.44	70	1	0.1551
H7	19.46	69.5	1	0.3085
H8	20	1	0.3834	1
H9	20	16	0.3834	1
H10	20	15	0.3834	1
H11	26.44	10.5	0.3834	0.196
H12	20	1	0.3834	1
Liquid-H2	20	1	1	0

Table 3.2: Helium-Neon stream state points in Kerkar et al. baseline model [97].

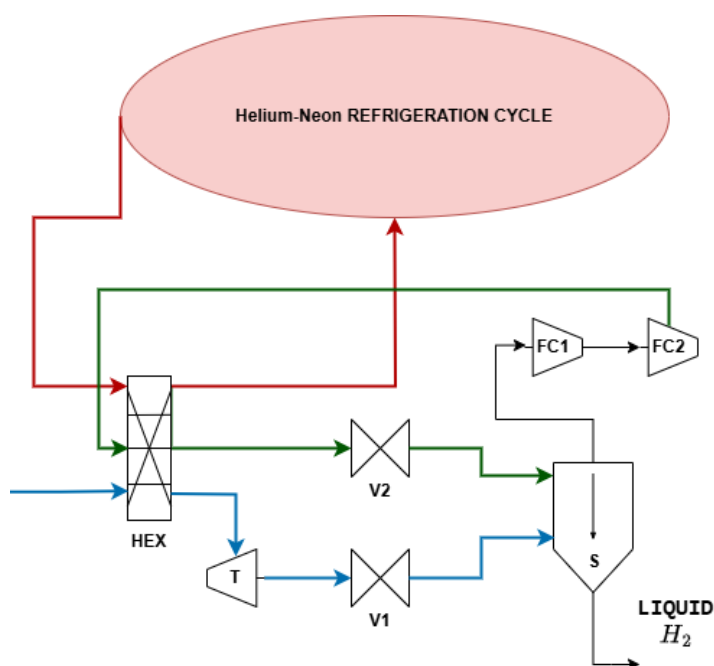
Stream	Temp. [K]	Press. [bar]	Flowrate [kg/s]	Vapour Quality
HENE1	310	73	9.7	1
HENE2	75	71.5	9.7	1
HENE3	41.84	13.3	9.7	1
HENE4	24.44	2.5	9.7	0.9807
HENE5	9.64	2.4	9.7	1
HENE6	310	2.4	9.7	1

One of the initial tasks involves modifying the existing Brayton cycle to simplify the final liquefaction stage. Specifically, towards the end of the current configuration, which can be seen in Figure 3.2 at cryogenic conditions, one turbine (T), two expansion valves (V1 & V2), and two flash compressors (FC1 & FC2) can be substituted.

After these components will be removed, two pathways are possible. The first one includes an ejector unit, while the second option aims to make a more efficient use of the excess H_2 gas. Both pathways have the intention to reduce both capital expenditure (CAPEX) and operational expenditure (OPEX). These reductions are expected to positively impact key performance indicators (KPIs) such as the Specific Liquefaction Cost (SLC) and the Specific Energy Consumption (SEC).

Table 3.3: Pre-cooling refrigerant stream state points in Kerkar et al. baseline model [97].

Stream	Temp. [K]	Press. [bar]	Flowrate [kg/s]	Vapour Quality
MR1	301.35	1.1	35	1
MR2	310	24.8	35	0.7197
MR3	310	24.8	20.656	1
MR4	255	24.4	20.656	0.716
MR5	255	24.4	12.4278	1
MR6	235	24	12.4278	0.9172
MR7	145	23.6	12.4278	0.3718
MR8	75	23.2	12.4278	0.0146
MR9	72	1.2	12.4278	0.0943
MR10	139.34	1.2	12.4278	0.6857
MR11	255	24.4	8.2278	0
MR12	235	24	8.2278	0
MR13	145	23.6	8.2278	0
MR14	144.44	1.2	8.2278	0.0301
MR15	142	1.2	20.656	0.4969
MR16	223.24	1.2	20.656	0.9402
MR17	310	24.8	14.344	1
MR18	255	24.4	14.344	0
MR19	235	1.2	14.344	0.1904
MR20	225.8	1.2	35	0.7397
MR21	235.7	1.1	35	0.8034

**Figure 3.2:** Current Configuration Brayton-Cycle Cryogenic Stage as per Kerkar et al. [97].

The ejector will utilize H₂ as the working fluid and will be modeled outside of Aspen HYSYS due to the lack of a native ejector unit operation in the software. The separate model, initially developed by Dr. Emanuele Zanetti, will serve as a base and be adapted to suit the process conditions of the H₂ Brayton cycle. Integration of this ejector into the overall process will involve thermodynamic consistency checks, performance benchmarking, and interface adjustments.

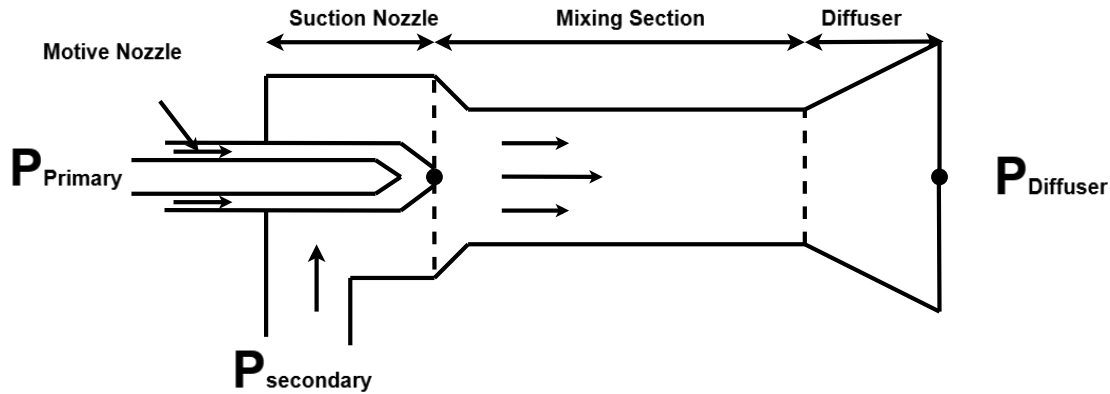


Figure 3.4: Schematic of Ejector Working Process, replicated from Liu et al. [98]

The ejector operates by using high-pressure flow from the primary inlet through a motive nozzle (H7), where the fluid undergoes isentropic expansion, converting pressure energy into kinetic energy. This high-velocity jet creates a low-pressure region at the entrance of the suction nozzle, which draws in a secondary, lower-pressure fluid from the secondary inlet (H12). Both streams then enter the mixing section, where momentum exchange occurs, and the flows merge into a single high-speed stream, reaching over Mach 1. This mixture is then decelerated in the diffuser, where part of the kinetic energy is recovered as static pressure, resulting in a mixed flow that exits at an intermediate pressure at the diffuser outlet (H8). This process enables compression of the secondary stream without mechanical work, relying solely on the energy of the motive flow.

Table 3.4: Operating conditions of streams H-7 and H-12.

Stream	Temp. (K)	Pressure (bar)	Mass Flow (kg s^{-1})	Vapour Quality (-)
Primary (H-7)	33.0	73.8	1	1
Secondary (H-12)	25.95	4.0	0.5403	0

Table 3.4 shows the working boundary conditions at the inlet of the ejector. These state conditions have been selected from Kerkar et al. previous work [97]. The $T_{\text{secondary}}$ has been calculated in the MATLAB code using the *refpropm* function calling REFPROP databank of NIST [99]. On the other hand, the mass flow rate of the secondary stream is derived after being iteratively adjusted based on vapor quality at the ejector outlet to satisfy energy and efficiency constraints. As can be seen, having 0.5403 kg s^{-1} of H_2 is almost a 41% increase compared to Kerkar et al. [97] flow rate of 0.3834 kg s^{-1} , meaning that less H_2 is being liquefied per second. As a result, the He–Ne cooling capacity will be insufficient, leading to both a proportional increase in the total compressor work, see Equation 2.9, and OPEX. This is explained in detail in Chapter 6.

To better understand this trade-off, the performance of the ejector was theoretically modeled in MATLAB using the framework established by Elbel et al. and Liu et al. [98, 100]. Two key metrics were considered:

$$\eta_{\text{ejector}} = \phi \cdot \frac{h'_{\text{s,isen}} - h_{\text{s}}}{h_{\text{p}} - h'_{\text{p,isen}}} \quad (3.4)$$

$$\phi = \frac{\dot{m}_{\text{s}}}{\dot{m}_{\text{p}}} \quad (3.5)$$

Here, ϕ is the entrainment ratio, h_p and h_s are the specific enthalpies of the primary motive and secondary suction streams, and h'_{isen} denotes isentropic enthalpies evaluated at the ejector outlet pressure. These equations were implemented in a MATLAB model used to simulate ejector performance across a range of operating conditions.

Typical efficiency values range below 20%, while entrainment ratio fluctuates around 0.5 as found by Elbel et al. [100] experimental data, with generally lower efficiency for a higher entrainment ratio. Nonetheless, Nakagawa et al. [101] conducted an experimental study on ejector geometry, revealing that ejector efficiency reaches a peak under high-pressure conditions, with efficiencies as high as 23%. This peak efficiency is influenced by factors such as the geometric configuration of the ejector, the temperature of evaporation, and the temperature of the gas cooler outlet. For the purpose of this investigation, the ejector target efficiency was chosen to be 20%, with a calculated entrainment ratio of 0.5403.

The ejector-based design was ultimately not selected for the final configuration due to an expensive OPEX. However, the insights gained from this research highlight the performance limitations of ejectors at cryogenic temperature hydrogen liquefaction environments. The results are further analyzed in Chapter 6 to better understand how the secondary inlet pressure affects the amount of H_2 being recirculated back, as well as the relationship between excess H_2 gas and the He–Ne cooling capacity, where trade-offs are quantified.

3.2.2. Final Process

Following the limitations observed in the ejector-based configuration, the final liquefaction process adopts a recirculation approach that enhances thermal integration and simplifies the cryogenic refrigeration loop. The resulting configuration, shown in Figure 3.5, builds upon the Brayton cycle architecture but incorporates strategic elements from Claude cycles, leading to a high-pressure H_2 liquefaction system best described as a Hybrid Brayton–Claude cycle.

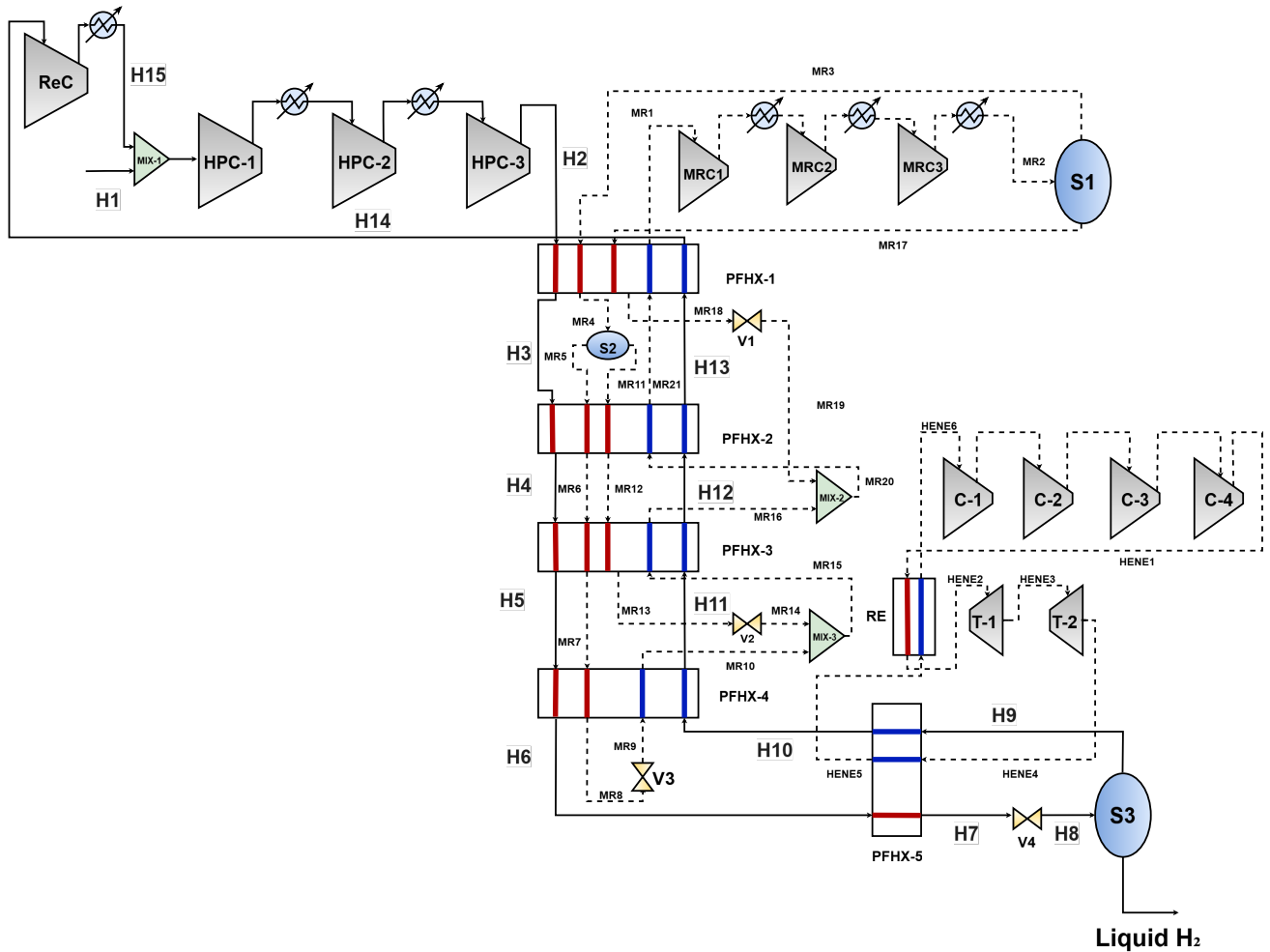


Figure 3.5: High-Pressure H₂ Liquefaction PFD Based on a Hybrid Brayton-Claude Cycle

Compared with the original design by Kerkar et al. [97], the cryogenic segment in this configuration has been significantly streamlined. The turbine, one expansion valve, and two flash compressors previously required to recompress and re-expand cold hydrogen have been eliminated. This simplification not only reduces CAPEX by removing critical rotating equipment and control elements, but also reduces OPEX by allowing a substantial decrease in the mass flow rate of the He–Ne coolant, from 9.7 kg s^{-1} in the original model to 6.9 kg s^{-1} in the final design.

The key innovation lies in the treatment of excess cold hydrogen (stream H-9), which exits the final separator (S3) at 20.2 K. Rather than being directly recompressed and looped through a dedicated path, this stream is redirected to pass sequentially through all PFHX units as a cold feed stream, recovering its residual cooling potential. Once completed in this internal heat exchange cascade, the stream is compressed from 1 bar to 21 bar and heated back to 310 K, aligned with the inlet conditions of the main hydrogen feed. Lastly, using a mixer (MIX-1), the main feed (H1) and the recirculation stream (H15) blend together before starting the liquefaction journey to 20.2 K.

This configuration provides three core benefits:

- **Component minimization** in the cryogenic zone, lowering CAPEX.
- **Reduced coolant flow rate**, decreasing Brayton cycle compressor work and hence OPEX.
- **Improved liquefaction performance**, as H₂ is now fully liquefied at high pressure: stream H-7 is entirely in the liquid phase at 73.8 bar.

The percentage of excess gas requiring recirculation also decreases from 33.14% in the baseline case

to 32.65% in the final configuration, yielding a modest improvement in net liquefaction rate. It is important to note that all MR compositions have been kept constant throughout this study; optimization of MR design was outside the scope of this thesis.

Table 3.5: Mixed Refrigerant Composition, Kerkar et al. [97].

Component	Mole Fraction [%]
Pentane	12.70
Butane	7.50
Propane	20.30
Ethylene	12.70
Ethane	9.50
Methane	17.50
Nitrogen	16.50
Neon	3.30

Process Description: The MR in the pre-cooling cycle is compressed to a pressure of 24.8 bar using three compressors: MRC1, MRC2, and MRC3. The flow rate required to achieve the cooling duty is 35 kg/s, which is used to lower the H₂ gas temperature to 74.15 K and 75 bar in the stream H6. Within the pre-cooling cycle, the refrigerant in the MR2 stream exists in both vapor and liquid phases. This mixture is sent to a separator, where MR3 is in the vapor phase and MR17 is in the liquid phase. To meet the cooling demands, four heat exchangers are employed: PFHX1, PFHX2, PFHX3 and PFHX4. The stream data for both the cooled hydrogen gas and the mixed refrigerant used in the cooling process are provided in Table 3.6 and Table 3.8.

Once the H₂ gas has been precooled to 74.15 K and 75 bar by the pre-cooling cycle, it enters the second refrigeration cycle for cryogenic liquefaction, which is based on the Brayton cycle concept. The cryogenic cycle uses a helium-neon mixture (Nelium) as the refrigerant, with a mole fraction distribution of 80% helium and 20% neon. This Nelium refrigerant is compressed from 3 bar to 60 bar by a four-unit intercooler compression system consisting of compressors C-1, C-2, C-3, and C-4. A recuperator is used to cool the compressed refrigerant from 310 K to 74.95 K by utilizing the cooling duty of the incoming Nelium refrigerant stream (HENE5), which has already been used to cool the hydrogen stream (H6) for cryogenic liquefaction.

The H₂ gas stream in H6, initially at 74.15 K, is cooled to 27.95 K in H7 using PFHX5. Both the cooling duty of the refrigerant (HENE4) and the recirculation stream of cold H₂ gas (H9) help to reach this low temperature; here it liquefies completely. After reaching this temperature, H7 is expanded using an isenthalpic valve reaching an atmospheric pressure of 1 bar for final storage purposes. The vapor quality of stream H8 is 0.3265, which is then recycled through all the PFHXs, making use of its cooling duty and being fed back into the initial loop; ultimately achieving the desired output of 86 TPD of liquid H₂.

Process Simulation: As discussed in subsection 2.2.4, recent conceptual studies have utilized commercial process simulation tools to evaluate plant operations in steady state conditions and, in some cases, estimate the total cost of the plant under varying scenarios. According to Al Ghafri et al. [18], the precision of the models used to represent the properties of H₂ and refrigerant fluids, as well as the design constraints imposed on key process equipment, such as heat exchangers, turbo-expanders, and compressors, is of high importance. This thesis makes use of steady-state process simulator Aspen HYSYS V14 process modeling.

Within the process simulation, Aspen HYSYS's built-in unit operation models are used to calculate the operation of components such as heat exchangers, compressors, and expanders. For the calculation of the multi-stream counter-current plate-fin heat exchanger (PFHX) in the hydrogen liquefaction process model, the built-in "LNG exchanger" model from Aspen HYSYS is utilized.

In addition, in the initial phase of modeling the liquefaction process, the efficiencies of the pressure-altering equipment are assumed. Specifically, the isentropic efficiencies of all compressors in the pre-

cooling cycles are assumed to be 85%, while compressors in the cryogenic cycle have polytropic efficiencies between 81.86 and 86.61%. Cryogenic turbo-expanders are assumed to have an isentropic efficiency between 87.4 and 90.0%. These efficiency values have been refined on the basis of the preliminary design outcomes and subsequently reincorporated into the simulation. Likewise, pressure drops associated with adsorbers and heat exchangers, including aftercoolers, which are initially considered negligible, have been adjusted and reintroduced into the simulation once preliminary equipment designs have been established.

Table 3.6: H₂ Stream State Points

Stream	Temp. [K]	Press. [bar]	Flowrate [kg s ⁻¹]	Vapour Quality [-]
H1	310	21	1	1
H2	310	80	1.4847	1
H3	255	78.7	1.4847	1
H4	235	77.5	1.4847	1
H5	144.95	76.2	1.4847	1
H6	74.15	75	1.4847	1
H7	27.95	73.8	1.4847	0
H8	20.2	1	1.4847	0.3265
H9	20.2	1	0.4847	1
H10	46.65	1	0.4847	1
H11	142.15	1	0.4847	1
H12	180	1	0.4847	1
H13	195	1	0.4847	1
H14	210	1	0.4847	1
H15	310	21	0.4847	1
Liquid-H ₂	20.2	1	1	0

Table 3.7: Helium-Neon Stream State Points

Stream	Temp. [K]	Press. [bar]	Flowrate [kg/s]	Vapour Quality
HENE1	310	60	6.9	1
HENE2	78.65	60	6.9	1
HENE3	46.75	13.3	6.9	1
HENE4	27.45	3.0	6.9	1
HENE5	74.05	3.0	6.9	1
HENE6	310	3.0	6.9	1

Table 3.8: Pre-cooling MR Stream State Points

Stream	Temp. [K]	Press. [bar]	Flowrate [kg/s]	Vapour Quality
MR1	288.2	1.1	35	1
MR2	310	24.8	35	0.7150
MR3	310	24.8	20.4583	1
MR4	254.30	24.4	20.4583	0.716
MR5	255	24.4	12.3083	1
MR6	235	24	12.3083	0.9172
MR7	144.95	23.6	12.3083	0.3718
MR8	74.95	23.2	12.3083	0.0146
MR9	71.85	1.2	12.3083	0.0943
MR10	139.35	1.2	12.3803	0.6857
MR11	255	24.4	8.1500	0
MR12	235	24	8.1500	0
MR13	144.95	23.6	8.1500	0
MR14	144.45	1.2	8.1500	0.0301
MR15	141.95	1.2	20.4583	0.4969
MR16	224.65	1.2	20.4583	0.9449
MR17	310	24.8	14.5417	1
MR18	255	24.4	14.5417	0
MR19	234.80	1.2	14.5417	0.1904
MR20	226.52	1.2	35	0.7401
MR21	235.21	1.1	35	0.7975

Fluid Properties Estimation: Since Aspen HYSYS is able to interface with the REFPROP database from NIST [99], the thermodynamic properties of normal, ortho, and para- H_2 can be accurately estimated using the state-of-the-art EOS developed by Leachman et al. [37]. After being cooled in the heat exchangers with continuous catalytic ortho-para- H_2 conversion, the H_2 feed stream, can be modeled assuming equilibrium- H_2 as described by Tamarona et al. [63]. For this purpose, the pseudo-equilibrium- H_2 heat capacity model developed by Valenti et al. [102] is utilized. The file can be seen in Appendix A.

The operational characteristics of gaseous H_2 entering the liquefaction plant at 21 bar, along with the properties of the resultant liquid product upon exiting the plant, are evaluated using the pseudo-equilibrium- H_2 model. Furthermore, the thermodynamic properties of the MR fluids in the pre-cooling and cryogenic liquefaction Brayton cycles are computed using the Peng-Robinson cubic equation of state, as implemented in Aspen HYSYS.

Calculation of Specific Energy Consumption The specific energy consumption (SEC) for the large-scale hydrogen liquefaction process is determined within the process simulation model using Aspen HYSYS's built-in spreadsheet functionality. The calculation is based on Equation 2.2. In this study, two different SEC values are evaluated: SEC with electric power recovery (SEC_{recov}) and SEC without power recovery from turbines (SEC_{base}). This comparison is made to assess the economic benefits of integrating turbine power recovery systems (turbo-generators) into the liquefaction process.

For SEC_{base} , the total power requirement is the sum of the electrical power required to operate the compressors within the liquefaction plant. In contrast, for SEC_{recov} , the net power requirement is calculated as the difference between the total electrical power consumption and the electrical power generated by turbo-generators installed in the plant. In this study, only turbo-expanders producing mechanical power greater than 100 kW are considered suitable for coupling with generators. The efficiency of converting mechanical energy from turbines to electrical energy is assumed to be 80%.

For research purposes, the primary reference process is the large-scale H_2 liquefier using mixed-refrigerant pre-cooling, developed by Berstad et al. [59], with a capacity of 86 TPD. The specific liquefaction power of this plant is calculated to be 3.014 kWh/kg $_{LH_2}$, while the SEC of the plant is

24.2 MW, or 6.723 kWh/kg_{LH₂}, when considering power recovery from turbine generators. Without turbine power recovery, the SEC value increases to 6.983 kWh/kg_{LH₂}. The exergy efficiency of the reported plant is 44.831%, based on the electric power recovery calculated using Equation 3.6.

$$\text{FOM} = \frac{w_{\text{ideal}}}{\frac{\dot{W}}{\dot{m}_f}} \quad (3.6)$$

4

Sizing of Preliminary Equipment

To evaluate the technical feasibility of the conceptual hydrogen liquefaction process developed in this work, preliminary sizing of major equipment is conducted. While detailed mechanical design and cost optimization are beyond the scope of this study, preliminary sizing ensures that all unit operations are physically realizable and appropriately integrated into the system. This step bridges process modeling and techno-economic evaluation by providing the necessary inputs, such as heat exchange areas, compressor stages, and turbine work outputs, for capital cost estimation. Unlike traditional manual design methods, this thesis leverages and adapts Python based computational frameworks developed by Tamarona, enabling a more efficient yet robust approach to equipment sizing. The methodology follows industry-relevant thermodynamic and empirical design criteria, and aligns with the assumptions applied in the process simulation and economic analysis chapters.

As mentioned in Section 2.3, the components analyzed include plate-fin heat exchangers (PFHX), compressors, and turbines, each essential to the reliable operation of the hydrogen Brayton-based liquefaction cycle. The pre-existing Python codes for compressor and turbine sizing were modified to incorporate state property calculations using REFPROP, employing the Peng–Robinson equation of state. The source code for the program is available on GitHub at: <https://github.com/pbtamarona/h2liquefaction>.

4.1. Plate-Fin Heat Exchanger Sizing

The preliminary design and sizing of the plate-fin heat exchangers (PFHXs) used in this study were carried out using Aspen EDR. Input parameters for the design were directly transferred from Aspen Hysys files which accurately recreated the PFHXs used in the H₂ liquefaction plant. Ortho-para H₂ conversion was not included in the sizing of the first 4 PFHXs, while for PFHX5, the length of the heat exchanger was selected based on the code developed by O'Neill et al. [22]. It is important to mention that the code initially supported only single-component cold streams. After the implementation of the He-Ne, multi-component streams were also supported. Nonetheless, the code focused only on the very final PFHX; where the previous heat exchangers used mixed refrigerants as cold stream. As a result, the model could not be applied to the first 4 PFHX. Moreover, the REFPROP interface lacks the required syntax to extract latent heat of vaporization data, which limits integration of phase change heat effects.

Although the ortho-to-para H₂ spin isomer conversion is essential to accelerate the liquefaction process, it is an exothermic reaction and thus contributes additional thermal load. This would require a higher cooling duty for the cold streams and consequently influence the design and sizing of the PFHX. However, as the excess p-H₂ gas stream (H9) is circulated back to the beginning of the liquefaction cycle as a cooling cold stream through all the PFHXs, its temperature increases, and the H₂ shifts towards to o-H₂, an endothermic process. This was proven by employing the pseudo-equilibrium-H₂ heat capacity model developed by Valenti et al. [102]. Subsequently, the entire liquefaction plant was evaluated running on n-H₂ and Aspen HYSYS gave no output errors. The pseudo-e-H₂ model proposed by Valenti et al. [102] is implemented by modifying the REFPROP fluid definition file (.FLD) for parahydrogen and

then accessing REFPROP's parahydrogen model directly within Aspen HYSYS [99]. It is important to mention that also in this reverted process, the catalysts are essential to favor the conversion, otherwise the process would require significantly more time.

Within Aspen EDR's design environment, a thorough evaluation of thermal, hydraulic, and mechanical aspects is performed. The software also reports detailed thermal and flow properties, including wall temperatures at each calculation node, which are used to verify compliance with design constraints. The key results from the preliminary design of the PFHX, as evaluated in Aspen EDR, are presented in Figure 4.1 and meet the design criteria summarized in Table 4.1.

Table 4.1: Design Limitations of plate-fin heat exchangers [63]

Parameter	Value	Unit
Max. Length	8.2	m
Max. Width	1.5	m
Max. Height	3.0	m
Volume	15–30	m ³
Specific Surface	500–1,800	m ² m ⁻³
Max. Temperature Difference	25.0	K
Min. Temperature Difference	1.00	K

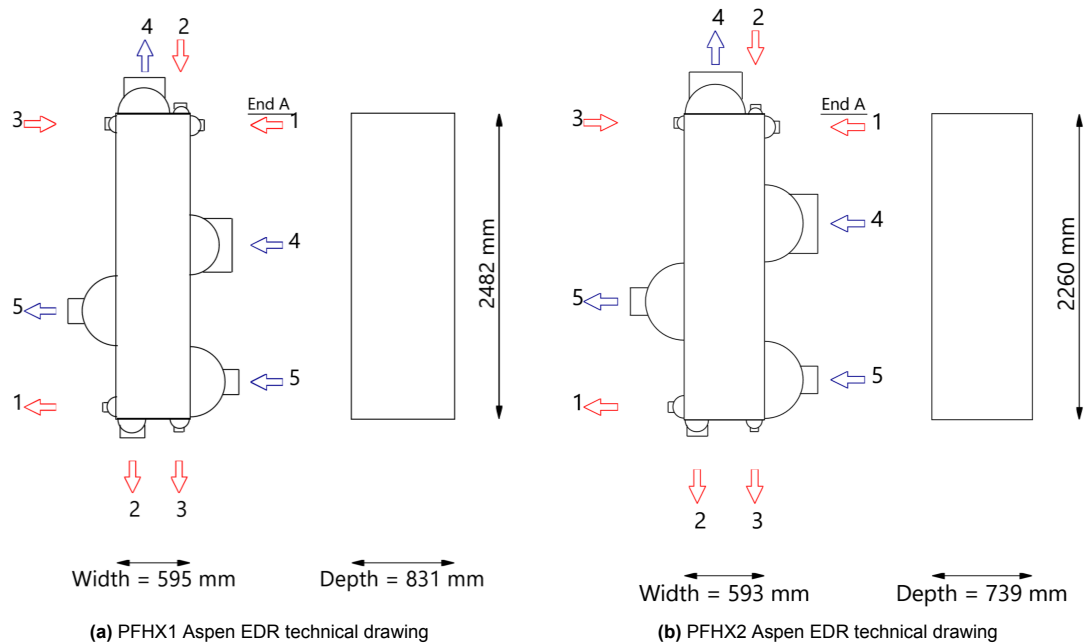


Figure 4.1: PFHX1 & PFHX2 Aspen EDR technical design results

4.2. Compressor Sizing

Based on the process simulation conducted in Aspen HYSYS, the inlet volumetric flow rates across the hydrogen feed, precooling refrigerant, and cryogenic liquefaction cycle compressor units were found to span from 2,200 to 73,000 m³ h⁻¹, with maximum discharge pressures reaching up to 80 bar. Centrifugal compressors were selected for all stages of compression due to their suitability within this operational range. To ensure thermal regulation, each centrifugal compressor is equipped with an aftercooler designed to reduce the discharge gas temperature to 310 K using cooling water.

The initial sizing of these compressors was carried out using Tamarona's GitHub repository mentioned above, as well as Kerkar et al. [97] work to adapt it to the Brayton cycle. Modifications were made to the

code to integrate the current state conditions from the Aspen HYSYS. The primary design constraints used in this preliminary sizing are summarized in Table 4.2.

Table 4.2: Design Limitations of Centrifugal Compressor [63]

Parameter	Value	Unit
Max. Number of Stages per Compressor	8	stages
Min. Volumetric Inlet Flowrate	1,000	$\text{m}^3 \text{h}^{-1}$
Max. Volumetric Inlet Flowrate	300,000	$\text{m}^3 \text{h}^{-1}$
Max. Compressor Power	37,000	kW
Max. Impeller Tip Velocity	500	m s^{-1}

4.2.1. Compressors in the Liquefaction Plant

To determine the appropriate number of stages and rotational speed (RPM) for the turbo-compressors, n – z (rotational speed–number of stages) diagrams are utilized. Figure 4.3, Figure 4.5, and Figure 4.7 illustrate these diagrams, which were generated using a custom Python script developed for the three compressor units in the liquefaction plant. For an in-depth explanation of the methodology and code used in the preliminary design and selection of compressors and turbines, Tamarona et al. research paper provides this [63].

Hydrogen Feed Compression Unit: In the liquefaction process, H_2 gas is compressed from 21 bar to 80 bar using three compressor units, with each unit's discharge subsequently cooled by aftercoolers, as illustrated in Figure 4.2.

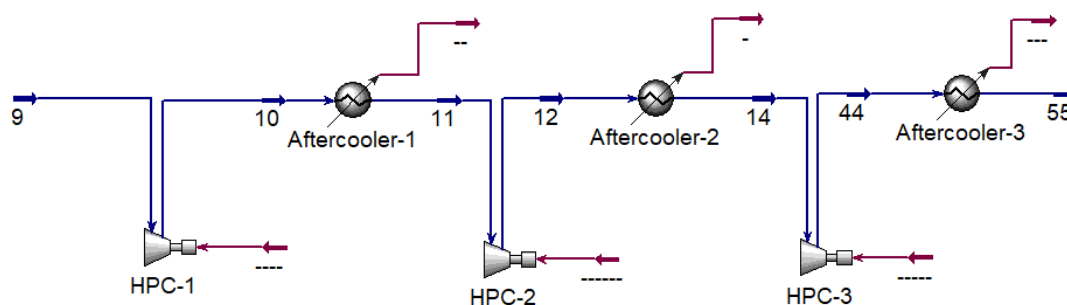


Figure 4.2: Aspen HYSYS modeling of feed compressor system.

This compression system operates with a pressure ratio of approximately four. The minimum number of stages for the centrifugal compressors is constrained by the maximum allowable impeller tip velocity, which is limited to 500 m s^{-1} in the base case scenario. Based on the Python-based design tool, six stages are selected for all the compressors.

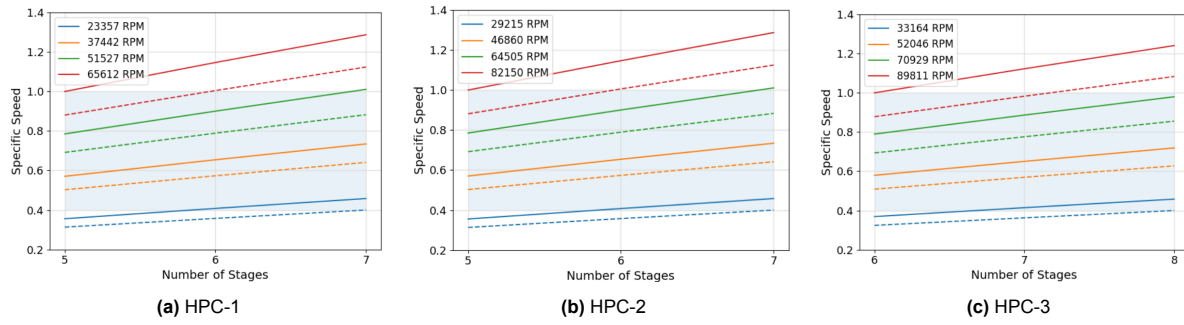


Figure 4.3: n-z diagrams for determining optimal number of stages and rotational speeds for HPC-1, HPC-2, and HPC-3.

MR Pre-Cooling Cycle Compression Unit: The mixed refrigerant, composed of eight different components, is pressurized from 1.1 bar to 24.8 bar through a series of three compressor units. Each compressor is designed to operate with two stages. Since the MR pre-cooling cycle was outside the scope of this thesis, the n-z plots, number of stages and RPM have not been changed from Kerkar et al. work [97].

Table 4.3: Mixed Refrigerant Compressors Design

Compressor	RPM	# of Stages
MRC1	4,200	2
MRC2	7,500	2
MRC3	12,500	2

Helium-Neon Cryogenic Liquefaction Cycle Compression Unit: Berstad et al. [59] reference plant described by Kerkar et al. [97], used a cryogenic cooling compression unit where the refrigerant is compressed from 2.4 bar to 73 bar using a single 15-stage compression system. In this study, after the optimization of the final liquefaction stage, and the reduction of the He-Ne coolant flow-rate, allowed for a smaller maximum discharge pressure in the cycle, with a final maximum pressure of 60 bar. Furthermore, the preliminary selection of thermodynamic conditions in the liquefaction stage provoked some liquid generation in Turbine-2 (T2); as a result, the outlet pressure has been increased from 2.4 bar to 3 bar. Since the outlet stream of the turbine circulates back into the first compressor of the helium-neon cycle, compressing from 3 bar to 60 bar allows for a reduction of the number of stages and thus, one less compressor, reducing CAPEX.

The compression system has been distributed across 4 separate compressors, where the first two use 4-stages, while the remaining two use three-stages. Figure 4.4 illustrates the first out of the four compressors, operating at a rotational speed of 8250 RPM with 4 stages. All compressors are equipped with intercoolers, which have been implemented accordingly in the Aspen HYSYS simulation environment.

Hence, a total of 14 stages have been used in this compression cycle. To calculate this number, the following equation has been used [103].

$$r_s = \left(\frac{P_{out}}{P_{in}} \right)^{1/n} \implies R_{tot} = (r_s)^n \quad (1)$$

1. **From the old design:**

$$R_{tot,old} = \frac{73 \text{ bar}}{2.4 \text{ bar}} \approx 30.417, \quad N_{old} = 15,$$

$$r_s = (R_{tot,old})^{1/N_{old}} = 30.417^{1/15} \approx 1.256.$$

2. **For the new case:**

$$R_{tot,new} = \frac{60 \text{ bar}}{3 \text{ bar}} = 20, \quad N_{new} = \frac{\ln R_{tot,new}}{\ln r_s} = \frac{\ln 20}{\ln 1.256} \approx 13.2 \implies 14 \text{ stages (round up)}.$$

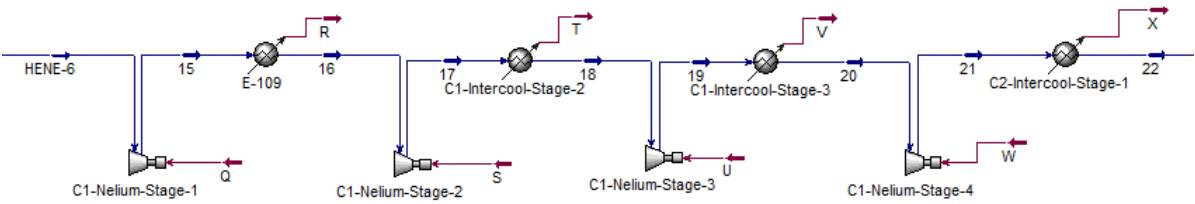


Figure 4.4: Aspen HYSYS modeling of feed compressor system

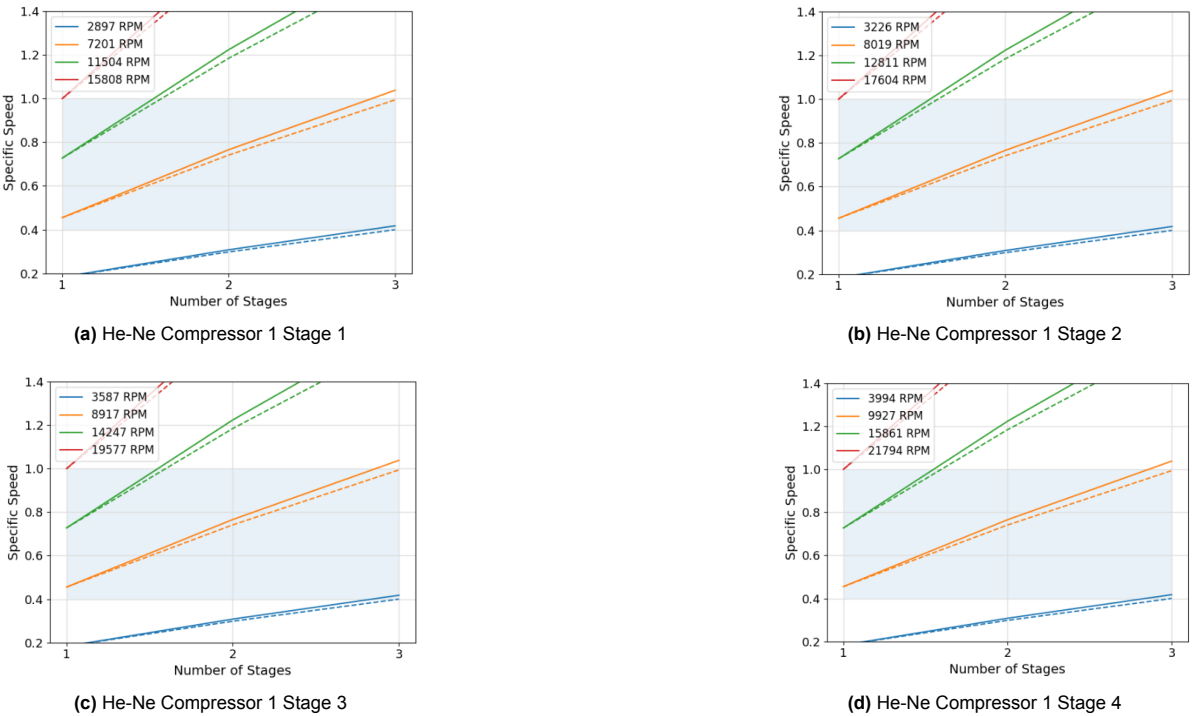


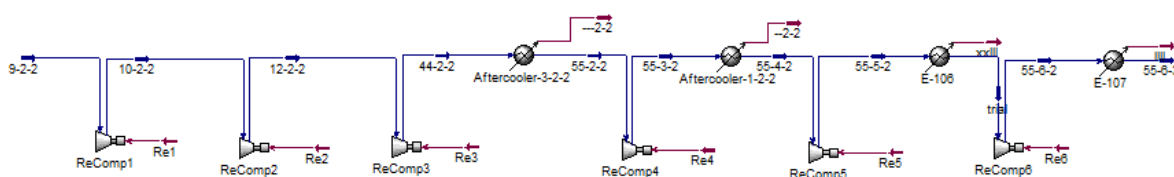
Figure 4.5: n–z diagrams for determining optimal number of stages and rotational speeds for Helium-Neon compressors.

Table 4.5 shows design parameters for each of the 4 multi-stages compression in the Helium-Neon cryogenic cycle. These values are lower than Kerkar et al. [97] because of a smaller mass-flow rate, as well as a lower maximum discharge pressure at the end of the cycle. Furthermore, an explanation on when a design limitation is suitable for a specific compressor will be given.

Table 4.4: Mixed Refrigerant Compressors Design

Compressor	RPM
He-Ne Compressor 1 Stage 1	8,250
He-Ne Compressor 1 Stage 2	8,250
He-Ne Compressor 1 Stage 3	8,250
He-Ne Compressor 1 Stage 4	9,500
He-Ne Compressor 2 Stage 1	9,500
He-Ne Compressor 2 Stage 2	9,500
He-Ne Compressor 2 Stage 3	9,500
He-Ne Compressor 2 Stage 4	9,500
He-Ne Compressor 3 Stage 1	9,500
He-Ne Compressor 3 Stage 2	9,500
He-Ne Compressor 3 Stage 3	9,500
He-Ne Compressor 4 Stage 1	20,000
He-Ne Compressor 4 Stage 2	20,000
He-Ne Compressor 4 Stage 3	20,000

Excess H₂ Recirculation Loop Compression Unit: As 32.65% of stream H8 is composed of very cold H₂ excess gas at 1 bar, its recirculation loop aims to mix it together with the initial feed stream H1. To do so, it must be under the same thermodynamic conditions. As a result, after making use of its cooling duty through all the PFHXs, it is compressed to 21 bar. 6 centrifugal compressors are needed to comply with compressor design limitations as well as maximum number of stages. Figure 4.6 shows the process simulation in Aspen HYSYS.

**Figure 4.6:** Excess H₂ Recirculation Compressors

As can be observed, only from the third compressor an aftercooler is required. This is due to the low inlet temperature of this compression system which is of 210K after going through all the heat exchangers. Figure 4.7 shows the n-z diagram for the first compressor.

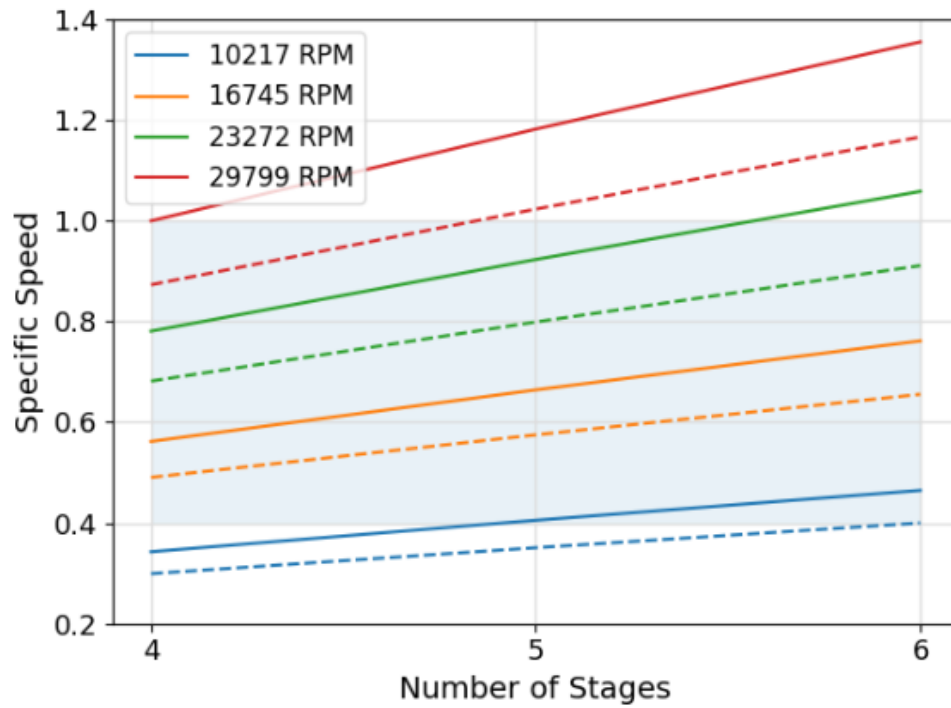
Figure 4.7: Excess H₂ Recirculation Compressor 1

Table 4.5: Mixed Refrigerant Compressors Design

Compressor	RPM	# of Stages
Recirculation Compressor 1	13,000	4
Recirculation Compressor 2	17,500	5
Recirculation Compressor 3	17,500	6
Recirculation Compressor 4	22,500	6
Recirculation Compressor 5	30,000	6
Recirculation Compressor 6	37,500	6

The preliminary design of centrifugal compressors is considered valid when the selected input parameters satisfy the defined constraints at all stages of the design process. Table 4.6 outlines the parameter limits applied for the multistage centrifugal compressor design. Although these constraints are not highly restrictive, they reflect recommended values intended to enhance efficiency. If certain values prove difficult to attain, a tolerance margin of up to 10% is permitted for specific parameters.

Table 4.6: Parameter constraints of centrifugal compressors [18]

Parameter	Range	Remark
Specific Speed, ω_s	0.4 – 1.0	
Flow coefficient, φ	0.2 – 0.3	
Work coefficient, ψ	0.5 – 0.6	
Degree of Reaction, R	0.6 – 0.7	10% tolerance
Rotor tip diameter ratio, δ_t	0.5 – 0.75	
δ_t/δ_h	0.2 – 0.7	
Rotor outlet absolute flow angle, α_2	60° – 70°	10% tolerance
Rotor outlet relative flow angle, β_2	0° – 60°	10% tolerance
Impeller tip speed velocity, u_2	< 500 m s ⁻¹	

4.3. Turbine Sizing

Compared to Kerkar et al. [97] work, this project uses two turbines instead of 3 as T-3 was removed from the final process due to the recirculation inefficiency. Furthermore, the issue with liquid generation within the turbine has been avoided since now the H_2 stream liquefies at high-pressures in PFHX5.

This thesis focuses exclusively on radial in-flow turbines, also known as centrifugal turbines, for the design of turbo-expanders used in the hydrogen liquefaction process. Compared to axial flow turbines, radial designs experience lower mechanical stresses at equivalent tip speeds, which enables operation at higher rotational velocities and consequently improves efficiency [104]. Although axial turbines may outperform in terms of efficiency at very high Reynolds numbers ($Re > 10^6$), they come with significantly higher capital costs [74]. Radial in-flow turbines are also known for their high reliability and low maintenance requirements.

Table 4.7: Design Limitations of radial-inflow turbines [63]

Parameter	Value	Unit
Max. Number of Stages per Turbine	1	stages
Min. Volumetric Inlet Flowrates	8.5	$m^3 h^{-1}$
Max. Volumetric Inlet Flowrates	339,800	$m^3 h^{-1}$
Max. Rotational Speed	100,000	rpm
Max. Turbine Power	1,500	
Max. Impeller Tip Velocity	500	$m s^{-1}$

The preliminary design of turbo-expanders in this study follows the same constraint-based methodology applied to compressors. The specific design limits adopted are outlined in Table 4.7. As shown in Figure 3.5, turbines T-1 and T-2 reach isentropic efficiencies of 87.38% and 90%, respectively. According to the n-z diagram illustrated in Figure 4.8, both turbines were configured with two stages. The parameter constraints guiding this preliminary design are summarized in Table 4.8. While these constraints are not strictly enforced, they represent best-practice values aimed at optimizing turbine performance. Where necessary, tolerance margins are permitted for certain parameters to accommodate practical design limitations.

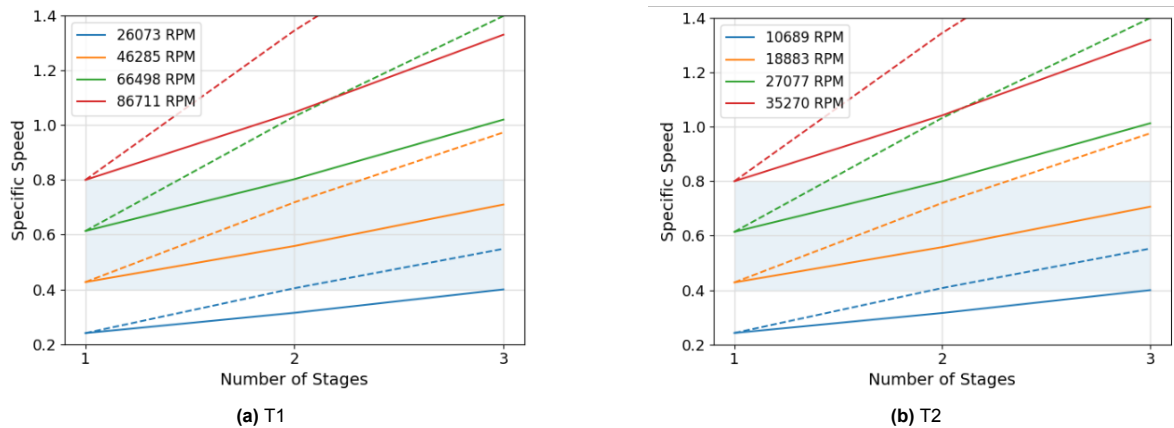


Figure 4.8: n-z diagrams for He-Ne turbines

Table 4.8 lists the constraints for the radial in-flow turbines parameters and their respective tolerances. All of these limits have been respected when developing the n-z diagrams.

Table 4.8: Parameter constraints of radial in-flow turbines [105]

Parameter	Range	Remark
Specific Speed, ω_s	0.4 – 0.8	
Flow coefficient, φ	0.2 – 0.3	25% tolerance
Work coefficient, ψ	0.8 – 1.0	20% tolerance
Degree of Reaction, R	0.45 – 0.65	15% tolerance
Rotor meridional velocity ratio, ζ	0.65 – 1.0	
Rotor tip diameter ratio, δ_t	< 0.7	10% tolerance
δ_t/δ_h	> 0.4	45% tolerance
Rotor inlet absolute flow angle, α_1	66° – 78°	
Rotor inlet relative flow angle, β_1	22° – 40°	
Impeller tip speed velocity, u_2	< 500 m s ⁻¹	

Techno-Economic Analysis

Techno-economic analysis (TEA) is a well-established approach to assess the economic viability of technological systems. In this thesis, a TEA is performed to estimate the specific liquefaction cost (SLC) of a large-scale H_2 liquefaction plant based on a Brayton cycle. The purpose of this analysis is to determine the economic value of liquid hydrogen by weighing the associated liquefaction costs against the expected benefits, using the hybrid Brayton-Claude cycle process described in Chapter 3.

To carry out this analysis, the Aspen Process Economic Analyzer (APEA) was employed. APEA was selected for its ability to generate equipment cost projections using up-to-date pricing data sourced from real-world EPC projects (Engineering-Procurement-Construction) and equipment suppliers [63]. Beyond estimating purchase costs, APEA also calculates total installed costs, accounting for both direct and indirect field expenses. In addition, Aspen HYSYS process simulations can be integrated with APEA to evaluate total capital investment, operating expenses, and utility consumption within the plant.

The following sections present the customized TEA workflow developed for this thesis, including detailed descriptions of general project specifications, equipment input data, and key process assumptions that form the basis for the economic evaluation of the H_2 liquefaction system.

5.1. APEA Workflow and Project Basis

The Aspen Process Economic Analyzer (APEA) facilitates the automated generation of detailed designs, cost estimates, investment analyses, and project timelines based on limited project scope information. This can include data from process simulations or a list of pre-sized equipment. The standard project workflow recommended for APEA is shown in Figure 5.1's left-hand-side. The tool allows users to revisit and revise previous steps to improve the accuracy of cost assessments.

In this thesis, the standard APEA workflow has been adapted to suit the specific needs of the H_2 liquefaction process under investigation. These modifications enable the calculation of the Specific Liquefaction Cost (SLC). The customized techno-economic analysis (TEA) workflow used for cost evaluation is presented in Figure 5.1.

5.1.1. Project Basis

The project basis establishes the overarching specifications for the entire project scenario, influencing both the design and cost estimation by setting system defaults and environmental parameters. In APEA, this includes key elements such as **Project Properties**, **General Project Data**, **Basis for Capital Cost**, **Investment Analysis**, **Process Design**, and **Streams**.

Project Properties and **General Project Data** allow users to define the project name, owner, unit system, country of reference, and currency used for cost evaluation. The choice of base country impacts several default parameters, including the cost index, labor wage rates, and costs related to engineering, construction management, and project oversight. For this thesis, the European Union (EU) is selected

as the project location, with the Euro (EUR) designated as the base currency.

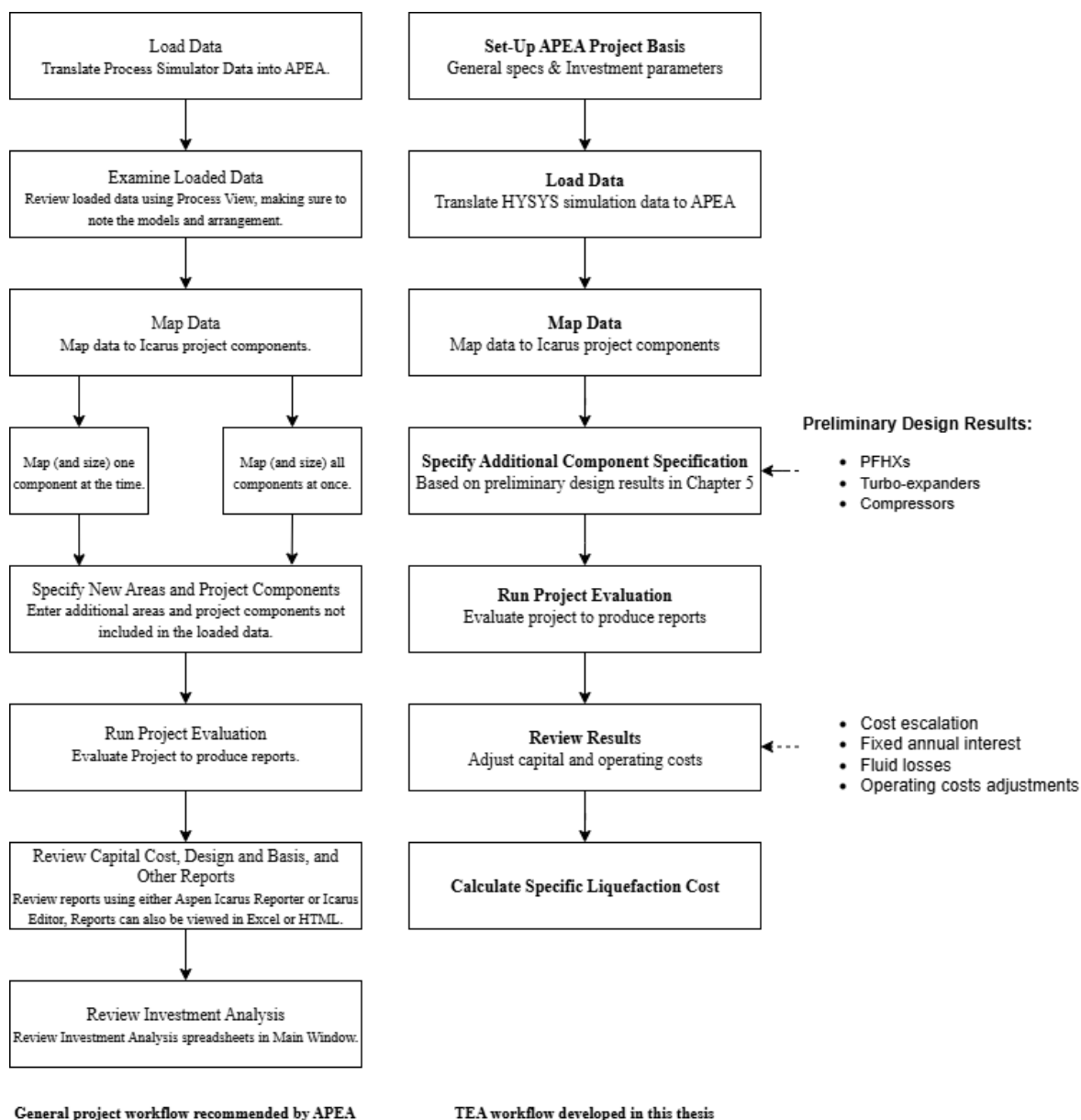


Figure 5.1: Techno-economic analysis workflow using APEA

In APEA projects that incorporate input from a process simulator, the **Process Design** specifications are used to map simulation equipment models to corresponding ICARUS project components. In this thesis, the Aspen HYSYS simulation developed in Chapter 3 and refined using the preliminary equipment sizing in Chapter 4 is imported directly into APEA's Process Design module.

Within the **Process Design** settings, the *Utility Specification* section defines the utilities used for process heating and cooling. This includes the details and unit cost of cooling water streams associated with the process aftercoolers. For both the simulation and the TEA model, cooling water is assumed to operate at a pressure of 3.45 bar, with inlet and outlet temperatures of 20° C and 35° C, respectively. The cooling water unit cost is established at $0.05 \times 10^{-4} \text{ €/m}^3$.

The **Streams** specification section in APEA allows users to create, modify, or delete material streams. However, in this case, no changes or additions are made to the streams imported from the Aspen HYSYS simulation.

Basis for Capital Costs: The **Basis for Capital Costs** section in APEA includes several configuration categories, such as *General Specs*, *Construction Workforce*, *Currency*, and *Input/Output Units of Measure Customization*. Among these, the **General Specs** have a major impact on both capital investment and operating cost estimates.

Within the *General Specs*, parameters like Process Description, Process Complexity, and Project Type are used to determine the contingency percentage applied to the total project cost. These variables interact in a non-linear way to produce the final contingency value. Additionally, the Process Description affects design allowances for equipment with system-generated material costs, while the Project Type influences the structure of the power distribution and process control systems for the plant. Furthermore, the Project Location setting also falls under *General Specs* and affects the estimation of domestic and international freight costs, taxes and duties (as a percentage of material cost), and rotating equipment spares (as a percentage of their purchase cost). Another important input in this section is the Estimated Starting Date of Basic Engineering, which defines the start of the project timeline, including design, procurement, delivery, site preparation, and construction phases. This date also determines whether capital cost escalation or de-escalation is applied, based on APEA's pricing basis.

In this thesis, APEA V14 was used, which is based on cost data from the first quarter of 2022. Capital cost escalation was manually applied using Equation 2.5.1 and the CEPCI indices for 2022 and 2024. Consequently, the Estimated Start Date for Basic Engineering was set to April 1st, 2022. Figure 5.2 shows the input values of APEA General Specs for the Base Scenario for this thesis.

Name	Item 1
Process Description	New process
Process Complexity	Typical
Process Control	Digital
PROJECT INFORMATION	
Project Location	Rotterdam
Project Type	Grass roots/Clear field
Contingency Percent	30
Estimated Start Day of Basic Engineering	1
Estimated Start Month of Basic Engineering	APR
Estimated Start Year of Basic Engineering	22
Soil Condition Around Site	SOFT CLAY
EQUIPMENT SPECIFICATION	
Pressure Vessel Design Code	ASME
Vessel Diameter Specification	ID
P and I Design Level	FULL

Figure 5.2: APEA General Specifications

Choosing "New Process" in the Process Description under General Specs, results in a 10% design allowance, with a contingency value as high as 30%.

Investment Analysis: Furthermore, APEA allows users to define *Construction Workforces*, which are categorized into General Rates and Craft Rates. When a country base is selected, APEA assigns default values for wage rates, crew sizes, productivity, and related parameters based on regional norms. Since this thesis does not involve a detailed labor analysis, the default workforce data for European projects, provided by APEA, are used.

The **Investment Analysis** section includes parameters such as *Investment Parameters*, *Operating Unit Costs*, *Raw Material Specifications*, and *Product Specifications*. However, since the main goal of this thesis is to estimate the Specific Liquefaction Cost (SLC) of a H₂ liquefaction plant, detailed data on raw materials and product characteristics, which are typically used for profitability or ROI analysis, are not relevant. Consequently, most investment parameters are omitted, except for the Operating Cost Parameters and Facility Operation Parameters, which are necessary to calculate the annual operating costs.

The Operating Cost Parameters include:

- Operating Charges, such as laboratory fees and operating supplies, are defined as a percentage of labor costs.
- Plant Overhead, covering services, facilities, payroll, and similar expenses, expressed as a percentage of operating labor and maintenance costs.
- G&A Expenses, representing general and administrative costs such as management salaries, R&D, and product distribution, also specified as a percentage of operating costs.

The Facility Operation Parameters include settings such as Facility Type, Operating Mode, Length of Start-Up Period, Operating Hours per Period, and Process Fluids. Relevant parameter values are illustrated in Figure 5.3.

Investment Analysis Parameters		
Name	Units	Item 1
Period Description		Year
Number of Weeks per Period	Weeks/period	52
Number of Periods for Analysis		20
Tax Rate	Percent/period	40
Interest Rate/Desired Rate of Return	Percent/period	20
Economic Life of Project	Period	10
Salvage Value (Percent of Initial Capital Cost)	Percent	20
Depreciation Method		Straight Line v
ESCALATION PARAMETERS		
Project Capital Escalation	Percent/period	5
Products Escalation	Percent/period	5
Raw Material Escalation	Percent/period	3,5
Operating and Maintenance Labor Escalation	Percent/period	3
Utilities Escalation	Percent/period	3
PROJECT CAPITAL PARAMETERS		
Working Capital Percentage	Percent/period	5
OPERATING COSTS PARAMETERS		
Operating Supplies	Cost/period	25
Laboratory Charges	Cost/period	25
Operating Charges	Percent/period	25
Plant Overhead	Percent/period	50
G and A Expenses	Percent/period	8
FACILITY OPERATION PARAMETERS		
Facility Type		Chemical Processing Facility v
Operating Mode		Continuous Processing - 24 Hou v
Length of Start-up Period	Weeks	20
Operating Hours per Period	Hours/period	8.322
Process Fluids		Liquids and Gases v

Figure 5.3: APEA Investment Analysis Parameters

The *Operating Unit Costs* section defines the unit labor costs for both operators and supervisors, as well as the costs of non-heat transfer utilities. The calculation of total labor cost involves three steps:

1. Estimating the number of operators and supervisors required to run the facility over a given duration.
2. Adjusting for the total operating hours within the defined period.
3. Multiplying the resulting labor hours by the respective unit costs and summing them.

Operator labor refers to all tasks directly involved in running the facility, while supervisor labor includes oversight and management of the operational staff. In this study, the unit labor costs for operators and supervisors are based on APEA's default values: 20 €/person·hour for operators and 35 /person·hour for supervisors.

The non-heat transfer utility costs include items such as electricity, potable water, fuel, and instrument air. However, only electricity costs are considered in this thesis, since the remaining utilities are not defined in the process simulation. For the base case, the electricity price is set at 0.1 €/kW h. A

sensitivity analysis on electricity price variations is conducted within the TEA, with results discussed in the following chapter.

5.2. Specifications of Mapped Equipment

Once the desired process components have been mapped, their specifications are established based on the process simulation results. Compressors, turbines, heat exchangers, and auxiliary components, are matched with its corresponding ICARUS component. These mappings are further refined using sizing results from preliminary equipment design to reflect realistic technical parameters. This step ensures that the cost estimations in APEA accurately represent the scale, configuration, and functionality of the H₂ liquefaction plant.

5.2.1. Compressors

In this study, compression systems modeled in Aspen HYSYS are mapped to the ICARUS Gas Centrifugal Compressor - Horizontal (DGC CENTRIF) model in APEA. Key input parameters for cost estimation, such as the inlet gas flow rate and design pressures, are automatically transferred from HYSYS. Although certain details like the number of impellers and rotational speed are not directly imported, they are determined based on the compressor design Python code. For instance, Figure 5.4 shows an example of a mapped centrifugal compressor to APEA.

HPC-1 - Centrifugal compressor - horizontal		
Name	Units	Item 1
Item Reference Number		3
Remarks 1		Equipment mapped from 'HPC-1'.
Remarks 2		
Item description		HPC-1
User tag number		GC3
Drawing reference number		
Structure tag		
Component WBS		
Quoted cost per item	EUR	
Currency unit for matl cost		
Source of quote		
Number of identical items		1
Installation option		
Code of account		
Icarus/User COA option		
Casing material		SS304
Actual gas flow rate Inlet	M3/H	3.622,525423
Design gauge pressure Inlet	KPAG	1.998,67108
Design temperature Inlet	DEG C	36,85
Design gauge pressure Outlet	KPAG	3.178,437076
Design temperature Outlet	DEG C	87,451896
Number of impellers		6
Compressor speed	RPM	35.000
Driver power	KW	1.116,304505
Molecular weight		2,01588
Specific heat ratio		1,385285
Compressibility factor Inlet		1,012084
Compressibility factor Outlet		1,017011
Intercooler required		NO
Intercooler type		WATER
Aftercooler required		YES
Aftercooler type		WATER
Inter/Aftercooler excess area	PERCENT	0
Maximum interstage temperature	DEG C	175
Intercooler outlet temperature	DEG C	30
Interstage pressure drop	KPAG	35
Driver type		MOTOR
Turbine gauge pressure	KPAG	2.050
Gear reducer type		
Lube oil system		YES
Equipment Footprint X	M	
Equipment Footprint Y	M	
Equipment Footprint Z	M	

Figure 5.4: HPC1 APEA specifications

Each compressor is equipped with an aftercooler to reduce the temperature of the outlet gas to 310K. Some compressors which require multi-stage compression system are also equipped with an intercooler for the same reason. In this work, the inter and aftercoolers are mapped in APEA by selecting Yes at each compressor specifications, intercoolers are only selected if required by the process design. APEA maps them by using the ICARUS TEMA Shell and Tube Heat Exchanger model (DHE TEMA EXCH). Furthermore, an important parameter is the Inter/Aftercooler excess area, expressed as a percent, however, since these aftercoolers were not part of the preliminary equipment design, their capital cost is estimated based on APEA's default sizing and design parameters. Once a component is mapped and specified in APEA, the item evaluation can be executed to produce a capital cost report, providing a direct cost estimate for that equipment.

5.2.2. Turbines

Expansion turbine models from Aspen HYSYS are mapped to the ICARUS Turbine Turbo-expander (DTURTURBOEXP) component in APEA. As with compressor mapping, key turbine specifications, such as inlet gas flow rate and gauge pressures at both inlet and outlet, are automatically filled based on HYSYS data. These inputs are essential for estimating the turbine's capital cost in APEA.

Turbine cost estimation can be refined by entering the inlet design temperature from the process specifications. Figure 5.5 illustrates an example of the two turbo-expanders specification interface in APEA.

TURBINE-1 - Turboexpander		
Name	Units	Item 1
Item Reference Number		25
Remarks 1		Equipment mapped from TURBINE-1'.
Remarks 2		
Item description		TURBINE-1
User tag number		TUR25
Drawing reference number		
Structure tag		
Component WBS		
Quoted cost per item	EUR	
Currency unit for matl cost		
Source of quote		
Number of identical items		1
Installation option		
Code of account		
Icarus/User COA option		
Actual gas flow rate Inlet	M3/H	394,521301
Design gauge pressure Inlet	KPAG	5,898,67108
Design temperature Inlet	DEG C	-194,5
Design gauge pressure Outlet	KPAG	1,228,67108
Power output	KW	579,183464
Molecular weight		7,238984
Specific heat ratio		1,592405
Compressibility factor Inlet		0,959135
Isentropic efficiency	PERCENT	87,381561
Number of spare cartridges		0
Equipment Footprint X	M	
Equipment Footprint Y	M	
Equipment Footprint Z	M	
Equipment Coordinate X	M	
Equipment Coordinate Y	M	
Equipment Coordinate Z	M	
Equipment Location Level		
Equipment Safety Dimension X	M	
Equipment Safety Dimension Y	M	
Equipment Safety Dimension Z	M	
Equipment Symbol Shape		
Allow resizing		

TURBINE-2 - Turboexpander		
Name	Units	Item 1
Item Reference Number		26
Remarks 1		Equipment mapped from TURBINE-2'.
Remarks 2		
Item description		TURBINE-2
User tag number		TUR26
Drawing reference number		
Structure tag		
Component WBS		
Quoted cost per item	EUR	
Currency unit for matl cost		
Source of quote		
Number of identical items		1
Installation option		
Code of account		
Icarus/User COA option		
Actual gas flow rate Inlet	M3/H	1,042,767912
Design gauge pressure Inlet	KPAG	1,228,67108
Design temperature Inlet	DEG C	-226,4
Design gauge pressure Outlet	KPAG	198,67108
Power output	KW	352,891727
Molecular weight		7,238984
Specific heat ratio		1,606103
Compressibility factor Inlet		0,944858
Isentropic efficiency	PERCENT	90
Number of spare cartridges		0
Equipment Footprint X	M	
Equipment Footprint Y	M	
Equipment Footprint Z	M	
Equipment Coordinate X	M	
Equipment Coordinate Y	M	
Equipment Coordinate Z	M	
Equipment Location Level		
Equipment Safety Dimension X	M	
Equipment Safety Dimension Y	M	
Equipment Safety Dimension Z	M	
Equipment Symbol Shape		
Allow resizing		

(a) APEA T1 component specs form

(b) APEA T2 component specs form

Figure 5.5: Turbines T1 & T2 specifications in APEA

As discussed in subsection 3.2.2, this study evaluates two specific energy consumption (SEC) values for the H₂ liquefier: one including power recovery from turbines (SEC_{recov}) and one without it (SEC_{base}). To calculate SEC_{recov}, the additional capital and operating expenses associated with power generators must be considered. Since generator models are not available in the HYSYS process simulation file, they are manually incorporated in APEA using the ICARUS project component *Electrical generator – Steam or gas turbine drive (EEG TURBO GEN)* for each turbo-generator unit. In the specification form, the driver type is set to "NONE" so that only the generator costs are accounted for, excluding turbine costs already modeled.

In this process, T1 and T2 operate on the same rotating shaft, thus a single generator is assigned to the system. This generator converts the combined mechanical output of both turbines into electrical power.

5.2.3. Plate-Fin Heat Exchangers

The LNG multi-stream heat exchanger models from Aspen HYSYS are mapped to the ICARUS Heat Exchanger, Plate Fin (DHE PLATE FIN) component in APEA. To estimate the direct cost of the plate-fin heat exchangers, APEA requires manual input of the PFHX core dimensions, as these are not automatically transferred during equipment mapping. Therefore, the core dimensions obtained from

the preliminary equipment sizing must be entered manually.

HX1 - Multi-stream, plate fin heat exchanger			HX2 - Multi-stream, plate fin heat exchanger		
Name	Units	Item 1	Name	Units	Item 1
Item Reference Number		6	Item Reference Number		7
Remarks 1		Equipment mapped from 'HX1'.	Remarks 1		Equipment mapped from 'HX2'.
Remarks 2			Remarks 2		
Item description		HX1	Item description		HX2
User tag number		HE6	User tag number		HE7
Drawing reference number			Drawing reference number		
Structure tag			Structure tag		
Component WBS			Component WBS		
Quoted cost per item	EUR		Quoted cost per item	EUR	
Currency unit for matl cost			Currency unit for matl cost		
Source of quote			Source of quote		
Number of identical items		7	Number of identical items		5
Installation option			Installation option		
Code of account			Code of account		
Icarus/User COA option			Icarus/User COA option		
Material		A3003	Material		A3003
Core length	M	2,482	Core length	M	2,26
Core width	M	0,595	Core width	M	0,593
Core height	M	0,831	Core height	M	0,739
Number of streams		5	Number of streams		5
Parting sheet thickness	MM	2	Parting sheet thickness	MM	2
Cap sheet thickness	MM	6,35	Cap sheet thickness	MM	6,35
End bar width	MM	38	End bar width	MM	38
Side bar width	MM	38	Side bar width	MM	38
Core weight	KG		Core weight	KG	
Pipe material		AL	Pipe material		AL
Layer data			Layer data		
Stream data			Stream data		
Equipment Footprint X	M		Equipment Footprint X	M	
Equipment Footprint Y	M		Equipment Footprint Y	M	
Equipment Footprint Z	M		Equipment Footprint Z	M	
Equipment Coordinate X	M		Equipment Coordinate X	M	
Equipment Coordinate Y	M		Equipment Coordinate Y	M	
Equipment Coordinate Z	M		Equipment Coordinate Z	M	
Equipment Location Level			Equipment Location Level		
Equipment Safety Dimension X	M		Equipment Safety Dimension X	M	
Equipment Safety Dimension Y	M		Equipment Safety Dimension Y	M	
Equipment Safety Dimension Z	M		Equipment Safety Dimension Z	M	
Equipment Symbol Shape			Equipment Symbol Shape		
Allow resizing			Allow resizing		

(a) PFHX1 APEA specifications

(b) PFHX2 APEA specifications

Figure 5.6: Plate-fin heat exchangers PFHX1 & PFHX2 specifications in APEA

For non-catalytic PFHX units, the cost estimate can be further refined by including additional parameters such as parting sheet thickness, cap sheet thickness, end bar width, side bar width, and core weight, based on results from Aspen EDR. In contrast, for catalytic PFHXs, only the core dimensions of the kinetic ortho-para conversion simulation are entered, while all other parameters are based on APEA's default values. Hence, as previously mentioned, since ortho-para conversion was only considered in PFHX5, only the length of this heat exchanger has been adjusted to the length needed to achieve 99.8% p-H₂. Figure 5.6 illustrate sample specification forms for noncatalytic and catalytic PFHXs, respectively. If multiple identical PFHX units are used in parallel, the total cost is determined by specifying the number of units within the APEA form.

5.2.4. Vapor-Liquid Separators

A total of three vapor-liquid separators are implemented in the H₂ liquefaction process in Aspen HYSYS; these are mapped to the ICARUS project as Vertical process vessel (DVT CYLINDER). As these components were not included in the sizing of preliminary equipment design, the default specifications from APEA are used.

5.3. Results and SLC Calculation

	A	B	C
1	EXECUTIVE SUMMARY		
2	=====		
3			
4	PROJECT NAME:	Final Analysis	
5			
6	CAPACITY:	ERR	
7			
8	PLANT LOCATION:	Rotterdam	
9			
10	BRIEF DESCRIPTION:		
13			
14	SCHEDULE:	=====	
15	Start Date for Engineering	1APR22	
16	Duration of EPC Phase	62,00	Weeks
17	Completion Date for Construction	Thursday, June 15, 2023	
18	Length of Start-up Period	20,00	Weeks
19			
20			
21	INVESTMENT:	=====	
22	Currency Conversion Rate	1,00	EUR/EUROPEAN EURO
23	Total Project Capital Cost	129.021.228,36	EUR
24	Total Operating Cost	28.074.908,63	EUR/Year
25	Total Raw Materials Cost	0,00	EUR/Year
26	Total Utilities Cost	21.473.839,14	EUR/Year
27	Total Product Sales	0,00	EUR/Year
28	Desired Rate of Return	20,00	Percent/Year
29	P.O. Period	0,00	Year
30			
31	PROJECT INFORMATION:	=====	
32	Simulator Type	Aspen HYSYS	

Figure 5.7: APEA executive summary report

Figure 5.7 presents a sample of the executive summary report generated by APEA upon completing the project evaluation. This summary highlights key outputs, specifically the Total Project Capital Cost and Total Operating Cost, which serve as the main inputs for calculating the Specific Liquefaction Cost (SLC) of the H₂ liquefaction plant.

Before performing the SLC calculation, adjustments to both the capital and operating costs reported by APEA are required. First, the total capital cost must be updated to include working capital, as APEA only factors this into its cash flow analysis during the commissioning stage. In this study, a working capital assumption of 10% of the total capital cost is applied.

Furthermore, the capital cost is subject to cost escalation. Since this thesis uses APEA Version 14, which is based on April 1st 2022 cost data, the reported capital cost reflects 2022 pricing. To align this with 2024 cost levels, an escalation is applied using Equation 2.5.1 and the CEPCI indices for 2022 (816.0) and April 1st 2024 (799.05) [106]. This adjustment ensures that the cost estimate reflects current market conditions. Nonetheless, to properly configure the CEPCI corrections, the analysis must be done in US\$. The following equation serves to adjust these costs to 2024 in € using both CEPCI and the exchange rates for the Start Date for Engineering [107].

$$f(C)_{\epsilon, 2024} = f(C)_{\epsilon, \text{base}} \cdot \left(\frac{\text{US\$}}{\epsilon} \right)_{\text{base}} \cdot \left(\frac{\text{CEPCI}_{2024}}{\text{CEPCI}_{\text{base}}} \right) \cdot \left(\frac{\epsilon}{\text{US\$}} \right)_{2024} \quad (5.1)$$

As defined in Equation Equation 5.1, the Specific Liquefaction Cost (SLC) calculation involves converting the total capital expenditure (CAPEX) into an equivalent annual cost. In this thesis, the capital

investment is treated as a fixed annuity over the project lifetime, representing the yearly payment covering the total CAPEX. The annualized capital cost is determined using the capital recovery factor:

$$\text{CAPEX}_a = \text{CAPEX}_{\text{tot}} \cdot \frac{I_{\text{fix}} \cdot (1 + I_{\text{fix}})^{t_p}}{(1 + I_{\text{fix}})^{t_p} - 1} \quad (5.2)$$

where I_{fix} represents the fixed annual interest rate. Based on literature recommendations by Syed et al. [85], a rate between 0.07 and 0.11 is typical; for this study, a value of 0.09 is selected, assuming a project lifetime of 20 years.

The Operating Cost reported by APEA corresponds to the annual operating expenditure of the H_2 liquefaction plant, as APEA's assessment is structured on a yearly basis. This cost includes both fixed and variable expenses, primarily categorized under Operation & Maintenance (O&M). However, one notable omission is the cost associated with fluid losses, especially from turbomachinery. Since these leakages are not modeled in the process simulation, their economic impact must be estimated separately.

To account for this, fluid loss is assumed to be 1.5% of the plant's daily liquefaction capacity in the base scenario (86 TPD). The cost of lost H_2 is taken as 3.0 €/kg H_2 . Furthermore, for mixed refrigerant systems, the makeup flow required to compensate for leakage is estimated at 1.5 kg h $^{-1}$, with a cost of 5.0 €/kg H_2 . These factors are included in the total annual operating cost of the liquefaction facility.

After applying the necessary adjustments to the annual CAPEX and OPEX of the H_2 liquefaction facility, the Specific Liquefaction Cost (SLC) can be determined using Equation 2.25. The annual production capacity of the liquefaction plant, denoted as $\dot{m}_{\text{LH}_2,a}$, is calculated as:

$$\dot{m}_{\text{LH}_2,a} = U_a \cdot 8760 \frac{\text{hours}}{\text{year}} \cdot 3600 \frac{\text{seconds}}{\text{hour}} \cdot \dot{m}_{\text{LH}_2} \quad (5.3)$$

Here, the plant utilization factor U_a is assumed to be 0.95, which translates to approximately 8,322 operational hours per year. A representative summary of the SLC calculation using APEA for total capital and operating costs is shown in Figure 5.8.

	Project Capital Cost	Plant Operating Cost
Values Obtained From APEA:	€ 129.021.228,36	€ 28.074.908,63
Parameter	Value	Remarks
Working Capital (10%)	€ 12.902.122,84	
Total CAPEX (2022)	€ 141.923.351,20	
CEPCI 2022	816,00	CEPCI Composite Index
CEPCI 2024	799,50	CEPCI Composite Index
Total CAPEX (2024)	€ 143.048.045,58	Equation 5.1
Payment period	20,00	in years
Fixed annual interest	0,09	
Annual CAPEX 2024	€ 15.670.409,15	Equation 5.2
Total annual OPEX	€ 28.074.908,63	
Hydrogen leaks	1.50%	of plant capacity
Refrigerant leaks	1.5	kg/h
Fluid Loss OPEX	€ 1.412.550,00	
Total annual OPEX with fluid loss	€ 29.487.458,63	
Total annual CAPEX+OPEX	€ 45.157.867,78	
Liquefier capacity	86,00	
Utilisation rate	0.950	
SLC	€ 1,51	per kg of LH2
Specific cost details		Remarks
Specific H ₂ cost	€ 3,00 per kg of H ₂	Assumption
Specific MR cost	€ 5,00 per kg of MR	Assumption
Electricity price	€ 0,10 per kWh	Base-scenario
Cooling water price	€ 0,050 per m ³	Assumption

Figure 5.8: Summary sheet of SLC calculation based on cost estimates from APEA

6

Results & Evaluation

This chapter presents and evaluates the key outcomes of the hydrogen liquefaction process modeling, with particular focus on the performance of the plate-fin heat exchanger (PFHX). The results are derived from simulations conducted using the enhanced Python-based model, incorporating a helium-neon coolant mixture and dynamic property updates. The primary outputs include the temperature and para-hydrogen conversion profiles along the exchanger, coolant-side behavior, and derived sizing metrics. These findings are then discussed in the context of their thermodynamic relevance and implications for equipment design.

Beyond the PFHX, the chapter also addresses process-wide performance as captured in the Aspen HYSYS simulation, comparing alternative configurations and highlighting the trade-offs encountered during optimization. The evaluation section reflects on the modeling approach, identifies limitations, and outlines possible areas for future refinement, such as improvements in property prediction, ejector feasibility, and system integration strategies.

6.1. PFHX Results

To evaluate the final stage ortho-to-para H_2 conversion, the enhanced PFHX Python model was executed using the configuration shown in Figure 3.5. The key input parameters were adapted to match those of the Aspen HYSYS V14 liquefaction process, ensuring consistency across both modeling domains. In particular, the reactant stream was initialized at 74.15 K with 53.39% para- H_2 content, while the helium-neon coolant mixture entered at 27.45 K and 300 kPa with a helium mole fraction of 0.8. The 53.39% para- H_2 content was selected after interpolating from Figure 2.4 at the inlet temperature of the reactant.

Figure 6.1 shows the simulation results for the length of the reactor. Panel (a) displays the temperature profiles of the reactant and coolant streams. The reactant cools from 74 K to below 30K over the 7-meter channel, closely approaching the coolant temperature due to effective thermal coupling.

Panel (b) illustrates the evolution of the para- H_2 mole fraction. Starting from an inlet concentration of 0.53, the H_2 stream undergoes spin-isomer conversion as temperature decreases, reaching a final para-fraction of 0.9815 at the reactor outlet. This result is nearly in equilibrium with the theoretical conversion curve, shown as a dashed line. The steep increase in para-fraction between 50–30 K is consistent with Figure 2.3 following standard statistical mechanics treatments.

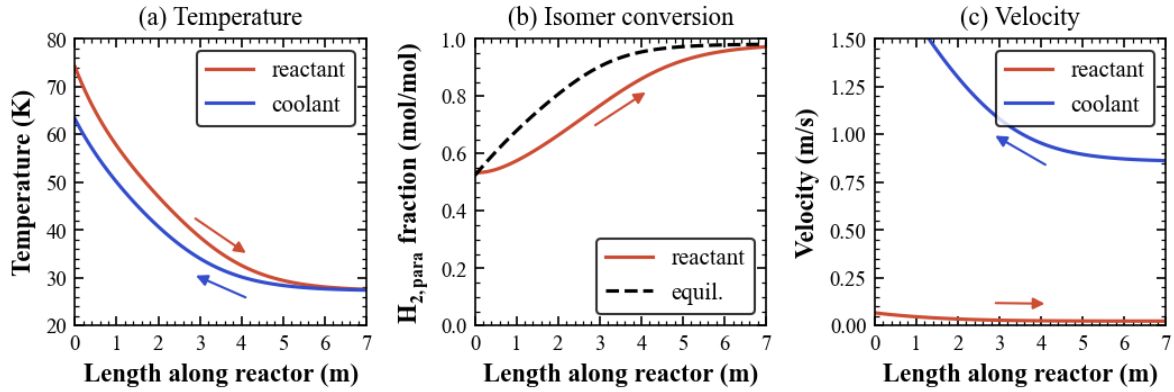


Figure 6.1: Simulation results along the PFHX length: (a) temperature profiles, (b) para-H₂ conversion, and (c) velocity profiles.

Panel (c) shows the velocity profiles of the streams. As expected, the reactant velocity remains nearly constant due to low compressibility, while the coolant decelerates as heat is absorbed, leading to modest density increases.

Table 6.1: Summary of PFHX model input parameters.

Parameter	Value	Parameter	Value
H ₂ $\dot{m}$	86 TPD	Length	7.0 m
Coolant $\dot{m}$	596.16 TPD	Width	1.5 m
Inlet para-H ₂ fraction	0.5339	Height	2.0 m
Outlet para-H ₂ fraction	0.9815	Fin Thickness	0.4
Inlet Coolant T	24.45 K	Fin Height	6.0
Inlet Coolant p	300 kPa	Fin Pitch	1.5
Inlet Reactant T	74.15 K	Separation Length	6.0
Inlet Reactant p	7500 kPa	Parting Sheet Thickness	1.5
Helium mole fraction (coolant)	0.80	Catalyst solid fraction	0.31
Simulation nodes	200	Catalyst diameter	424.0 μm
Tolerance	0.0001	Catalyst density	4.0 g cm ⁻³

Overall, the model 98.15% para-H₂ conversion having a PFHX with the following dimensions: $7.0 \times 1.5 \times 2.0$ m. This size is subsequently implemented into the APEA to accurately validate its total costs. These results serve as the basis for thermal sizing and subsequent capital cost estimation.

6.2. Process Simulation Results

This section presents and discusses the results derived from the Aspen HYSYS process simulation, focusing on two primary analyses. Initially, the ejector performance results are explored to clarify why this design was ultimately not adopted in the final liquefaction system, despite its potential CAPEX advantages. Subsequently, a comprehensive techno-economic analysis of the final hybrid Brayton-Claude liquefaction cycle is detailed. This analysis constitutes the core of the results and emphasizes improvements in specific energy consumption (SEC) and specific liquefaction cost (SLC), providing key insights into the effectiveness and economic viability of the optimized process.

Ejector Performance Analysis

This subsection presents the findings from the analysis of the ejector performance in the modified hydrogen liquefaction process. Initially, the ejector concept was considered promising for reducing CAPEX by eliminating the requirement for a turbine, a valve, and a flash compressor in the cryogenic cooling stage. T3, V5, and FC2, respectively, from Figure 3.1. However, a detailed analysis reveals critical operational constraints that limit its viability as part of the optimized final system.

Aspen HYSYS V12 does not have an ejector block, and as a result, Aspen HYSYS V14 was installed while trying to model this component. A .dll Aspen extension file was found that was later added to the process engineering program. However, while the extension addition worked, the ejector block was not sufficient for such a cryogenic condition. Only gas-to-gas modeling was possible, while the secondary inlet stream was modeled as liquid. However, the MATLAB code provides scientific details and insights into its performance.

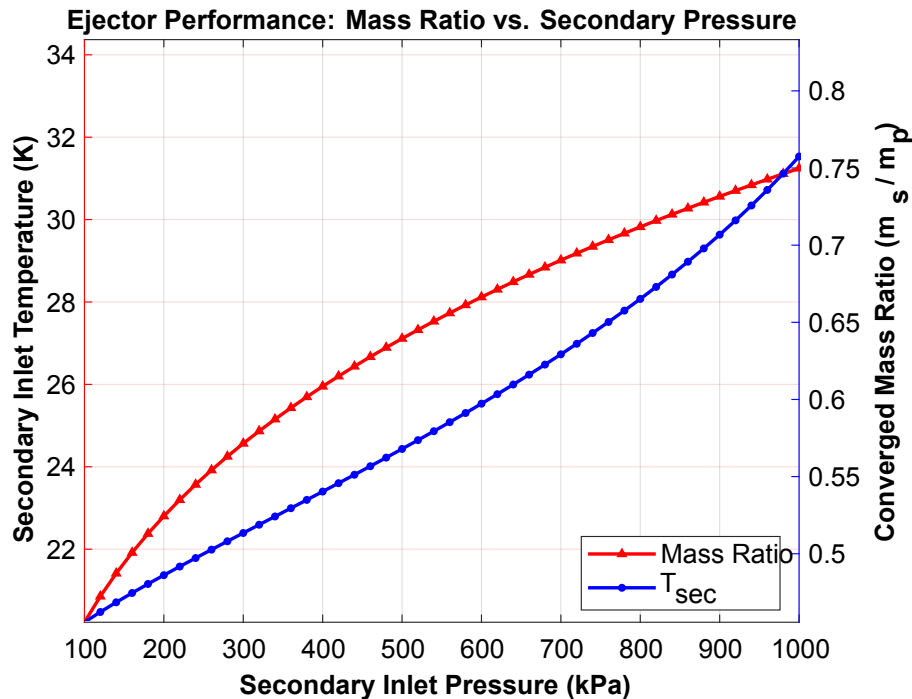


Figure 6.2: Ejector secondary pressure versus mass entrainment ratio and secondary temperature.

Figure 6.2 shows the relationship between the secondary inlet pressure of the ejector, the secondary temperature, and the resulting mass entrainment ratio. These values were obtained using a MATLAB simulation, where the mass entrainment ratio is defined as the mass flow of the secondary (entrained) fluid relative to the primary (motive) fluid. The plot provided clearly indicates that increasing the secondary inlet pressure of the ejector leads to a corresponding increase in both the mass entrainment ratio and the secondary outlet temperature. The mass entrainment ratio exhibits a nonlinear behavior, initially gradually increasing and subsequently accelerating sharply as it approaches the critical pressure (12.97 bar). In addition, the secondary outlet temperature increases consistently with increasing secondary inlet pressure, becoming particularly pronounced at higher pressures. Thus, a higher secondary inlet pressure results in greater entrainment capacity but is accompanied by elevated secondary outlet temperatures, which affects subsequent process requirements and efficiency.

In particular, the lowest gas production was found at a secondary inlet pressure of approximately 1.1 bar (0.4589 kg s^{-1}), still resulting in roughly 20% more vapor generation than in the baseline process designed by Kerkar et al. [97]. Additionally, this low secondary pressure configuration introduced critical temperature crossover issues within the PFHX, as the ejector outlet temperature decreased considerably.

The increase in vapor generation by the ejector can be attributed to its working principle. As discussed by Elbel et al. [100], the ejector uses the kinetic energy of a high-pressure motive fluid to entrain and compress a lower-pressure fluid, resulting in intermediate pressure fluid flow with mixed phases at the diffuser outlet. This inherent mixing and compression process generates additional vapor due to the high shear and turbulent mixing effects within the ejector's mixing chamber and diffuser.

The generated additional vapor, once separated, must then be recompressed to achieve the required

liquefaction conditions. Taking for example, a secondary inlet pressure of 4 bar, the resulting H_2 cold gas flow rate increased to 0.5403 kg s^{-1} compared to Kerkar et al. value of 0.3834 kg s^{-1} [97]. This increase requires a substantial increase in the mass flow of helium-neon coolant, from a baseline of 9.7 kg s^{-1} to 23.3 kg s^{-1} , representing an approximate 140% increase. This increase directly amplifies the demand for compression power, significantly elevating OPEX, especially considering that the compressor input power scales proportionally with the increase in coolant mass flow. This phenomenon underscores a fundamental disadvantage of the ejector-based configuration, where OPEX escalations outweigh initial capital cost reductions.

Given these constraints, the ejector-based system, despite promising reductions in CAPEX, was not selected for the final optimized design. However, these insights provide valuable guidance for future techno-economic evaluations of ejector applications in similar cryogenic hydrogen liquefaction processes.

6.2.1. Baseline Scenario

Figure 6.3, Table 6.2, Table 6.3, and Table 6.4 outline the final process flow diagram and its working points. The capacity of the liquefaction plant is of 86 TPD, while its exergy efficiency with power recovery is 44.83%

The process flow diagram depicted in Figure 6.3 comprises a range of components including compressors with interstage aftercooling, heat exchangers, turbines, isenthalpic expansion valves, separators, mixers, and a recuperator. In comparison to the baseline layout shown in Figure 2.7, where a single 15-stage Nelium compressor was used, the current design opts for a configuration of four compressors, consisting of four-four-three-three stages respectively, coupled with intercooling to condition the refrigerant prior to entry into the recuperator and subsequent expansion in turbines T1 and T2. These turbines deliver the necessary cryogenic cooling capacity for hydrogen liquefaction.

For hydrogen feed gas compression, the system employs three high-pressure compressors (HPCs) to increase the pressure from 21 bar to 80 bar. The isentropic efficiencies of the respective compressors are:

- HPC-1: 81.93%
- HPC-2: 81.95%
- HPC-3: 79.51%

In parallel, the mixed refrigerant compression system, made up of an eight-component refrigerant mixture, is also comprised of three compressor units that elevate the pressure from 1.1 bar to 24.8 bar. The isentropic efficiencies of these mixed refrigerant compressors (MRCs) are:

- MRC-1: 84.00%
- MRC-2: 84.51%
- MRC-3: 83.95%

To achieve liquid H_2 , the process requires reducing the H_2 gas temperature from 74.15 K to below 28 K. This cryogenic cooling duty is fulfilled by a Nelium refrigerant cycle. The Nelium mixture is compressed from 3 bar to 60 bar through four dedicated compressor units, labeled C-1 to C-4. The first two units comprise four compression stages, while the final two units consist of three stages each; and are all equipped with interstage intercoolers to ensure thermodynamic efficiency and temperature control during compression.

In parallel, the flash gas stream (H9) after being used as a coolant throughout all PFHXs, composed of low pressure p- H_2 , is compressed from 1 bar to 21 bar by compressor unit (ReC) which consists of six compressors. These compressors operate with isentropic efficiencies between the range of 71.39% and 76.62%, and are employed to recover and reintegrate the H_2 vapor into the process.

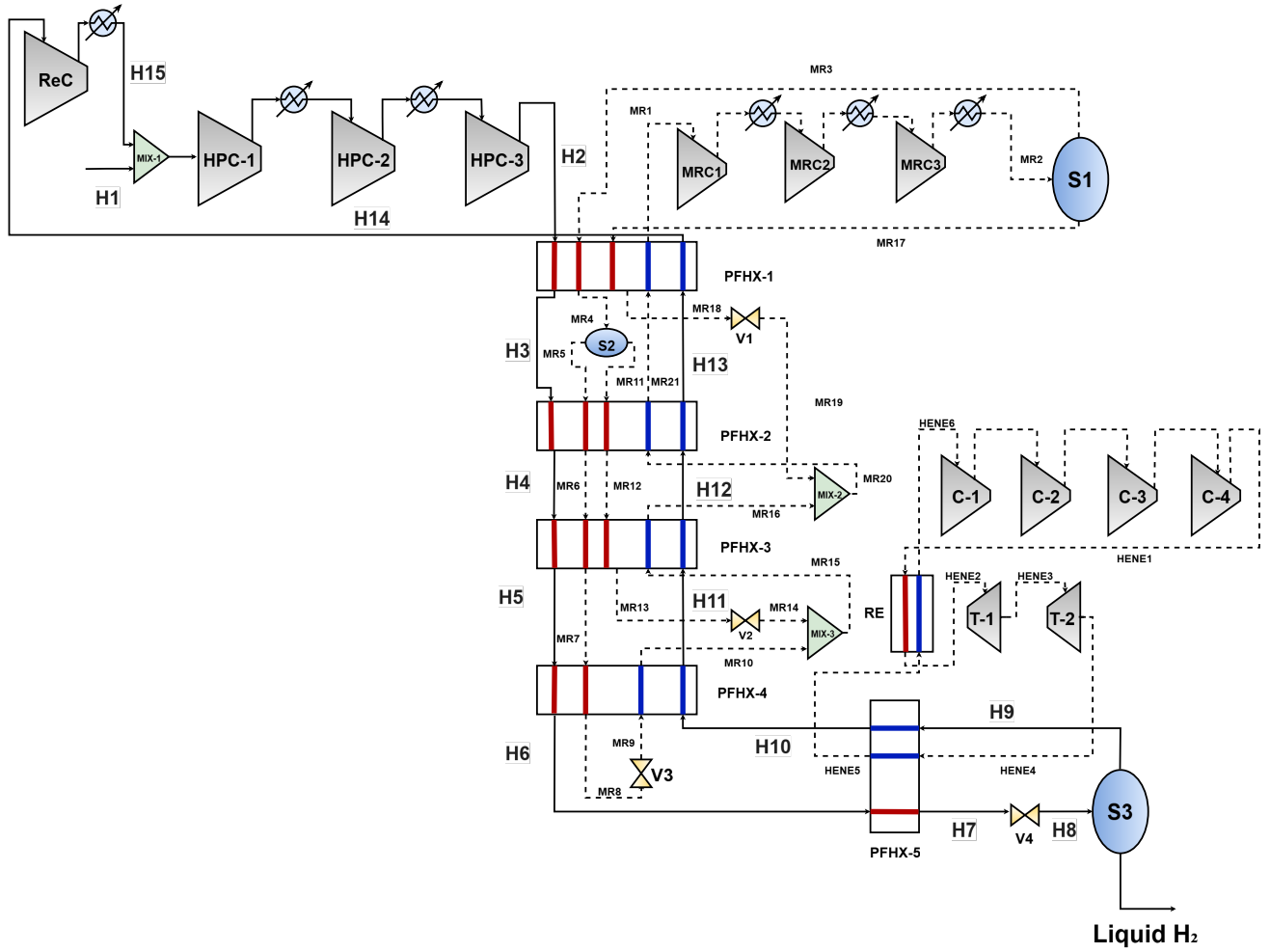


Figure 6.3: High-Pressure H₂ Liquefaction Based on a Hybrid Brayton-Claude Cycle Process Flow Diagram

Table 6.2: H₂ Stream State Points

Stream	Temp. [K]	Press. [bar]	Flowrate [kg s ⁻¹]	Vapour Quality [-]
H1	310	21	1	1
H2	310	80	1.4847	1
H3	255	78.7	1.4847	1
H4	235	77.5	1.4847	1
H5	144.95	76.2	1.4847	1
H6	74.15	75	1.4847	1
H7	27.95	73.8	1.4847	0
H8	20.2	1	1.4847	0.3265
H9	20.2	1	0.4847	1
H10	46.65	1	0.4847	1
H11	142.15	1	0.4847	1
H12	180	1	0.4847	1
H13	195	1	0.4847	1
H14	210	1	0.4847	1
H15	310	21	0.4847	1
Liquid-H ₂	20.2	1	1	0

Table 6.3: Helium-Neon Stream State Points

Stream	Temp. [K]	Press. [bar]	Flowrate [kg/s]	Vapour Quality
HENE1	310	60	6.9	1
HENE2	78.65	60	6.9	1
HENE3	46.75	13.3	6.9	1
HENE4	27.45	3.0	6.9	1
HENE5	74.05	3.0	6.9	1
HENE6	310	3.0	6.9	1

Table 6.4: Pre-cooling MR Stream State Points

Stream	Temp. [K]	Press. [bar]	Flowrate [kg/s]	Vapour Quality
MR1	288.2	1.1	35	1
MR2	310	24.8	35	0.7150
MR3	310	24.8	20.4583	1
MR4	254.30	24.4	20.4583	0.716
MR5	255	24.4	12.3083	1
MR6	235	24	12.3083	0.9172
MR7	144.95	23.6	12.3083	0.3718
MR8	74.95	23.2	12.3083	0.0146
MR9	71.85	1.2	12.3083	0.0943
MR10	139.35	1.2	12.3803	0.6857
MR11	255	24.4	8.1500	0
MR12	235	24	8.1500	0
MR13	144.95	23.6	8.1500	0
MR14	144.45	1.2	8.1500	0.0301
MR15	141.95	1.2	20.4583	0.4969
MR16	224.65	1.2	20.4583	0.9449
MR17	310	24.8	14.5417	1
MR18	255	24.4	14.5417	0
MR19	234.80	1.2	14.5417	0.1904
MR20	226.52	1.2	35	0.7401
MR21	235.21	1.1	35	0.7975

As seen in Figure 6.3, the Hybrid Brayton-Claude cycle uses 2 turbines in the cryogenic cycle, T-1 and T-2. Used to cool down the He-Ne coolant, their isentropic efficiencies are 88.30% and 89.95% respectively.

The Specific Energy Consumption (SEC) value of this H₂ liquefaction plant is of 6.723 kWh/kg_{LH₂} with power recovery, while 6.983 kWh/kg_{LH₂} when the power recovery is not taken into account. The He-Ne cycle is the liquefaction stage that consumes most energy with a total of 2.89 kWh/kg_{LH₂}, followed by the MR compression cycle accounting for 2.50 kWh/kg_{LH₂}. This leads to an exergy efficiency value of 44.831% which reduces to 43.162% when the power recovery is not accounted.

The evaluation of both CAPEX and OPEX for the plant is conducted using the techno-economic framework outlined in chapter 5. In the baseline scenario, the specific liquefaction cost is estimated at 1.51€/kg_{LH₂}. The annual capital expenditure (CAPEX) for the liquefaction facility in 2024 amounts to M€ 15.67. The operating expenditure (OPEX), which includes losses from hydrogen and refrigerant leaks as well as energy recovery from turbine shafts, is estimated at M€ 28. The total project cost is M€ 129.0 based on 2022 price levels. Using the cost-scaling methodology described in Equation 5.2, the cost is adjusted to 2024 pricing, resulting in a total value of M€ 143.0.

Table 6.5: Compressor Units main cost parameters and direct costs for 86 TPD

Compressors	Items in Parallel	Inlet Flowrate [kg/s]	Power Input [kW]	Total Direct Costs [€]
HPC-1	1	1.4847	1116	2,126,300.00
HPC-2	1	1.4847	1125	2,183,800.00
HPC-3	1	1.4847	1142	2,220,000.00
MRC1	1	35	2819	9,234,700.00
MRC2	1	35	3270	4,463,700.00
MRC3	1	35	2923	2,361,100.00
C-1	1	6.9	2973	6,888,800.00
C-2	1	6.9	3064	3,636,000.00
C-3	1	6.9	2210	2,278,700.00
C-4	1	6.9	2157	2,327,300.00
ReC1	1	0.4847	138.2	1,488,700.00
ReC2	1	0.4847	162.9	1,565,300.00
ReC3	1	0.4847	193.6	1,642,800.00
ReC4	1	0.4847	224.3	1,667,100.00
ReC5	1	0.4847	224.4	1,665,400.00
ReC6	1	0.4847	226.1	1,678,400.00

Table 6.6: Turbine units main cost parameters and direct costs for 86 TPD

Turbines	Inlet Flowrate [kg/s]	Power Output [kW]	Total Direct Costs [€]
T-1	6.9	579.2	489,200.00
T-2	6.9	352.9	365,400.00

Table 6.7: Heat Exchanger units main cost parameters and direct costs for 86 TPD

Heat Exchangers	Items in Parallel	Dimensions [m]	Total Direct Costs [€]
HEX1	7	2.482 × 0.595 × 0.831	4,692,300.00
HEX2	5	2.26 × 0.593 × 0.739	3,218,200.00
HEX3	7	4.516 × 0.594 × 0.89	6,652,700.00
HEX4	4	3.098 × 0.596 × 0.691	2,290,200.00
HEX5	1	7.000 × 1.500 × 2.000	2,490,200.00

Table 6.5, Table 6.6, and Table 6.7 present the total direct costs of the compressor, turbine, and heat exchanger units based on key design parameters. Among these components, the compressor units represent the highest expenditure, with an aggregated direct cost of approximately M€ 50. Figure 6.4 illustrates the cost distribution across the four major subsystems of the liquefaction plant: the H₂ feed and recirculation compression system, the precooling compression system, the cryogenic compression and expansion system, and the heat exchanger system (comprising five PFHXs). Of these, the heat exchangers incurs the highest cost, with an estimated total of M€ 19.34, a substantial change from Kerkar et al. [97] plant as the cryogenic compression and expansion system took the biggest portion of direct costs. This reflects the optimization work done on the He-Ne cryogenic cycle. Within the heat exchanger subsystem, HEX3 is the most expensive one with a total direct cost of M€ 6.65, while MRC1 is identified as the costliest individual unit of the entire liquefaction plant, with a direct cost of M€ 9.23.

Figure 6.5 shows the breakdown of annual operating expenses (OPEX) for the base case, based on 2022 pricing in the APEA model. The total annual OPEX is M€ 28.05, of which M€ 21.47 million, accounting for 72.8%, is attributed to utility costs. Under this scenario, the annual expenditure is divided into approximately 33% CAPEX and 67% OPEX relative to the total yearly costs.

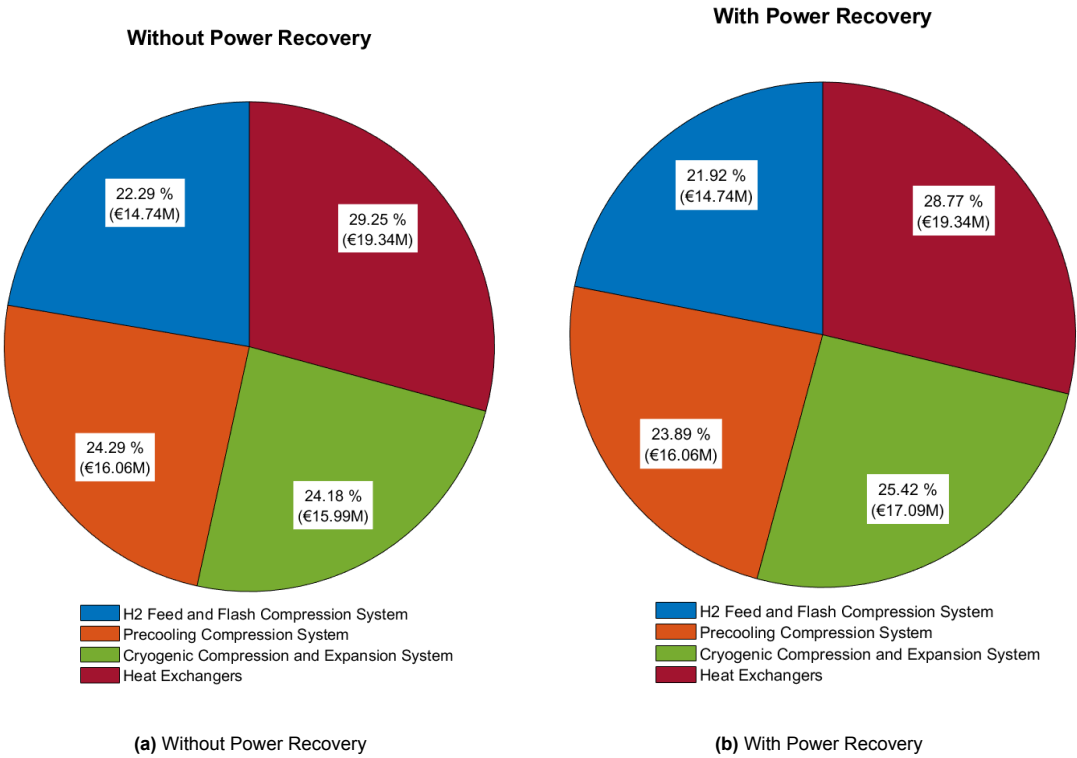


Figure 6.4: CAPEX Distribution per Process Equipment Direct Cost (using APEA's 2022 pricing basis)

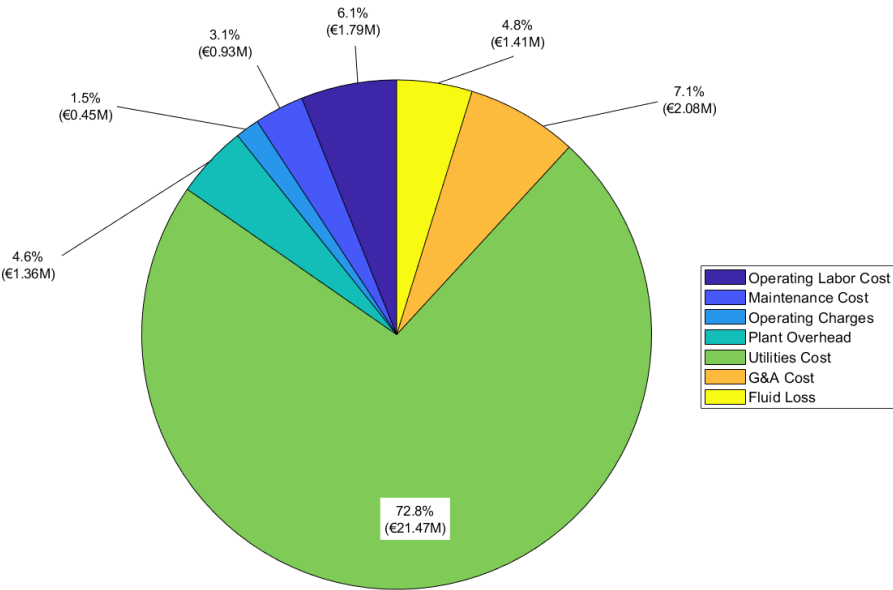


Figure 6.5: Annual OPEX Distribution Pie Chart

Baseline Scenario Analysis: The final H₂ liquefaction plant based on a hybrid Brayton-Claude cycle is summarized using 3 main KPIs: SEC, SLC, and exergy efficiency. After detailed optimization and

refinement, these values are compared to Kerkar et al. [97] plant and the reference case by Berstad et al. [59].

In the reference case by Berstad [22], the exergy efficiency is reported at 44.7%, while Kerkar et al. [97] plant, the calculated exergy efficiency is 43.5%. Similarly, this thesis yielded to an exergy efficiency of 43.16% for the baseline scenario. The specific energy consumption (SEC) values derived from this large-scale liquefaction design are notably below those of conventional hydrogen liquefiers, which generally range between 10 to 12 kWh/kg_{LH₂}. In this work, the SEC is 6.723 kWh/kg_{LH₂} with power recovery and 6.983 kWh/kg_{LH₂} without power recovery, representing roughly a 39% reduction compared to current industrial benchmarks. Lastly, this thesis achieved a SLC of €1.49/kg_{LH₂}, a 5.1% decrease from Kerkar et al. Brayton cycle [97]. Nonetheless, a detailed discussion is given in the following chapter.

6.2.2. Sensitivity Analyses

Sensitivity analysis is a critical step in techno-economic evaluations of H₂ liquefaction because it identifies the parameters to which the process economics and efficiency are most responsive. In line with findings from literature, the cost of electricity emerges as the dominant factor influencing the specific liquefaction cost (SLC). Variations in electricity price strongly affect operational expenditure, reflecting the high share of power consumption in overall costs. Furthermore, CAPEX optimization can also lead to a reduced final SLC as seen in the He-Ne cryogenic compression cycle. The following analyses compare the baseline scenario with that in which power recovery is included in the total final costs.

Electricity Cost Analysis: The specific liquefaction costs is evaluated assuming an electricity price of 0.1 €/kWh. The sensitivity analysis extends this by calculating the SLC of the 86 TPD conceptual plant over a range of electricity prices from 0.01 to 0.3 €/kWh. The results are displayed in Figure 6.6, where the plot depicts the influence of variation in electricity cost on the economics of the plant.

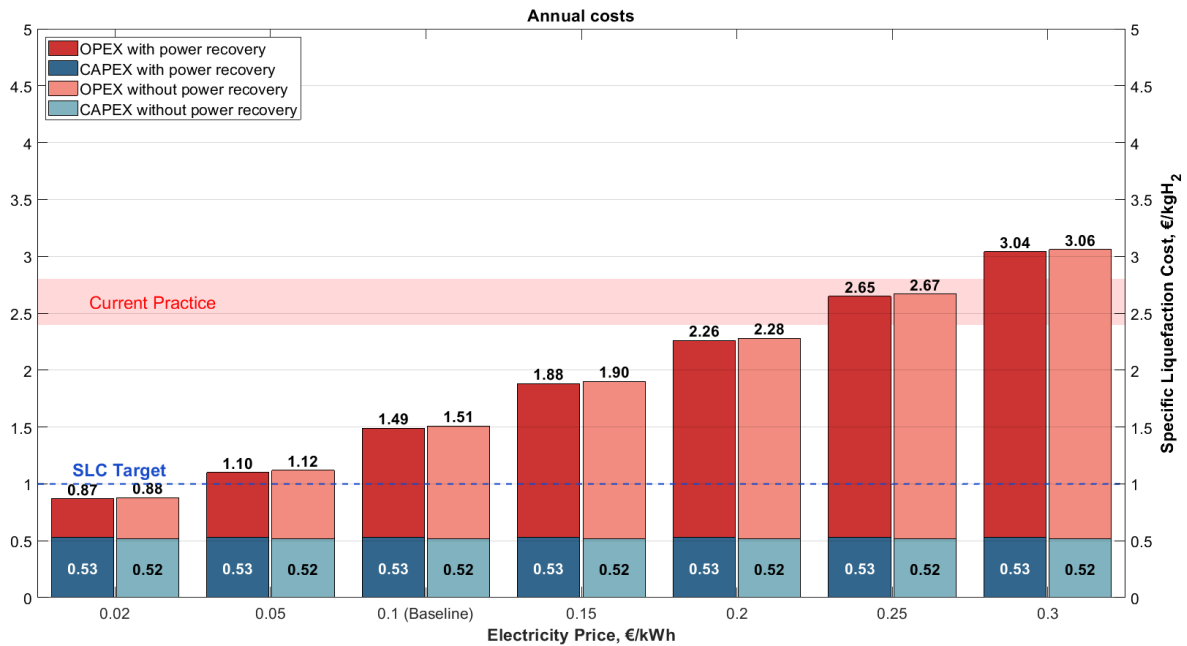


Figure 6.6: SLC of the H₂ liquefaction plant as a function of electricity price

The results displayed in Figure 6.6 highlight the strong dependence of the specific liquefaction cost (SLC) on electricity price. As shown, the SLC rises nearly linearly with increasing electricity costs, reaching values between 2.24 and 2.65 €/kg_{LH₂} at 0.20–0.25 €/kWh. This corresponds closely to the current industrial cost range of 2.37–2.85 €/kg_{LH₂}. At lower electricity prices, however, the SLC decreases significantly, approaching the long-term cost reduction targets of below 1.00 €/kg_{LH₂}. An electricity price of 0.035 €/kWh gives the desired SLC result for the baseline scenario.

Figure 6.6 also shows the difference contributions CAPEX and OPEX give to the final SLC. As can be observed, as electricity price increases, the OPEX contribution becomes the main driver to the SLC; reaching over 80% of the total cost at 0.30 €/kWh. Power recovery is considered and compared to the baseline scenario. As can be noticed, CAPEX is 0.01€ higher in this case due to the capital cost of the electric generators coupled to the turbines, while the OPEX contribution is less compared to the baseline scenario. The SLCs of these two scenarios do not differ as much compared to Tamarona et al. claudé cycle as in this process, just two turbo-expanders are implemented, while in the claudé cycle up to 4 are used; recovering more total power and further decreasing the total OPEX.

The electricity price range considered extends beyond typical EU averages to capture the potential effect of sharp price escalations, such as those observed during recent energy market volatility, on hydrogen liquefaction economics. This sensitivity underlines the critical role of stable, low-cost electricity supply in achieving competitive large-scale LH₂ production. It is important to mention that electricity price only affects the OPEX, with the CAPEX keeping a constant SLC contribution despite changes in the electricity cost.

SLC & CAPEX as a Function of Annual Interest Rate & Payment Period: The interest rate and annual payment period are key financial parameters that directly influence the capital recovery factor and consequently the levelized cost of H₂ liquefaction. Evaluating their impact through sensitivity analysis is essential, as variations in financing conditions can significantly alter project feasibility and investment attractiveness, even when technical performance remains unchanged. As a result, more analysis has been done on these two variables, as they were initially taken as an average of current practices.

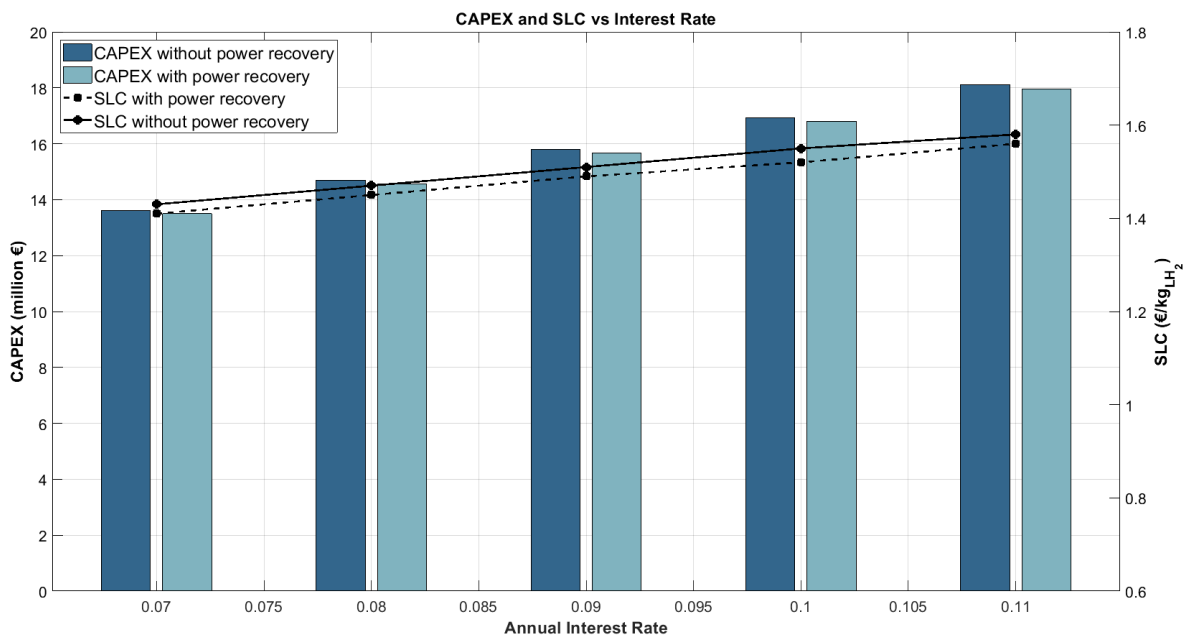


Figure 6.7: SLC & CAPEX of the H₂ liquefaction plant as a function of interest rate

Figure 6.7 shows a decrease in both CAPEX and SLC as the interest rate decreases. This reduction is relatively linear and does not cause serious variations as does the price of electricity, see Figure 6.6. Similarly, it is important to note that the interest rate only affects the annual CAPEX.

Additionally, Figure 6.7 visualizes the difference in CAPEX between the two scenarios, where the one with power recovery included has a slightly higher CAPEX due to the single electric turbo generator, coupled to the two iso-axial He-Ne turbines (T1 & T2).

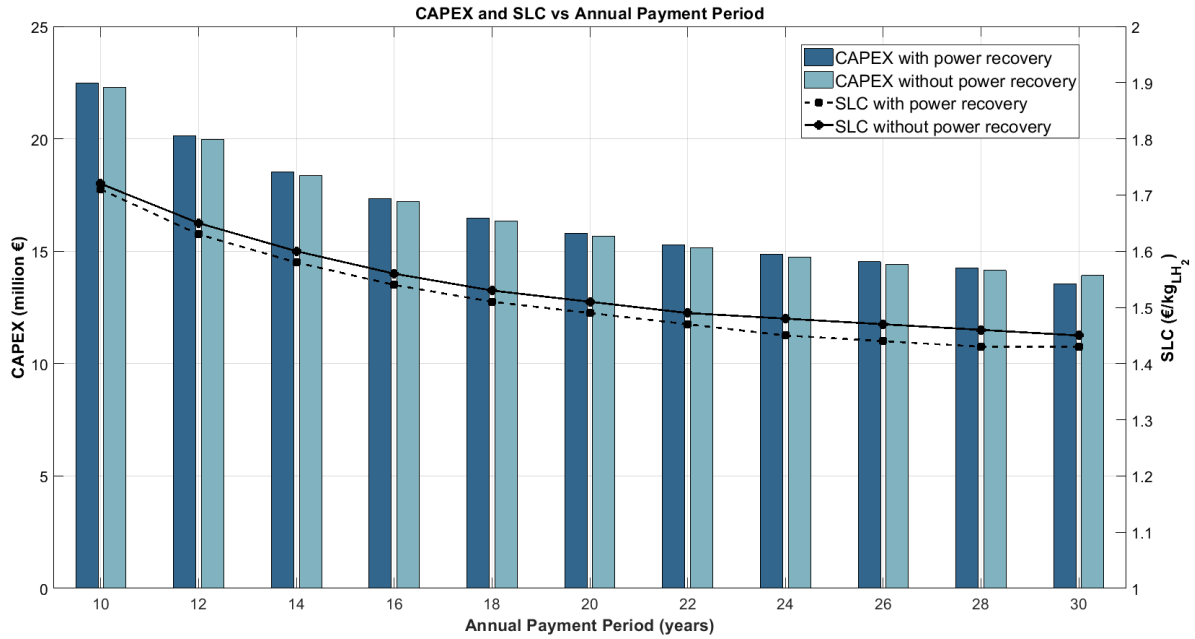


Figure 6.8: SLC & CAPEX of the H₂ liquefaction plant as a function of annual payment period

Figure 6.8 shows a decrease in both CAPEX and SLC as the annual payment period in years increases. However, it is possible to observe a logarithmic-like function for the SLC since after 22 years the change in this indicator is only 0.01€. As a result, keeping 20 years for this variable is determined to be a great choice, as it balances between a cheaper liquefaction cost while avoiding too lengthy contract period, exposing the financing to uncontrollable variables such as crisis.

He-Ne Cryogenic Compression Cycle, Compressor Analysis: As previously described, optimization of the He-Ne cryogenic cycle was achieved after modifying the final liquefaction stage of the plant by recirculating back the excess H₂ gas. This allowed the He-Ne coolant mass flow rate to drop to 6.9 kg s⁻¹ while keeping the same cooling capacity but at a lower discharge pressure. In fact, the He-Ne mixture was compressed from 3 bar to 60 bar (from a previous 2.4 bar to 75 bar from the reference model described by Berstad et al. [59]).

This new and lower pressure ratio led to the selection of a total of 4 compressors with 14 stages in total, as seen in Equation 1. However, the same compression system could have been achieved with five compressors with three stages for the first four compressors and two stages for the last compressor. In doing so, the CAPEX increases as a result of the presence of an additional compressor. Nevertheless, curious insights about the SEC are found as this KPI is subject to a decrease. Figure 6.9 provides insights for the two configurations under both scenarios with and without power recovery.

As can be observed, SLC is 0.09 € cheaper per kilogram of liquid produced H₂, which means M€ 2.68 saved per year under both scenarios. The difference between these two scenarios lays under the CAPEX difference of indeed 0.1 € per kilogram of produced liquid H₂. However, using five compressors in the He-Ne leads to a lower SEC value, with a reduction of 2.68%, reaching a value of 6.796 kW kg⁻¹ LH₂ without power recovery and down to 6.537 kW kg⁻¹ LH₂ when power recovery is considered, a reduction of 3.88% from the same scenario but with four compressors. This is because in the Aspen HYSYS simulation, implementing five compressors in the He-Ne compression cycle leads to a slightly smaller total power input, decreasing both the final SEC and the OPEX, although minimal.

This provides valuable insights into further optimization of the H₂ liquefaction plant, not only in the He-Ne cycle, but also in other stages of the plant, such as the feed compression stage, mixed refrigerant pre-cooling, or the final recirculation compression unit. Lastly, adding more compressors in a multi-stage compression train may lead to lower SEC and OPEX, but the CAPEX increase because of the additional component will result in a higher SLC, as well as more maintenance work.

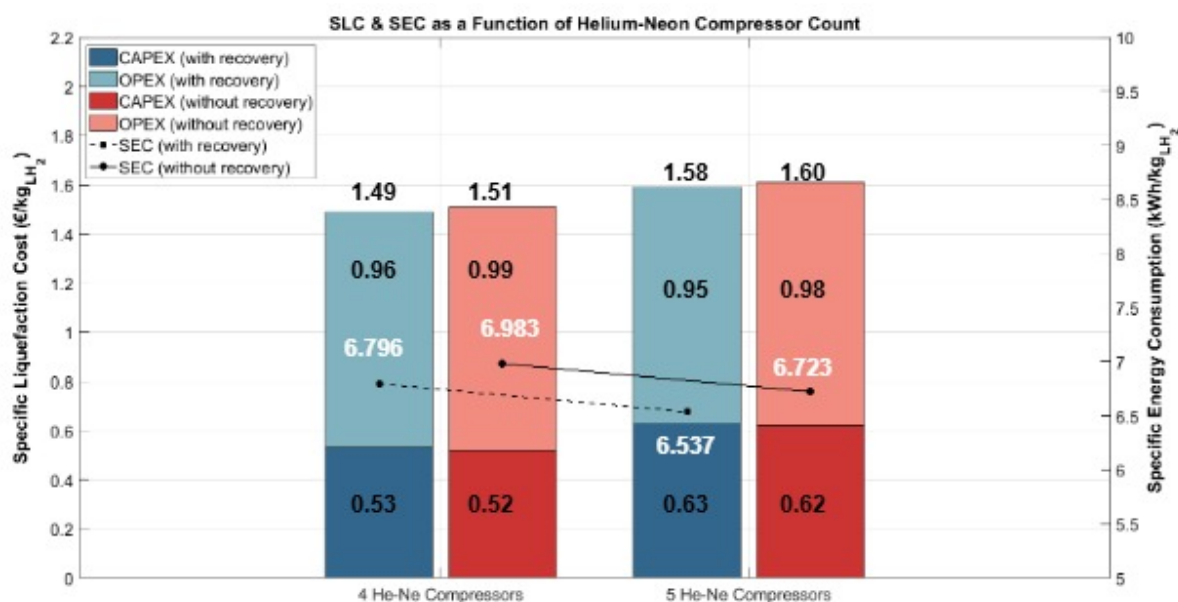


Figure 6.9: SLC & SEC of the H₂ liquefaction plant as a function total number of He-Ne compressors

Future Scenarios: Previous APEA results have all been performed under "New Process" selection in the *General Specs* for Process Description. Table 6.8 outlines the various options for the Process Description that APEA offers, and their respective design allowance and contingency percentages.

Table 6.8: Equipment design allowance and contingency percentages following Process Description

APEA Process Description	Design Allowance %	Contingency %
New & Unproven Process	15	36
New Process	10	30
Redesigned Process	7	26.4
Licensed Process	5	21.6
Proven Process	3	18

As the market for H₂ is expected to significantly increase over the coming decades, up to approximately 20 Mtpa of liquid H₂ traded in 2050 [93], this technology is expected to develop 360°. Major industry players will see a boost in confidence when economical factors such as design allowances and contingency percentages, become more favorable. As a result, Figure 6.10 shows the final analysis on these options.

As can be observed, at an electricity price of 0.1€/kWh, the Proven Process is the cheapest configuration, as expected, with a difference of 0.07€ compared to the current baseline scenario, resulting in M€ 2.09 saved annually.

With increasing confidence within market players and public entity support, an 86 TPD H₂ liquefaction plant is expected to see a 4.64% decrease in specific liquefaction cost. Nevertheless, it is important to mention that the combination of scaling up to higher capacity, and the inclusion of power recovery can further reduce this KPI.

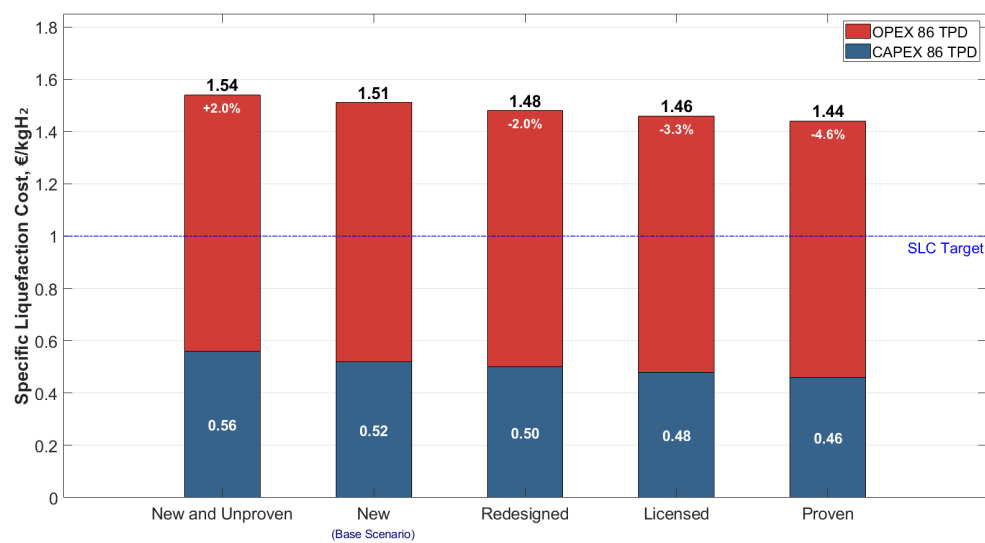


Figure 6.10: SLC results for H₂ liquefaction plant under different Process Description - APEA General Specifications

Conclusion & Recommendation

This chapter synthesizes the main findings of the thesis by addressing the sub-research questions introduced in chapter 1. Through these questions, the analysis connects the thermodynamic and process design aspects of H_2 liquefaction with its economic feasibility and its broader role in future energy systems. Drawing together these perspectives is essential because it not only clarifies the technical and economic boundaries of current liquefaction concepts, but also highlights their implications for scalability, competitiveness, and sustainability. Finally, the chapter formulates recommendations for research and industry, ensuring that the insights gained are translated into directions relevant for advancing both academic knowledge and practical implementation.

7.1. Conclusions

How can a conceptual high-capacity Brayton cycle H_2 liquefaction plant be accurately modeled using a commercial process simulation tool, including ortho-para conversion reaction kinetics, while addressing equations of state limitations?

To accurately model a high-capacity Brayton-cycle LH_2 plant in a commercial simulator, this work used Aspen HYSYS (v14) as the flow sheet backbone and split the problem into (i) thermodynamic modeling throughout the plant with industrially available EOS options and (ii) dedicated modeling of ortho-para (o-p) kinetics and catalytic PFHX performance outside HYSYS, with iterative data exchange. The main flow sheet comprises H_2 compression, MR pre-cooling to roughly 80K, a cryogenic Brayton loop (He-Ne/Nelium), catalytic PFHXs for o-p conversion, final expansion/liquid handling, following the large-scale concepts adopted by prior TU Delft studies, and excess H_2 gas modeling and management.

EOS limitations in standard process simulators were explicitly addressed. For H_2 , HYSYS was interfaced to NIST REFPROP [99] so that normal-/ortho-/para- H_2 properties are computed; the H_2 process stream that undergoes continuous o-p conversion in the PFHX network was treated with the pseudo-equilibrium- H_2 heat-capacity implementation of Valenti et al. [102] by modifying the REFPROP fluid file (PARAHYD.FLD) called from HYSYS. Mixed-refrigerant and Nelium loops were modeled with Peng-Robinson in HYSYS for cycle-level balances, consistent with earlier TU Delft implementations.

Because commercial simulators cannot currently resolve spatial o-p kinetics in multi-stream PFHXs, the kinetics were embedded via a dedicated catalytic PFHX model (boundary-value problem) of O'Neill et al. [22]. This was used to compute catalyst inventory, parafraction profiles, and temperature evolution in a 2-stream surrogate of each catalytic core, while the full geometry and hydrodynamics were sized in Aspen EDR design mode. EDR sizing outputs were fed to both HYSYS and APEA; the catalytic model ensured that the para- H_2 content at cold-end targets 99.5–99.8%.

This hybrid workflow balances accuracy and tractability. REFPROP in HYSYS provides reliable H_2 thermodynamics over 20–300 K, while first-order o-p kinetics validated for industrial catalysts (per Donaubauer/Wilhelmsen and recent reassessments). Similarly, compressors and turbines size was achieved by running the python code from Tamarona et al.[63] according to the process conditions.

An ejector-based cold end recirculation was explored to simplify the loop and cut CAPEX by removing a turbine, an expansion valve, and a compressor. In practice, Aspen HYSYS offered no robust cryogenic two-phase ejector: V12 had no block and the V14 extension handled only gas–gas service, preventing a faithful implementation. A standalone MATLAB entrainment/efficiency model, evaluated at the HYSYS state points, showed the concept required a substantially higher H_2 cold gas flow rate (0.5403 kg s^{-1} against previous results of 0.3834 kg s^{-1} from Kerkar et al. [97]), which in turn drove up Brayton He–Ne duty and compressor work; even at low suction pressures, vapor generation remained above baseline and caused PFHX ΔT crossovers. The implied coolant flow increase toward 23.3 kg s^{-1} (previously 9.7 kg s^{-1}) negated any CAPEX benefit, so the ejector option was discarded in favor of the simpler recirculation design.

In summary, an accurate conceptual model is achieved by (1) using HYSYS with REFPROP (Leachman EOS with implementation of the modified .FLD file by Valenti et al. [102]) for H_2 , PR for MR/Nelium, (2) implementing pseudo-e- H_2 properties in the main H_2 path, (3) sizing PFHXs in Aspen EDR, and compressors and turbines as per the model developed by Tamarona et al. [63], and (4) embedding o–p reaction kinetics via a catalytic PFHX model with iterative reconciliation of para-fraction. This structure follows and extends the reference model by Berstad et al. [59] methodology to the Brayton concept and provides a defensible basis for equipment design and TEA at large capacity, with process optimization done to achieve better techno-economical results.

How can the excess H_2 gas be effectively managed and modeled in the liquefaction process?

Excess H_2 generated at the cold end (flash gas and residual vapor after J–T throttling) is recovered rather than vented or inefficiently re-used, and is modeled explicitly as a recycle loop. In the baseline Brayton concept developed by Kerkar et al. [97], this stream is recompressed by two flash-gas compressors and then re-expanded, an energy-intensive arrangement that nevertheless captures the vapor and returns it to the coldbox as coolant. In this thesis final configuration, excess cold H_2 leaving the last separator is first routed counter-currently through all PFHXs as an additional cold stream to reclaim its exergy, and only then recompressed from 1 bar to initial feed pressure of 21 bar, heated to 310 K, and mixed with the fresh feed upstream of the main liquefaction path. This *cold-recovery + recycle-to-feed* strategy is implemented in Aspen HYSYS using the LNG multi-stream exchanger model and a dedicated recycle compressor train (ReC) made of a six-stage compression system; is iteratively fed back into the simulation via the HYSYS tool *Recycle* (RCY). The approach mirrors industrial practice where H_2 recycle stream provides further cooling duty in the exchangers.

Technically, this treatment lowers Nelium demand, eliminating superfluous cryogenic rotating equipment and the formation of liquids and solids. In this model the He–Ne flow decreases from 9.7 to 6.9 kg s^{-1} , and the fraction of excess gas requiring recirculation drops from 38.34% to 32.65%, improving net liquefaction without compromising cold-end temperatures; the ReC bank (six compressors, 71–77% isentropic efficiency) closes the mass balance of the recycle. Importantly, as the recycled p- H_2 warms through the PFHX network, it partially reconverts endothermically toward o- H_2 , which offsets part of the sensible heat gain, an effect represented via the pseudo-equilibrium- H_2 property method within HYSYS which was validated by testing n- H_2 in the same process simulation.

A key limitation of the He–Ne refrigeration loop is the scarcity of experimental transport property data at cryogenic conditions, which restricts validation of mixture behavior in compressors and turbines. Although entropy-scaling refinements improve the prediction of thermophysical properties compared to Peng–Robinson EOS, their reliability ultimately depends on benchmarking against high-fidelity experimental measurements. This uncertainty affects the accuracy of the prediction of their transport properties. To improve confidence in Nelium cycle modeling, targeted experimental campaigns are needed to validate entropy-scaling correlations and optimize mixture composition for large-scale H_2 liquefaction.

Overall, this integrated management and modeling of excess H_2 provides a thermodynamically and economically superior solution to ejector-based or re-expand/compress schemes for Brayton-based liquefiers.

How to effectively estimate the capital and operating costs of a conceptual hydrogen liquefier based on the process simulation model and preliminary equipment designs, and what are the key factors influencing the economic feasibility of the plant?

The CAPEX and OPEX of the large-scale Brayton liquefier were estimated by coupling the converged Aspen HYSYS model to Aspen Process Economic Analyzer (APEA) and feeding in the preliminary equipment designs. HYSYS unit operations were mapped to ICARUS cost objects, centrifugal compressors (GC CENTRIF), turbo-expanders (TUR TURBOEXP), and plate-fin heat exchangers (HE PLATE FIN). PFHX core dimensions and counts came from Aspen EDR and were manually entered to refine direct costs; inter-and-aftercoolers were included the option within each compressor that required one. Where power recovery is considered, generators were added explicitly as EG TURBO GEN items (one per shaft line), nonetheless, for the baseline scenario, power recovery was not considered. This workflow mirrors Tamarona et al. [63] and keeps process-to-cost data consistent across compressors, expanders and multistream PFHXs.

The basis of the APEA project and the investment settings were aligned with the European context (EU-/EUR), with cooling water utility assumptions consistent between the simulator and the TEA. Because APEA v14 uses a pricing basis for April 2022, capital costs were increased to 2024 using the respective CEPCI; working capital (10% of total capital) was added prior to annualization. Annual CAPEX was computed with the capital recovery factor (20-year life; interest rate within 0.07–0.11 according to the literature; 0.09 used in this case). These adjustments and equations are retained in the SLC worksheet that uses APEA's executive summary outputs (Total Project Capital Cost, Total Operating Cost) as inputs.

For the 86 TPD base case, equipment-level direct costs show (i) PFHXs as the largest subsystem (M€ 19.34) after design refinements in the cryogenic loop, and (ii) compressors as the single largest unit class, with MRC1 M€ 9.23. On the operating side, total annual OPEX M€ 28.05, of which utilities account for 72.8%; the annual spend splits to roughly 33% CAPEX and 67% OPEX. The integrated design achieves a SEC value of $6.983 \text{ kW h kg}^{-1}_{\text{LH}_2}$ and $\text{SLC} = 1.51 \text{ €/kg}_{\text{LH}_2}$ in the baseline (electricity at 0.10 €/kW h), improving on prior Brayton figures. Furthermore, the scenario in which power recovery is included is also analyzed, resulting in a 0.02 € cheaper SLC value ($1.49 \text{ €/kg}_{\text{LH}_2}$), which translates into almost 600,000.00€ saved per year, a reduction of 2.72% in the SEC value ($6.796 \text{ kW h kg}^{-1}_{\text{LH}_2}$), and an exergy efficiency of 44.35%.

Sensitivity studies identify the dominant economic levers. Electricity price drives SLC nearly linearly; SLC reaches $€2.24\text{--}2.65/\text{kg}_{\text{LH}_2}$ at $0.20\text{--}0.25 \text{ €/kW h}$ and approaches $€1/\text{kg}_{\text{LH}_2}$ only at very low tariffs (0.035 €/kW h). Financing terms affect only the annualized CAPEX: increase in the interest rate (0.07–0.11) raises the annual CAPEX by 33% and the SLC by 10%, while extending the payment period (10 to 30 years) reduces the annual CAPEX by 37.5%, although with an almost null effect on SLC beyond 22 years (0.01 € less for every two years). Together, these results confirm that cheap, stable electricity, favorable financing (low interest rate/adequate tenor), front-end compression strategy, and heat exchanger optimization are the principal feasibility drivers.

Scope boundaries and data handling were kept explicit: items not sized in detail (inter-and-aftercoolers) are treated according to the APEA defaults, while other components such as valves and mixers are not taken into account in the APEA output. PFHX geometries from EDR feed back into APEA for consistent OSBL/ISBL allowances and ΔT constraints. Similarly, compressors and turbines have also been adapted to their respective constraints and tolerances. This end-to-end chain, from HYSYS to EDR to APEA, with CEPCI escalation and annualization, provides a transparent and reproducible basis for SLC and for comparing design options at conceptual stage.

7.2. Recommendations

The recommendations consolidate actionable future steps to strengthen the technical credibility and economic robustness of the large-scale Brayton H_2 liquefaction concept developed in this thesis.

1. Capacity scale-up and benchmarking against Claude-cycle liquefier

- This thesis presents a liquefier with a capacity of 86 TPD. It is possible, and insightful, to produce a consistent scale-up set at 125, 250, and 500 TPD for the Brayton concept under identical costing rules, pressure levels, utility prices, and loss assumptions, allowing for a comparison with the Claude cycle (developed by Tamarona et al. [63]). Report SLC and SEC against capacity with and without power recovery, and discuss direct scale-up.

2. Native ortho–para (o–p) conversion in Aspen HYSYS

- Another potential improvement would be to implement o–p conversion natively inside HYSYS to remove the need for modifying REFPROP .FLD files and iterative external coupling. Retaining the validated kinetic form but eliminate file edits to improve reproducibility, model portability, and auditability of heat-release coupling across the PFHX network. The current approach relied on REFPROP integration and .FLD modification (pseudo-equilibrium heat capacity), which should be replaced by an explicit kinetic source term in the HYSYS flow-sheet. This is possible in Aspen HYSYS V14 by adding a Conversion Reactor unit operation, defining inlets and outlets, and setting up the conversion [108].

3. Thermophysical data for He–Ne (Nelum) at cryogenic conditions

- Furthermore, there is area for improvement within the PFHX model by O'Neill et al. [22]. Currently, the transport properties use ideal gas transport coefficients. A solution for their better prediction would be using the entropy-scaling method or experimentally fitted correlations for viscosity and thermal conductivity of He–Ne mixtures at cryogenic conditions.

4. Extend the O'Neill catalytic PFHX model to multistream

- Generalize the reactive PFHX model from two streams to 3–4 or more streams to reflect the real thermal integration (reactant H_2 , He–Ne coolant, and excess cold H_2 used as an auxiliary coolant). Move from a 2×2 to 4×4 segment-wise formulation; this will better predict o–p conversion, temperature cross-overs, and pressure-drop, thus de-risking sizing and catalyst loading. Tamarona et al. [63] explicitly recommend the construction of a steady-state continuum kinetic model for three or four PFHXs filled with catalyst.

5. Prioritize PFHX optimization (largest CAPEX driver)

- As PFHXs have become the most expensive component group in the liquefier after optimizing the He–Ne compression unit, a further optimization study can be done in the heat exchangers. This can be achieved by aiming to have the least amount of units in parallel, aiming to precisely distribute ΔT through all the PFHXs while ensuring enough heating/cooling duty; with the purpose of having smaller final sizes.

6. Integration with global H_2 deployment strategies

- Market studies by the Hydrogen Council, IRENA, and the IEA show that achieving competitive LH_2 costs ($<1.5 \text{ €}/\text{kg}$) is essential for large-scale trade and long-distance transport [1, 93, 109]. The Brayton-cycle concept aligns with these targets but requires coupling with low-cost renewable electricity and supportive policy frameworks to reach commercial viability. Embedding techno-economic models within broader H_2 value-chain analyses would provide a more realistic assessment of its role in global supply chains.

7. Boil-off management and infrastructure readiness

- Effective management of boil-off gas (BOG) is critical for maintaining the economic and technical viability of liquid H_2 supply chains. Literature indicates that uncontrolled ortho–para conversion and heat ingress can lead to daily losses of stored LH_2 , which significantly impacts long-distance transport economics. While large-scale shipping concepts (Suiso Frontier [110]) demonstrate that advanced insulation can reduce losses, further improvements in boil-off reliquefaction, vent gas utilization, and integration with port-side recovery systems are needed. Extending the Brayton liquefaction framework to include BOG reliquefaction or power generation pathways would enhance the robustness of techno-economic assessments. Coordinating liquefier design with downstream storage and transport systems is therefore essential to ensure that infrastructure developments can minimize evaporative losses and support reliable LH_2 delivery at scale.

In conclusion, this thesis shows that a large-scale, high-pressure H_2 Brayton liquefaction concept, integrating catalytic ortho–para conversion in a multistream PFHX network, reaches an SLC value of $1.49 \text{ €}/\text{kg}_{LH_2}$, with the potential to approach the targeted SLC of $1.00 \text{ €}/\text{kg}_{LH_2}$ under favorable electricity prices ($0.037 \text{ €}/\text{kW h}$) and offers a credible pathway for cost-effective H_2 transport and storage.

The techno-economic results highlight electricity price as the dominant driver of SLC, with PFHXs and compressors constituting the principal CAPEX contributors.

Building on this framework, immediate improvements should focus on (i) native implementation of o–p kinetics within Aspen HYSYS, (ii) validated thermophysical and transport properties for He–Ne mixtures at cryogenic conditions, and (iii) extension of the O'Neill PFHX model to true multistream operation. Finally, scaling the design to 125, 250, and 500 TPD will enable rigorous benchmarking against Claude-cycle baselines and sharpen the SEC/SLC–CAPEX trade-offs. Together, these advances will enhance predictive fidelity, narrow cost uncertainty, and accelerate the translation of large-scale hydrogen liquefaction into a viable element of future energy systems.

7.3. Safety Aspects

Last but not least, safety is a critical issue whenever working with H₂. This final section aims to inform the reader about its risks and hazards.

Hazards Associated with Liquefaction and Liquid Hydrogen: As previously discussed, the hydrogen liquefaction process is inherently energy-intensive and operates under severe cryogenic conditions. This, coupled with the unique properties of hydrogen, introduces a number of potential hazards:

- **Flammability and Explosivity:** Hydrogen has a wide flammability range (4–75%) and a low ignition energy (<0.02 μJ) [111]. Therefore, even small leaks can easily form flammable mixtures with air, with numerous potential ignition sources [111]. In confined or congested environments, hydrogen flames can accelerate rapidly, leading to Deflagration-Detonation Transition (DDT) with very high overpressures [111] safety.
- **Cryogenic Hazards:** The extremely low temperatures of LH₂ present significant risks. Skin contact can cause severe cold burns, similar to thermal burns [111]. Condensation and solidification of air at these temperatures can clog pipes and valves, and the formation of solid oxygen in the presence of LH₂ creates a shock-sensitive material with detonation potential [111]. Material compatibility with cryogenic temperatures and resistance to hydrogen embrittlement are crucial aspects in the design and operation of plants.
- **High Pressure Operations:** Some liquefaction processes involve high-pressure operations, increasing the potential release rate in the event of a leak. Pressure increases due to the vaporization of LH₂ or the ortho-para conversion can exceed the limits of the equipment, causing physical ruptures or explosions [111].
- **Hydrogen Embrittlement:** Hydrogen can cause embrittlement in some metals, leading to failure at stresses below the yield strength. Selecting compatible materials, such as 316 grade stainless steel, is critical [111].
- **Boil-off:** LH₂ is subject to boil-off. This leads to product losses and the need to safely manage the released gas through venting devices.

Safety Considerations and Risk Assessment: Effective safety management requires the identification and thorough understanding of hazards. The IDEALHY project conducted HAZID (Hazard Identification) for the new liquefaction process, storage, and transportation of LH₂. However, a review of available experimental data highlighted a general lack of relevant data for liquid hydrogen releases, especially at large scales [111]. Consequently, the validity of current mathematical models for predicting the consequences of such releases may be limited.

Advanced risk assessment methodologies, such as the combination of System-Theoretic Process Analysis (STPA) and Best-Worst Method (BWM), have been applied to identify system-level hazards and quantify the associated risk in hydrogen liquefaction systems [112]. This approach allows classification of hazards and identification of appropriate control actions and safety constraints.

Existing regulations and standards, such as DSEAR in the UK, provide a framework for controlling fire and explosion risks arising from the transport and storage of LH₂. However, there is a need for further development of specific standards and guidelines for liquid hydrogen, particularly with regard to safe separation distances in urban settings and material compatibility. Understanding the consequences of

LH₂ leaks and spills, the dispersion of the vaporized cold gas, and the behavior of hydrogen flames require further experimental research and model validation [111].

In conclusion, although liquid hydrogen offers significant advantages for hydrogen storage and transportation, it is imperative to rigorously and systematically address the multiple safety issues associated with LH₂ liquefaction and handling. Continued research, development of reliable predictive models, and implementation of robust risk management strategies are essential to ensure the safe deployment of this promising energy technology.

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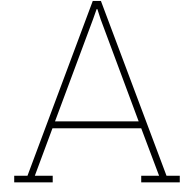
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Appendix A

Below, O'Neill et al. system condition for their ortho-para boundary value problem [22].

$$\frac{\partial x_p}{\partial z} = \frac{M_{H_2} r A_f}{\dot{m}_f}$$

$$\frac{\partial P_f}{\partial z} = -f \frac{\rho_f u_f^2}{2d_f}$$

$$\text{System conditions: } \frac{\partial T_f}{\partial z} = \frac{1}{\dot{m}_f c_{p,f}} \left(-\frac{\partial Q_{\text{trans}}}{\partial z} - \frac{\partial Q_{\text{conv}}}{\partial z} \right)$$

$$\frac{\partial P_c}{\partial z} = -f \frac{\rho_c u_c^2}{2D_h}$$

$$\frac{\partial T_c}{\partial z} = \frac{1}{\dot{m}_c c_{p,c}} \frac{\partial Q_{\text{transfer}}}{\partial z}$$

In the following, the MATLAB code calling REFPROP is displayed to model the ejector.

Listing A.1: Ejector modeling in MATLAB

```
1 clear all
2 clc
3 fluid='parahyd';
4 Pprimary=73.8*100; % pressure in kilopascals
5 Tprimary=33; %temperature in kelvin
6 hprimary=refpropm('h','t',Tprimary,'p',Pprimary,fluid);
7 sprimary=refpropm('s','t',Tprimary,'p',Pprimary,fluid);
8 xprimary=refpropm('q','p',Pprimary,'h',hprimary,fluid);
9
10 msecondary=0.3;
11 error2=1;
12 while error2>0.00001
13 Psecondary=110; %pressure in kilopascals
14 hsecondary=refpropm('h','p',Psecondary,'q',0,fluid);
15 Tsecondary=refpropm('T','p',Psecondary,'q',0,fluid);
16 ssecondary=refpropm('s','p',Psecondary,'q',0,fluid);
17 %xs=refpropm('q','p',ps,'h',hs,fluid);
18 mprimary=1;
19
20 massratio=msecondary/mprimary;
21 eta=0.2;
22
23 error=2;
```

```

24 pmin=Psecondary;
25 pmax=Pprimary;
26     %pressure in kilopascals
27 while abs(error)>0.00001
28     pdiffusor=(pmin+pmax)/2;
29     h_iss=refpropm('h','p',pdiffusor,'s',ssecondary,fluid);
30     h_isp=refpropm('h','p',pdiffusor,'s',sprimary,fluid);
31     etaCalc=massratio*(h_iss-hsecondary)/(hprimary-h_isp);
32
33     error=(etaCalc-eta);
34 if error>0
35     pmax=pdiffusor;
36 else
37     pmin=pdiffusor;
38 end
39 end
40
41 hdiffusor=(mprimary*hprimary+msecondary*hsecondary)/(mprimary+msecondary);
42 xdifusor=refpropm('q','p',pdiffusor,'h',hdiffusor,fluid);
43 Tdifusor=refpropm('T','p',pdiffusor,'h',hdiffusor,fluid);
44 pfinal=100; %pressure in kilopascals equal to 1 bar
45 xfial=refpropm('q','p',100,'h',hdiffusor,fluid);
46
47 msNew=(mprimary+msecondary)*xfial;
48 mpNew=(mprimary+msecondary)*(1-xfial);
49 ratioNew=msNew/mpNew;
50 error2=abs(ratioNew-massratio);
51 msecondary=ratioNew;
52 end

```

Below, the Valenti et al. REFPROP .FLD file for para-H₂ is displayed [102].

Listing A.2: PARAHYD REFPROP .FLD Content

```

1 parahydrogen      !short name
2 1333-74-0p        !CAS number
3 parahydrogen      !full name
4 H2                !chemical formula
5 R-702p            !synonym
6 2.01588           !molecular weight [g/mol]
7 13.8033           !triple point temperature [K]
8 20.271            !normal boiling point [K]
9 32.938            !critical temperature [K]
10 1285.8           !critical pressure [kPa]
11 15.538           !critical density [mol/L]
12 -0.219           !acentric factor
13 0.0              !dipole moment [Debye]
14 MBP              !default reference state
15 8.0              !version number
16 1049             !UN Number
17 other            !family
18 285.83           !heating value (gross or superior) [kJ/mol]
19
20
21 ! compiled by E.W. Lemmon, NIST Physical and Chemical Properties Division, Boulder, Colorado
22 ! 04-06-98 EWL, original version
23 ! 11-13-98 EWL, update format to version 6.1
24 ! 10-14-99 EWL, change ECS reference fluid to nitrogen
25 ! 10-20-99 EWL, add tcx and eta formulations from NIST12, Version 3.1
26 ! 11-02-99 EWL, reduce sigma0 by factor of 1000.
27 ! 11-10-99 EWL, add extra digits to melting equation to get ptp at ttp
28 ! 03-20-00 EWL, change max density to the density at p=2*pmax along melting line
29 ! 11-06-00 EWL, switch transport equations to hardcoded models
30 ! 03-21-02 MLH, put LJ parameters in ECS transport
31 ! 09-13-02 EWL, changed dmax to 44. See comments in TRNS_VIS under ETAH2.
32 ! 08-05-04 EWL, add Harvey and Lemmon dielectric correlation
33 ! 03-27-07 JW, add equation of state of Leachman et al. (2009)
34 ! 10-16-07 EWL, change upper temp. limit of vis and tcx from 400 to 1000 K
35 ! based on good graphical extrapolation
36 ! 07-17-08 EWL, change incorrect molecular weight from 2.01594 to 2.01588
37 ! 07-21-08 EWL, add vapor pressure ancillary

```

```

38 ! 06-17-10 CKL, add ancillaries for sat. densities
39
40
41 #EOS                      !equation of state specification
42 FEQ Helmholtz equation of state for parahydrogen of Leachman et al. (2009).
43 ?LITERATURE REFERENCE \
44 ?Leachman, J.W., Jacobsen, R.T, Penoncello, S.G., Lemmon, E.W.
45 ?"Fundamental Equations of State for Parahydrogen, Normal Hydrogen, and
46 ?Orthohydrogen,"
47 ?J. Phys. Chem. Ref. Data, 38(3):721-748, 2009.
48 ?\
49 ?The uncertainty in density is 0.1% at temperatures from the triple point
50 ?to 250 K and at pressures up to 40 MPa, except in the critical region,
51 ?where an uncertainty of 0.2% in pressure is generally attained. In the
52 ?region between 250 and 450 K and at pressures to 300 MPa, the
53 ?uncertainty in density is 0.04%. At temperatures between 450 and 1000
54 ?K, the uncertainty in density increases to 1%. At pressures between 300
55 ?and 2000 MPa, the uncertainty in density is 8%. Speed of sound data are
56 ?represented within 0.5% below 100 MPa. The estimated uncertainty for
57 ?heat capacities is 1.0%. The estimated uncertainties of vapor pressures
58 ?and saturated liquid densities calculated using the Maxwell criterion
59 ?are 0.1% for each property.
60 ?\
61 !end of info section
62 13.8033                    !lower temperature limit [K]
63 1000.0                    !upper temperature limit [K]
64 2000000.0                 !upper pressure limit [kPa]
65 104.0                     !maximum density [mol/L]
66 CPP                      !pointer to Cp0 model
67 2.01588                   !molecular weight [g/mol]
68 13.8033                   !triple point temperature [K]
69 7.041                     !pressure at triple point [kPa]
70 38.185                    !density at triple point [mol/L]
71 20.271                    !normal boiling point temperature [K]
72 -0.219                   !acentric factor
73 32.938                    1285.8    15.538    !Tc [K], pc [kPa], rhoc [mol/L]
74 32.938                    15.538    !reducing parameters [K, mol/L]
75 8.314472                 !gas constant [J/mol-K]
76 9 4 5 12 0 0            !# terms, # coeff/term for: "normal" terms, critical,
77 spare
78 -0.733375D+01 0.6855 1.0 0.0 !a(i),t(i),d(i),l(i)
79 0.100000D-01 1.000 4.0 0.0
80 0.260375D+01 1.000 1.0 0.0
81 0.466279D+01 0.489 1.0 0.0
82 0.682390D+00 0.774 2.0 0.0
83 -0.147078D+01 1.133 2.0 0.0
84 0.135801D+00 1.386 3.0 0.0
85 -0.105327D+01 1.619 1.0 1.0
86 0.328239D+00 1.162 3.0 1.0
87 -0.577833D-01 3.960 2.0 2.0 2.0 -1.7437 -0.1940 0.8048 1.5487 0. 0. 0.
88 0.449743D-01 5.276 1.0 2.0 2.0 -0.5516 -0.2019 1.5248 0.1785 0. 0. 0.
89 0.703464D-01 0.990 3.0 2.0 2.0 -0.0634 -0.0301 0.6648 1.2800 0. 0. 0.
90 -0.401766D-01 6.791 1.0 2.0 2.0 -2.1341 -0.2383 0.6832 0.6319 0. 0. 0.
91 0.119510D+00 3.190 1.0 2.0 2.0 -1.7770 -0.3253 1.4930 1.7104 0. 0. 0.
92
93 #AUX                      !auxiliary model specification
94 CPP ideal gas heat capacity function
95 ?LITERATURE REFERENCE \
96 ?Leachman, J.W., Jacobsen, R.T, Penoncello, S.G., Lemmon, E.W.
97 ?"Fundamental Equations of State for Parahydrogen, Normal Hydrogen, and
98 ?Orthohydrogen,"
99 ?J. Phys. Chem. Ref. Data, 38(3):721-748, 2009.
100 ?\
101 !end of info section
102 13.8033                    !lower temperature limit [K]
103 1000.0                    !upper temperature limit [K]
104 0.0                      !upper pressure limit [kPa]
105 0.0                      !maximum density [mol/L]
106 1.0                      8.314472    !reducing parameters for T, Cp0
107 1 7 0 0 0 0 0            !Nterms: polynomial, exponential, cosh, sinh

```

```

108 2.5      0.0
109 57.207   204.9
110 -66.923  227.5
111 10.745   345.5
112 1.2206   1025.4
113 -1.2785   1191.9
114 1.2527   5398.8
115 0.9175   10185.0
116
117
118 #AUX      !auxiliary model specification
119 PH0 Helmholtz form for the ideal-gas state
120 ?LITERATURE REFERENCE \
121 ?Leachman, J.W., Jacobsen, R.T, Penoncello, S.G., Lemmon, E.W.
122 ?"Fundamental Equations of State for Parahydrogen, Normal Hydrogen, and
123 ?Orthohydrogen,"
124 ?J. Phys. Chem. Ref. Data, 38(3):721-748, 2009.
125 ?\
126 !end of info section
127 13.8033    !lower temperature limit [K]
128 1000.0      !upper temperature limit [K]
129 0.0         !upper pressure limit [kPa]
130 0.0         !maximum density [mol/L]
131 1 2 7 0 0 0 0 0 !Nterms: ai*log(tau**ti); ai*tau**ti; ai*log(1-exp(bi*tau))
132 1.5000000000 1.0000000000 !ai, ti for [ai*log(tau**ti)] terms
133 -1.4485891134 0.0000000000 !aj, ti for [ai*tau**ti] terms
134 1.8845212390 1.0000000000
135 4.3025600000 -15.1496751472 !aj, ti for [ai*log(1-exp(ti*tau))] terms
136 13.0289000000 -25.0925982148
137 -47.7365000000 -29.4735563787
138 50.0013000000 -35.4059141417
139 -18.6261000000 -40.7249984820
140 0.9939730000 -163.7925799988
141 0.5360780000 -309.2173173842
142
143
144 @EOS      !equation of state specification
145 BWR MBWR equation of state for parahydrogen of Younglove (1982).
146 ?LITERATURE REFERENCE \
147 ?Younglove, B.A.,
148 ? "Thermophysical Properties of Fluids. I. Argon, Ethylene,
149 ? Parahydrogen, Nitrogen, Nitrogen Trifluoride, and Oxygen,"
150 ? J. Phys. Chem. Ref. Data, Vol. 11, Suppl. 1, pp. 1-11, 1982.
151 ?\
152 ?The uncertainties in density are 0.1% in the liquid phase, 0.25% in the
153 ?vapor phase, and 0.2% in the supercritical region. The uncertainty in
154 ?heat capacity is 3% and the uncertainty in speed of sound is 2% in the
155 ?liquid phase and 1% elsewhere.
156 ?\
157 ?Note:\
158 ?The ideal gas equation was refit from calculations of piecewise Cp0 equation
159 ?of McCarty
160 ?\
161 !end of info section
162 13.8      !lower temperature limit [K]
163 400.0      !upper temperature limit [K]
164 121000.0   !upper pressure limit [kPa]
165 44.0       !maximum density [mol/L]
166 CP1      !pointer to Cp0 model
167 2.01594    !molecular weight [g/mol]
168 13.8      !triple point temperature [K]
169 7.042      !pressure at triple point [kPa]
170 38.21      !density at triple point [mol/L]
171 20.2769    !normal boiling point temperature [K]
172 -0.218     !acentric factor
173 32.938     1283.77 15.556 !Tc [K], pc [kPa], rhoc [mol/L]
174 32.938     15.556 !reducing parameters [K, mol/L]
175 15.6173762 !gamma
176 0.0831434  !gas constant [L-bar/mol-K]
177 32 1       !Nterm, Ncoeff per term
178 0.4675528393416d-03 0.4289274251454d-01 -0.5164085596504d-00

```

```

179 0.2961790279801d+01 -0.3027194968412d+02 0.1908100320379d-04
180 -0.1339776859288d-02 0.3056473115421d-00 0.5161197159532d+02
181 0.1999981550224d-06 0.2896367059356d-03 -0.2257803939041d-01
182 -0.2287392761826d-05 0.2446261478645d-04 -0.1718181601119d-02
183 -0.5465142603459d-06 0.4051941401315d-08 0.1157595123961d-05
184 -0.1269162728389d-07 -0.4983023605519d+02 -0.1606676092098d+03
185 -0.1926799185310d-00 0.9319894638928d+01 -0.3222596554434d-03
186 0.1206839307669d-02 -0.3841588197470d-06 -0.4036157453608d-04
187 -0.1250868123513d-09 0.1976107321888d-08 -0.2411883474011d-12
188 -0.4127551498251d-12 0.8917972883610d-11
189
190
191 #AUX !auxiliary model specification
192 CP1 ideal gas heat capacity function
193 ?LITERATURE REFERENCE \
194 ?refit by EWL from calculations of piecewise fit of Cp0 by McCarty
195 ?\
196 !end of info section
197 13.8 !lower temperature limit [K]
198 400.0 !upper temperature limit [K]
199 0.0 !upper pressure limit [kPa]
200 0.0 !maximum density [mol/L]
201 1.0 8.31434 !reducing parameters for T, Cp0
202 6 4 0 0 0 0 0 !Nterms: polynomial, exponential, cosh, sinh
203 0.24995169d+01 0.00
204 -0.11125185d-02 1.00
205 0.27491461d-03 1.50
206 -0.10005269d-04 2.00
207 0.22695404d-08 3.00
208 -0.21031029d-12 4.00
209 0.12353388d+02 598.00
210 -0.17777676d+02 778.00
211 0.64309174d+01 1101.00
212 0.73347521d+01 6207.00
213
214
215 #TCX !thermal conductivity model specification
216 TC0 pure fluid thermal conductivity model of McCarty and Weber (1972).
217 ?LITERATURE REFERENCE \
218 ?McCarty, R.D. and Weber, L.A.,
219 ? "Thermophysical properties of parahydrogen from the freezing liquid line to
220 ? 5000 R for pressures to 10,000 psia,"
221 ? Natl. Bur. Stand., Tech. Note 617, 1972.
222 ?\
223 ?The uncertainty in thermal conductivity below 100 K is estimated to be 3%
224 ?below 150 atm and up to 10% below 700 atm. For temperatures around 100 K
225 ?at low densities, the uncertainty is about 1%. Above 100 K, the uncertainty
226 ?is estimated to be on the order of 10%.
227 ?\
228 !end of info section
229 13.8 !lower temperature limit [K]
230 1000.0 !upper temperature limit [K]
231 121000.0 !upper pressure limit [kPa]
232 66.94 !maximum density [mol/L]
233 H2 !pointer to hardcoded thermal conductivity model
234 9 9 0 0 0 0 0 !number of terms for various pieces
235 1.0 1.0 1.0 !reducing parameters for T, rho, eta
236 .5878519061667D3 0. 0. 0. 0 !dilute gas coeffs
237 -.1182164836398D4 0. 0. 0. 0
238 .9115082518407D3 0. 0. 0. 0
239 -.2936280009103D3 0. 0. 0. 0
240 -.4946317819761D1 0. 0. 0. 0
241 .3299549945380D2 0. 0. 0. 0
242 -.1016429439989D2 0. 0. 0. 0
243 .1335625725585D1 0. 0. 0. 0
244 -.6700700939493D-1 0. 0. 0. 0
245 1. 0. 0. 0. 0 !OP para fraction
246 -.1443670778633D2 0. 0. 0. 0 !residual coeffs
247 .4951795243651D-3 0. 0. 0. 0
248 .1015887358687D2 0. 0. 0. 0
249 -.3043737455575D0 0. 0. 0. 0

```

```

250 -.3776977405391D2      0. 0. 0. 0
251 .7750113657427D1      0. 0. 0. 0
252 .7015931751234D-2     0. 0. 0. 0
253 .1555600000000D2      0. 0. 0. 0
254 NUL                    !pointer to critical enhancement auxiliary function
255
256
257 #ETA                    !viscosity model specification
258 VSO pure fluid viscosity model of McCarty and Weber (1972).
259 ?LITERATURE REFERENCE \
260 ?McCarty, R.D. and Weber, L.A.,
261 ? "Thermophysical properties of parahydrogen from the freezing liquid line to
262 ? 5000 R for pressures to 10,000 psia,"
263 ? Natl. Bur. Stand., Tech. Note 617, 1972.
264 ?\
265 ?The uncertainty in viscosity ranges from 4% to 15%.
266 ?\
267 !end of info section
268 13.8                    !lower temperature limit [K]
269 1000.0                  !upper temperature limit [K]
270 121000.0                !upper pressure limit [kPa]
271 66.94                   !maximum density [mol/L]
272 H2                      !pointer to hardcoded thermal conductivity model
273 0 9 8 0 0 0 0 0        !number of terms for various pieces
274 1.0      1.0      1.0      !reducing parameters for T, rho, eta
275 -0.1841091042788D2     0. 0. 0. 0 !dilute gas coeffs
276 0.3185762039455D2     0. 0. 0. 0
277 -0.2308233586574D2     0. 0. 0. 0
278 0.9129812714730D1     0. 0. 0. 0
279 -0.2163626387630D1     0. 0. 0. 0
280 0.3175128582601D0     0. 0. 0. 0
281 -0.2773173035271D-1    0. 0. 0. 0
282 0.1347359367871D-2    0. 0. 0. 0
283 -0.2775671778154D-4    0. 0. 0. 0
284 -0.1099981128000D2     0. 0. 0. 0 !residual coeffs
285 0.1895876508000D2     0. 0. 0. 0
286 -0.3813005056000D3     0. 0. 0. 0
287 0.5950473265000D2     0. 0. 0. 0
288 0.1099399458000D1      0. 0. 0. 0
289 0.8987269839000D1      0. 0. 0. 0
290 0.1231422148000D4      0. 0. 0. 0
291 0.3110000000000D0      0. 0. 0. 0
292 NUL                    !pointer to critical enhancement auxiliary function
293
294
295 @TRN                    !transport model specification
296 ECS Extended Corresponding States model (Nitrogen reference); predictive mode.
297 ?LITERATURE REFERENCES \
298 ?Klein, S.A., McLinden, M.O., and Laesecke, A.,
299 ? "An improved extended corresponding states method for estimation of
300 ? viscosity of pure refrigerants and mixtures,"
301 ? Int. J. Refrigeration, 20:208-217, 1997.
302 ?\
303 ?McLinden, M.O., Klein, S.A., and Perkins, R.A.,
304 ? "An extended corresponding states model for the thermal conductivity
305 ? of refrigerants and refrigerant mixtures,"
306 ? Int. J. Refrigeration, 23:43-63, 2000.
307 ?\
308 ?Lennard-Jones parameters are for h2 from Reid, Prausnitz, and Poling,
309 ? Properties of Gases and Liquids, 4th ed, McGraw-Hill (1987)
310 ?\
311 !end of info section
312 13.8                    !lower temperature limit [K]
313 400.0                   !upper temperature limit [K]
314 121000.0                !upper pressure limit [kPa]
315 66.94                   !maximum density [mol/L]
316 FEQ nitrogen.fld
317 VS1                     !model for reference fluid viscosity
318 TC1                     !model for reference fluid thermal conductivity
319 1                        !Lennard-Jones flag (0 or 1) (0 => use estimates)
320 2.827                   !Lennard-Jones coefficient sigma [nm] for ECS method

```

```

321 59.7          !Lennard-Jones coefficient epsilon/kappa [K] for ECS method
322 1 0 0          !number of terms in f_int term in Eucken correlation, spare1,
      spare2
323 1.32d-3        0.0 0.0 0.0 !coeff, power of T, spare 1, spare 2
324 1 0 0          !number of terms in psi (visc shape factor): poly,spare1,spare2
325 1.0            0.0 0.0 0.0 !coeff, power of Tr, power of Dr, spare
326 1 0 0          !number of terms in chi (t.c. shape factor): poly,spare1,spare2
327 1.0            0.0 0.0 0.0 !coeff, power of Tr, power of Dr, spare
328 NUL            !pointer to critical enhancement auxiliary function
329
330
331 #STN            !surface tension specification
332 ST1 surface tension model of McCarty et al. (1981).
333 ?LITERATURE REFERENCE \
334 ?McCarty, R.D., Hord, J., and Roder, H.M.,
335 ? "Selected Properties of Hydrogen (Engineering Design Data),"
336 ? NBS Monograph 168, National Bureau of Standards, Boulder, 1981.
337 ?\
338 !end of info section
339 13.8            !lower temperature limit [K]
340 32.976          !upper temperature limit [K]
341 0.0             !dummy upper pressure limit
342 0.0             !dummy maximum density
343 1               !number of terms in surface tension model
344 32.976          !critical temperature used in fit (dummy)
345 0.005328 1.065 !sigma0 and n
346
347
348 #DE             !dielectric constant specification
349 DE3 dielectric constant model of Harvey and Lemmon (2005).
350 ?LITERATURE REFERENCE \
351 ?Harvey, A.H. and Lemmon, E.W.
352 ? "Method for Estimating the Dielectric Constant of Natural Gas Mixtures,"
353 ? Int. J. Thermophys., 26(1):31-46, 2005.
354 ?\
355 !end of info section
356 0.0             !lower temperature limit [K]
357 2000.0          !upper temperature limit [K]
358 0.0             !dummy upper pressure limit
359 0.0             !dummy maximum density
360 273.16 1000.0 1.0 !reducing parameters for t and d
361 0 2 3 0 0 0      !number of terms in dielectric constant model
362 2.0297          0. 1. 0. !coef, t exp, d exp
363 0.0069          1. 1. 0. !coef, t exp, d exp
364 0.181           0. 2. 0.
365 0.021           1. 2. 0.
366 -7.4            0. 3. 0.
367
368
369 #MLT            !melting line specification
370 MLP melting line model of Younglove (1982).
371 ?LITERATURE REFERENCE \
372 ?Younglove, B.A.,
373 ? "Thermophysical Properties of Fluids. I. Argon, Ethylene,
374 ? Parahydrogen, Nitrogen, Nitrogen Trifluoride, and Oxygen,"
375 ? J. Phys. Chem. Ref. Data, Vol. 11, Suppl. 1, pp. 1-11, 1982.
376 ?\
377 !end of info section
378 13.8            !lower temperature limit [K]
379 400.0           !upper temperature limit [K]
380 0.0             !dummy upper pressure limit
381 0.0             !dummy maximum density
382 1.              !reducing temperature and pressure
383 4 0 0 0 0 0      !number of terms in melting line equation
384 -0.265289115d2 0. !coefficients and exponents
385 0.248578596d0 0.1764739d1
386 -0.21272389d2 0.
387 0.125746643d0 0.1955000d1
388
389
390 #SBL            !sublimation line specification

```

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391 SB2 sublimation line model of McCarty et al. (1981)
392 ?LITERATURE REFERENCE \
393 ?McCarty, R.D., Hord, J., and Roder, H.M.,
394 ? "Selected Properties of Hydrogen (Engineering Design Data),"
395 ? NBS Monograph 168, National Bureau of Standards, Boulder, 1981.
396 ?\
397 !end of info section
398 13.8 !lower temperature limit [K]
399 13.8 !upper temperature limit [K]
400 0.0 !(dummy) upper pressure limit
401 0.0 !(dummy) maximum density
402 1. 0.13332237 !reducing temperature and pressure
403 2 0 1 0 0 0 !number of terms in sublimation line equation
404 4.009857354 0.0 !coefficients and exponents
405 -90.77568949 -1.0
406 2.489830940 1.0
407
408
409 #PS !vapor pressure equation
410 PS5 vapor pressure equation
411 ?LITERATURE REFERENCE \
412 ?Leachman, J.W., Jacobsen, R.T, Penoncello, S.G., Lemmon, E.W.
413 ?"Fundamental Equations of State for Parahydrogen, Normal Hydrogen, and
414 ?Orthohydrogen,"
415 ?J. Phys. Chem. Ref. Data, 38(3):721-748, 2009.
416 ?\
417 !end of info section
418 13.8033 !lower temperature limit [K]
419 32.938 !upper temperature limit [K]
420 0.0 !(dummy) upper pressure limit
421 0.0 !(dummy) maximum density
422 32.938 1285.8 !reducing parameters
423 4 0 0 0 0 0 !number of terms in equation
424 -0.487767D+01 1.0000000
425 0.103359D+01 1.5000000
426 0.826680D+00 2.6500000
427 -0.129412D+00 7.4000000
428
429
430 #DL !saturated liquid density equation
431 DL1 saturated liquid density equation of Lemmon (2010).
432 ?LITERATURE REFERENCE \
433 ?Lemmon, C.K. and Lemmon, E.W., 2010.
434 ?\
435 !end of info section
436 13.8033 !lower temperature limit [K]
437 32.938 !upper temperature limit [K]
438 0.0 !(dummy) upper pressure limit
439 0.0 !(dummy) maximum density
440 32.938 15.538 !reducing parameters
441 5 0 0 0 0 0 !number of terms in equation
442 -0.13509D+00 0.15 !coefficients and exponents
443 0.40739D+01 0.44
444 -0.53985D+01 0.7
445 0.55230D+01 0.99
446 -0.23643D+01 1.31
447
448
449 #DV !saturated vapor density equation
450 DV3 saturated vapor density equation of Lemmon (2010).
451 ?LITERATURE REFERENCE \
452 ?Lemmon, C.K. and Lemmon, E.W., 2010.
453 ?\
454 !end of info section
455 13.8033 !lower temperature limit [K]
456 32.938 !upper temperature limit [K]
457 0.0 !(dummy) upper pressure limit
458 0.0 !(dummy) maximum density
459 32.938 15.538 !reducing parameters
460 6 0 0 0 0 0 !number of terms in equation
461 -0.57545D+01 0.53 !coefficients and exponents

```

```
462 0.38153D+01 0.7
463 -0.12293D+02 1.7
464 0.15095D+02 2.4
465 -0.17295D+02 3.3
466 -0.34190D+02 10.
467
468
469 @END
470 c 1 2 3 4 5 6 7 8
471 c234567890123456789012345678901234567890123456789012345678901234567890
```

B

Appendix B

Figure B.1 and Figure B.2 show a planning visualization as per March 30th, 2025. Further changes will be made as the Thesis develops.

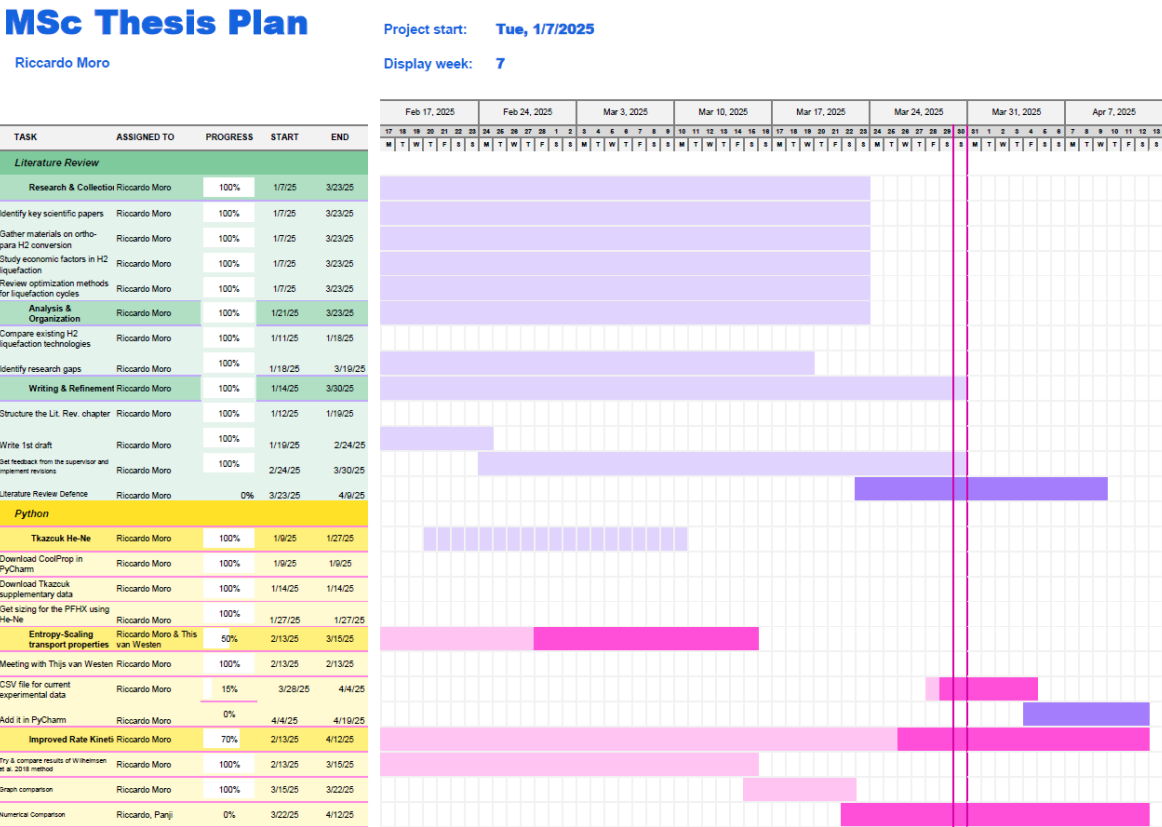


Figure B.1: Gantt chart of the project timeline (Page 1)

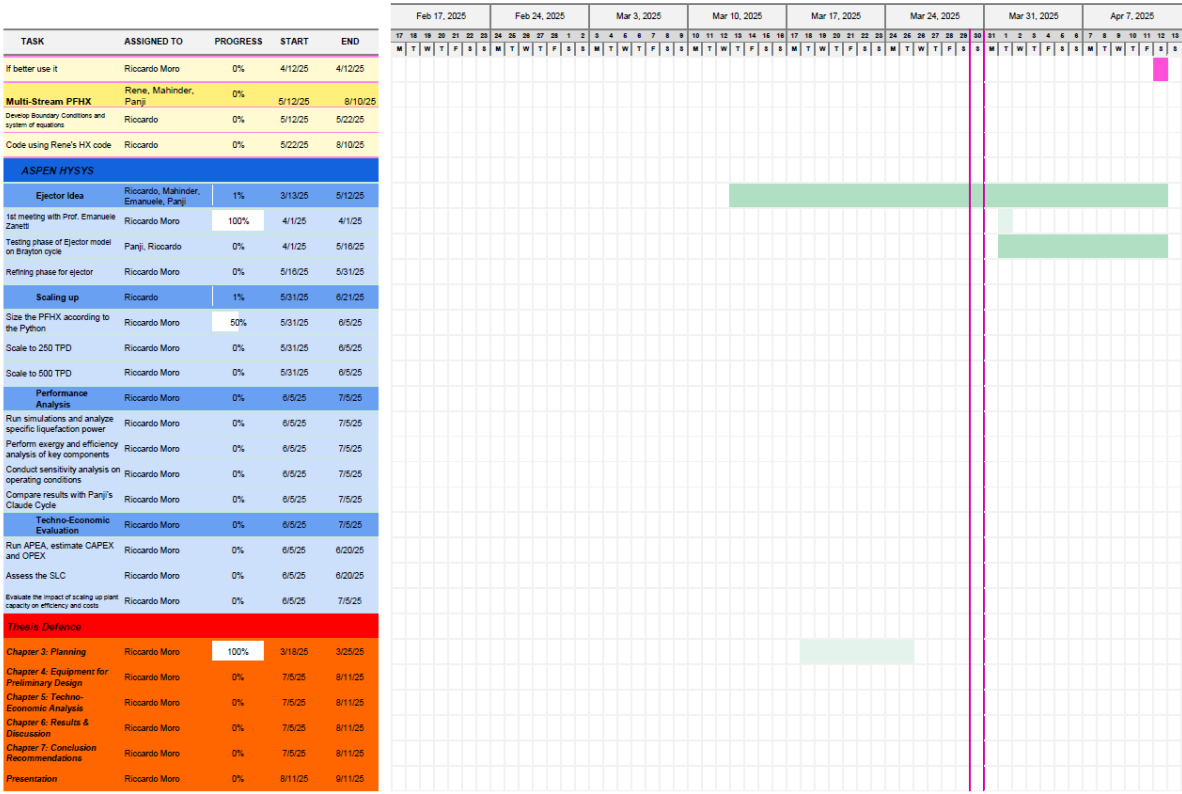


Figure B.2: Gantt chart of the project timeline (Page 2)