Hydrogen Liquefaction
Life Cycle Analysis

T. A. van Oeveren

Timman A. van Oeveren

Student Number: 4551710

Faculty:	Mechanical Engineering
Department:	Process & Energy
Track:	Energy, Flow & Process Technology

Thesis Supervisors & Comittee Members:

Dr.ir. Othon Moutlos,	TU Delft
Dr.ir. Mahinder Ramdin,	TU Delft
Dr.ir. Joost Vogtländer,	TU Delft

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Abbreviations

BOG	Boil-Off Gas
CML	Centrum voor Milieukunde Leiden (Center for Environmental Sciences Leiden)
GWP	Global Warming Potential
IDEMAT	Integrated Design and Engineering Materials Assessment Tool
IPCC	Intergovernmental Panel on Climate Change
ISO	International Organization for Standardization
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
LNG	Liquefied Natural Gas
MR	Mixed Refrigerant
PV	Photovoltaic
SEC	Specific Energy Consumption
SMR	Steam Methane Reforming
TPD	Tons Per Day
LH_2	Liquid Hydrogen
CGH_2	Compressed Gaseous Hydrogen
LOHC	Liquid Organic Hydrogen Carriers
NH_3	Ammonia
ReCiPe	An impact assessment method that transforms a long list of life cycle inventory results into a limited number of indicator scores
Eco-Indicator 99	A method for life cycle impact assessment
SimaPro	Software for analyzing life cycle assessments
openLCA	Open source software for life cycle assessment
Ecoinvent	A database for life cycle inventory data
VDI	Verein Deutscher Ingenieure

1. Introduction

1.1 Liquid hydrogen and the liquefaction process

Hydrogen is seen as a clean energy carrier that will aid in phasing out fossil fuels. Life Cycle Assessment (LCA) studies have shown the environmental potential of adopting sustainable hydrogen-based technologies. Sustainable hydrogen can potentially replace fossil fuels in our vehicles. Hydrogen is already used in various applications such as chemical production and various other niche industries, but these still use primarily non-sustainable hydrogen [1], [2]. To support global environmental goals, the European Union is increasingly supporting sustainable hydrogen technology development. The complete hydrogen value chain has many different pathways, and global adoption will require expansive and multifaceted infrastructure [3]. Major seaports, with their already robust infrastructure are being developed to become hydrogen hubs. The Port of Rotterdam, as an example, has numerous companies that are actively investing into the hydrogen economy within this port through various ongoing plans and projects. Prominent examples are plans for a hydrogen pipe network [4] and a 200 MW Electrolyzer [5][6].

Liquid hydrogen (LH_2) will play a key role in this global hydrogen economy, according to researchers [7]. The use cases of LH_2 in the hydrogen value chain are its long-distance transport and storage capabilities [8]. Like Liquid Natural Gas (LNG), LH_2 can be efficiently transported over long distances by ship, due from its high gravimetric density at atmospheric pressure. This density is approximately 1.5–2.0 times greater than that of compressed hydrogen which is typically stored at pressures up to 700 bar. Moreover, storing hydrogen as a liquid at atmospheric pressure significantly enhances the safety of storage and transport vessels compared to high-pressure storage [9]. Additionally, LH_2 is non-corrosive [10], enabling the use of containment vessels made from stainless steel or aluminum alloys that will not degrade easily. Also, LH_2 has a very high purity, which is a requirement for many of its applications.

Despite the potential of LH_2, its adoption faces major challenges. The liquefaction process is highly energy-intensive due to the need to cool hydrogen to -253°C. Roughly 30% to 40% of the net heating value is lost during liquefaction [11], [12]. A simplified schematic diagram of a basic hydrogen liquefaction cycle is shown in Figure 1.1. Hydrogen gas enters the system to be liquefied. The conditions of the feed gas can vary, so it is vital that it is compressed and purified to the required conditions for this thermodynamic cycle. Then, there are two stages of cooling, i.e., pre-cooling and cryogenic cooling. Distinction is made between these two phases due to different refrigerants being used, which are optimized for thermodynamic efficiency. Inside the heat exchangers, catalysts ensure that ortho-to-para conversion of hydrogen takes place, which reduces boil-off gas (BOG) during storage after the hydrogen has been liquefied and stored [13].

For both storage and transport, the phenomenon of BOG in LH_2 storage remains a significant efficiency issue. During storage, a daily 1.5%–3.0% of LH_2 is vaporized. This BOG effect occurs due to ortho-to-para conversion, heat leak, thermal stratification, flashing, and sloshing during transport [14]. A large effort is being made to increase the thermodynamic efficiency of hydrogen liquefiers, as this would reduce both economic and environmental costs [13]. Despite the large amount of literature, researchers note that there has been a noticeable lack of understanding of the environmental impacts of hydrogen liquefaction. This step is often overlooked and simplified, leading to potentially incorrect environmental impact assessments. The usual target for liquefaction research is optimizing the specific exergy consumption (SEC) of the thermodynamic cycle. This SEC addresses only the thermodynamic efficiency of the liquefaction cycle. Using solely the SEC to address environmental impacts neglects many other aspects of an industrial liquefaction plant, i.e., raw material costs [15], plant operation auxiliary electricity usage [16], and refrigerant production and leakage [10], [13]. Because of this lack of attention to climate aspects, the majority of previous LCA studies fail to properly address the impacts of hydrogen liquefaction. This Thesis aims to address this knowledge gap and provide clear guidelines for properly addressing and calculating the total environmental impacts of hydrogen liquefaction in future research.

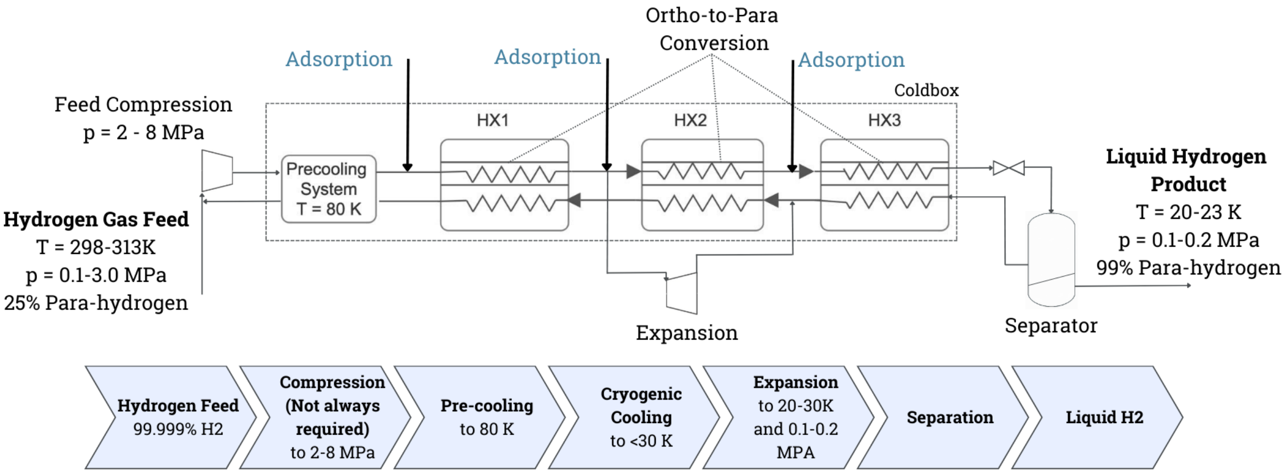

Figure 1.1: Flowsheet of a baseline hydrogen liquefaction cycle. from left to right, hydrogen feed gas is compressed to the required pressure and entered into the cooling cycle. the hydrogen is then cooled by first the pre-cooling cycle and then the cryogenic cooling cycle. Some turbines are included to regain some power and liquid hydrogen is separated from the remaining gas which is re-entered into the cooling system. Based on Al-Ghafri et al. [13].

1.2 Life Cycle Assessment

LCA is climate impact research that is subject to international standards. These standards are defined in the International Organization for Standardization (ISO) 14040 [17], and ISO 14044 [18]. They are systematic methods for defining the environmental impacts of products, processes, and their usage and/or disposal. LCA has four steps, Goal and Scope, Life Cycle Inventory (LCI), Life Cycle Inventory Assessment (LCIA) and Interpretation. The framework of these four steps is shown in Figure 1.2.

Goal and scope

The purpose of the study is established in this phase. It should be tailored to a specific audience and have clearly defined goals. To achieve the goals, system boundaries are determined. During this phase, the functional unit is also defined. The functional unit is the reference point against which the environmental impacts are measured [19].

Life Cycle Inventory

LCI is a list of material and energy inputs, emissions, waste, and products of a product, system or process. This list is produced according to the boundaries set in phase one. This accumulation of data allows for the assessment of climate impacts. In practice, creating an accurate and exhaustive list of relevant material and energy inputs can be quite challenging. Material selection, material origin, energy usage, travel distances, and everything else that affects environmental impacts needs to be reasonably modeled. LCI requires large databases, such as Ecoinvent [20] or the Integrated Design and Engineering Materials Assessment Tool (IDEMAT) [21], which are commercial databases specifically tailored to LCA. These databases contain individual entries of materials and processes. In these entries, energy and material inputs and outputs are documented. An LCI for a given project is constructed by combining and quantifying entries from these databases, ultimately calculating the total material and energy inputs and outputs of the process or product.

Life Cycle Impact Assessment

Environmental impacts are calculated during this phase. An appropriate LCIA method is chosen that fits the goals of the research. This method will interpret the LCI and calculate the environmental costs in defined categories. Environmental cost categories are defined in unique ways in the various LCIA methods that have been developed. No consensus exists among the academic community as to which is the correct approach, as different methods have unique upsides and downsides. As an example, ReCiPe [22], developed by the Rijksinstituut voor Volksgezondheid en Milieu (RIVM), attempts to calculate the harm done to the environment. It defines 17 ”Midpoint impact categories” which can be quantified and linked to three areas of harm, namely, damage to human health, damage to ecosystems, and damage to resource availability. This method is detailed but its hard to compare each of the 17 metrics to each other due to their different units and scales of measurement. this multidimensional complexity requires sophisticated analytical approaches to interpret the results. Another method is Eco-indicator 99 [23]. This method attempts to weigh all harmful effects and attribute all environmental harm to a single score. In this way, comparing results is much easier. However, it can be argued that this approach is short-sighted, since it obscures the type of harm that is caused, potentially leading policymakers to the wrong conclusions. Eco-cost, the LCIA primarily used in this work, has an innovative approach that solves these issues, which is explained in detail in section 2.1. It measures the monetary cost of pollution prevention.

Interpretation

Interpretation should not be seen as a final phase of LCA research. Rather interpretation is a general practice. Results can lead to invalidation of previous assumptions and sensitivity analyses can lead to the re-evaluation of LCI development. This step emphasizes the cyclical nature of LCA. Ultimately, conclusions and recommendations can be made based on the interpretation of results.

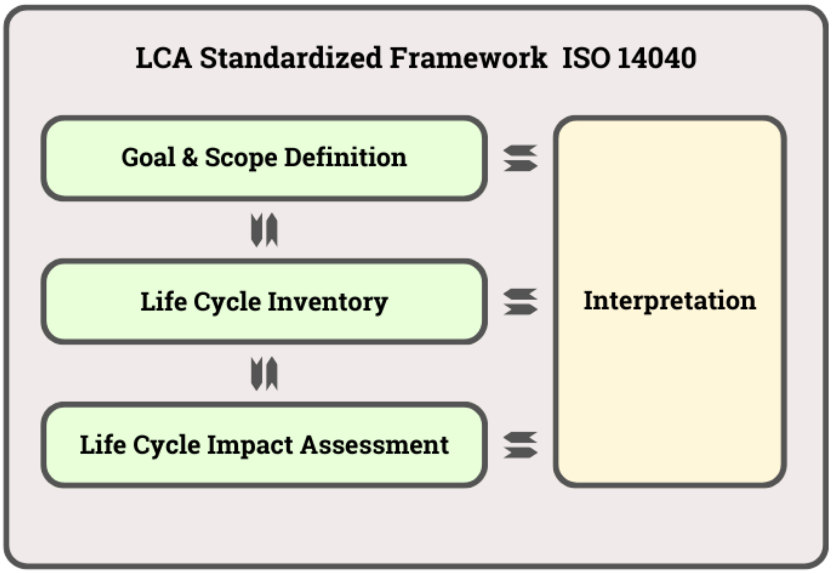

Figure 1.2: General framework of LCA, according to ISO 14040 standards [17] ,[18].

1.3 Hydrogen liquefaction LCA overview

Koroneos et al. [24] conducted an LCA of hydrogen production methods that included various energy sources used for production. Steam Methane Reforming (SMR), biomass, wind, solar and hydropower were analyzed. Koroneos et al. concludes that hydrogen production from photovoltaic (PV) modules has the highest total impact when considering all environmental categories, which it attributed to the manufacturing process of the PV cells, which had low efficiency as of 2004. SMR thus has a lower impact overall when compared to PV cells, but the contribution of greenhouse gas emissions is higher. Hydrogen production from wind or hydropower scores the lowest overall and seem very attractive for sustainable hydrogen. The study used Eco-indicator 95 as methodology. The optimal method of liquefying hydrogen is a two-step process of isothermic compression followed by adiabatic expansion. A total quantity of 1.3 kWh/kg is needed for this process in the form of heat, condensation enthalpy and ortho-to-para conversion. However, when taking into account the Carnot-efficiency, the minimum amount of energy required for liquefaction increases to 4 kWh/kg. This makes 4 kWh/kg the theoretical maximum achievable exergy efficiency. Koroneos et al. further notes how commercial plants are currently unable to reach efficiencies close to this number and states that the database they used in this paper, GEMIS [25], attributes 2.44 kWh/kg to the liquefaction process. This figure is roughly 39% lower than the theoretical optimum, and thus, seemingly false.

Bhandari et al. [26] extensively discussed hydrogen production methods and reviews LCA in this field. A visualization of Global Warming Potential (GWP) was produced, shown in Figure 1.3. It shows the emissions of various hydrogen gas production methods, and their equivalent emissions denoted in kg CO2 equivalent. SMR is still by far the most common production method, as of 2024 [13], yet it is one of the most polluting. Alternatives like wind-based electrolysis are far less polluting. The data shows that grid electricity should not be used for hydrogen production using electrolysis, as this emits far more CO_2 than SMR. This article reiterates the difficulty of comparing LCA studies to each other, due to differences in assumptions, methods and data. So any figure that attempts to compare results from different studies should be taken with healthy scepticism. It should also be noted that GWP is only one of many possible LCA indicators, and thus, gives an incomplete picture of total environmental impacts.

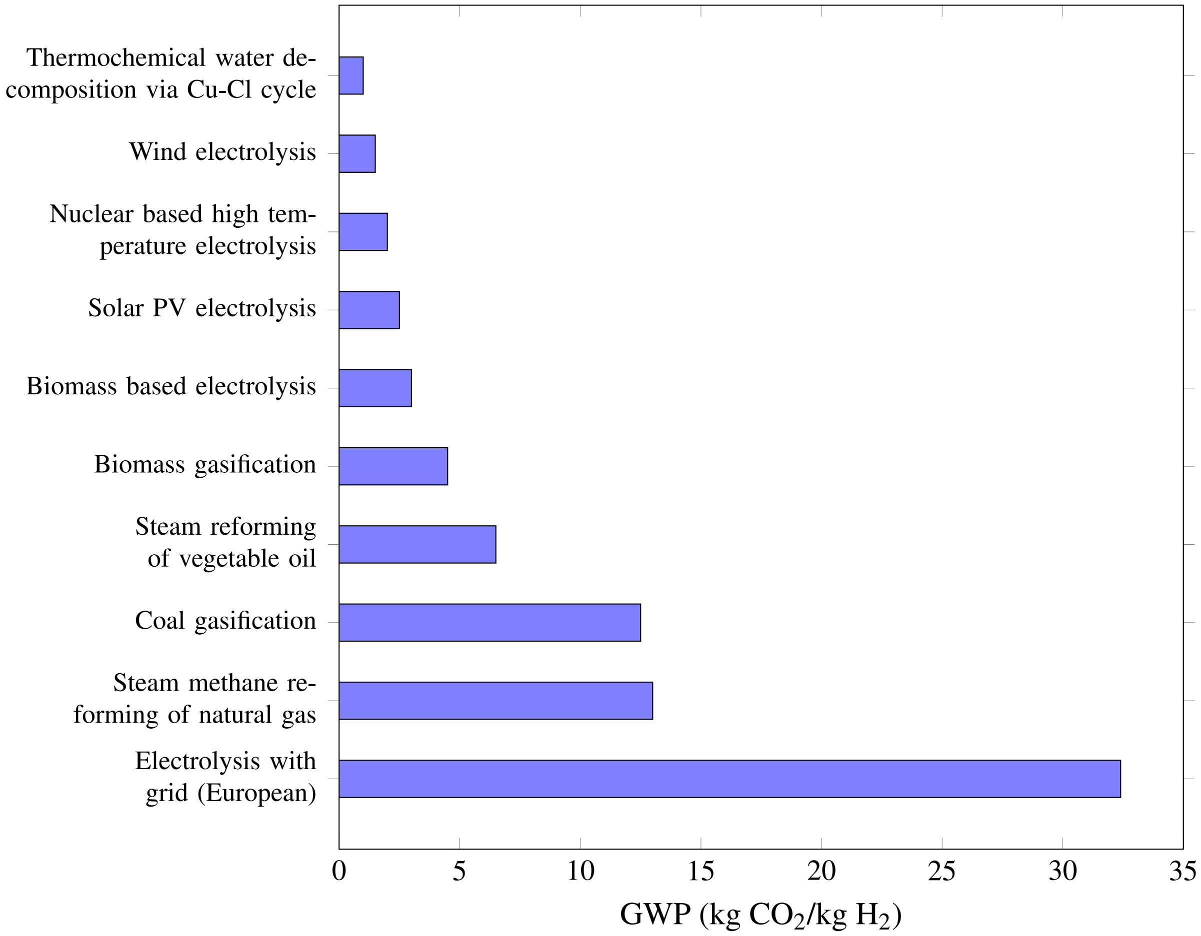

Figure 1.3: GWP of various hydrogen production methods, taken from different studies, reproduced from Bhandari et al. [26].

Al-Breiki et al. [27] does comparative LCA on different sustainable marine fuel alternatives, and compares them to the current baseline of LNG. The study uses the GREET model, which has an in-built calculation for LH_2 production which only uses the electricity costs for LCIA. This paper also gives details on ocean tankers specific parameters, which is a useful figure for transport related calculations.

IDEALHY [28] was a European project in 2013 aimed at developing and optimizing hydrogen liquefaction technologies. The project generated multiple scientific publications, including a notable work by Stolzenburg et al. [29]. This work contains a section on hydrogen liquefaction LCA. The liquefier in Stolzenburg et al. has a production capacity of 50 tons per day (TPD). It is assumed to have a lifespan of 30 years, and an annual operating time of 8000 hours. The document includes several details that are usually an oversight in hydrogen liquefaction LCA. It contains a list with the required materials for liquefier construction (shown in Table 1.1). This list can be used in combination with an LCA database to identify the environmental impacts associated with industrial manufacturing using these materials. The study also estimates the required power for plant operations, which are outside the thermodynamic process of liquefaction, such as a vacuum pump for the cold box, control systems, safety devices and other power consuming machinery that is used to operate an industrial liquefier, which is shown in Table 1.2. This additional information is vital for LCA, as additional power usage will likely affect the environmental impacts of hydrogen liquefaction, and is often overlooked in other literature. The report also mentions the possible LNG regasification cold exergy potential, in the case that hydrogen is produced using SMR, for which LNG can be used as an input. This step vastly reduces the electricity input requirement of precooling, which is shown in Table 1.3. The report reinforces the importance of hydrogen feed gas assumptions. This work uses an input pressure of 20 bar, but notes that 40 bar or even 80 bar input pressure would decrease power requirements substantially, reducing the feed compressor stages or allowing for its complete removal. The latter would reduce the total power consumption by roughly 9%, similar to LNG integration.

Table 1.1: Materials used for constructing a 50 TPD hydrogen liquefier according to Stolzenburg et al. [29].

Material	**Weight [ton]**
Carbon Steel	380
Stainless Steel	595
Copper	150
Aluminum	140
Concrete	46,620

Table 1.2: Total power consumption of hydrogen liquefaction associated with auxiliary plant operations according to Stolzenburg et al. [29].

Plant production capacity	**40 TPD**	**50 TPD**
Liquefaction process total (kW)	10,666	13,332
Liquefaction plant total (kW)	11,276	14,057
Percentage increase	5.72%	5.44%

Table 1.3: Power consumption comparison with and without LNG integration.

LNG Integration	**Total Power Consumed (kW)**
No	14,057
Yes	12,775
Percentage Decrease	9.12%

Ansarinasab et al. [30] studied the development of an improved hydrogen liquefaction cycle, with LCA included. It uses Eco-Indicator 99 for LCIA. It assumed equipment lifetime lasts twenty-five years at 7300 hours per annum. For LCI, it provides a material list which is reproduced in this work. Unconventionally, they include three air coolers in their liquefaction cycle, during the cryogenic stage. The paper suggests cooling hydrogen gas from a temperature of 80 Kelvin to 23 Kelvin using ambient air as the cooling medium. Furthermore, they use specific Eco-indicator 99 values sourced from Cavalcanti et al. [31], which are reproduced in Table 4. The table shows the material makeup of some relevant liquefaction equipment types, as well as their respective Eco-indicator point values. These points represent various environmental impacts, which are weighted and combined to become a single output metric, ”points”. These points are meant to be combined with the weights of the individual components to come to a final result for environmental impacts as they are given in points per kilogram. We were unable to find the weights used in this work.

Table 1.4: Liquefier component material composition and weighted Eco-Indicator 99 approach, Cavalcanti et al. [31]. The points represent a combination of all environmental harm.

Component	Material	Mat. %	EI99 (mPts/kg)	Points (mPts/kg)
Compressor	Steel	33%	86	131
	Steel low alloy	45%	110	
	Cast iron	22%	240	
Air Cooler	Steel	66%	86	519
	Copper	33%	1400	
Pump	Steel	35%	86	186
	Cast iron	65%	240	
Turbo-Expander	Steel	25%	86	201
	Steel high alloy	75%	240	
Heat Exchanger	Steel	66%	86	519
	Copper	33%	1400	

Frank et al. [32] is an LCA comparing GH2 and LH_2 pathways. It uses GREET [33] and HDSAM [34] as tools for hydrogen liquefaction LCIA. HDSAM uses a 10 TPD liquefier with a 11 kWh/kg SEC. According to HDSAM, hydrogen liquefaction should be attributed an additional 0.5% energy loss factor based on industry experience. Akhtar et al. [15], [35] analyses various hydrogen carrier pathways with truck transport. It utilizes the Ecoinvent 3.6 database through the SimaPro software and chooses the Centrum voor Milieukunde Leiden (CML) method for LCIA. This study used the Stolzenburg et al. [29] values for liquefier construction materials, shown in Table 1.1. They also provide A liquefaction cycle which was calculated at an SEC value of 6.76 kWh/kg LH_2.
Yilmaz et al. [36] presents a study on the geothermal-to-LH_2 pathway. A liquefaction SEC of 8.6 kWh/kg was calculated. This work doesn't address the environmental impacts of this geothermal-to-LH_2 pathway. Kolb et al. [37] is an LCA on hydrogen imports to Germany. It uses German-made compressed gaseous hydrogen (CGH_2) and compares it to LH_2 imports from high potential countries, namely Quebec for its wind power, Morocco for its solar power, and Chile for both its wind and solar power. For the liquefaction of hydrogen, the paper uses an SEC of 13.8 kWh/kg, and uses this figure to represent all emissions of this stage. The method used for impact assessment was CML-IA [38]. The database was Ecoinvent [20] and the software used was openLCA [39]. Furthermore, this paper addresses a glaring problem with an assumption in most hydrogen LCAs. Namely that liquefaction is a continuous process, and the assumption that a standalone system can use 100% green electricity cannot be fulfilled. Wind and solar are themselves not continuous power generation technologies, so either overproduction is needed where the base power generation is used and excess energy has to be produced and sold or utilized, or grid energy needs to be purchased to fill the energy gaps.
Kim et al. [16] produces an LCA on the impacts of using mixed refrigerants (MRs) for hydrogen liquefaction. The MRs contained methane, ethane, helium, hydrogen, nitrogen and propane. It used the ReCiPe Hierarchist [22] method for LCIA, using the Ecoinvent v3.0 database [20] in the SimaPro software [40]. The results are an emission of 67.85 kg CO_2 eq. on day 1 due to the acquisition of the refrigerants, and then a daily emission of 0.253 kg CO2 eq. due to assumed refrigerant leakage. This approach seems questionable, as counting refrigerants acquired on day 1 as "emissions" seems counterintuitive, rather, this could be categorized as raw resource costs. This study does not compare the MR to the base case in terms of environmental impact assessment. They also provide the SEC and an LCI of the raw construction materials used for the liquefier, which is reproduced in Table 5. The raw construction materials represent the raw material costs per ton of

LH_2 produced, instead of the total materials used for construction, which is a different approach to other works that produce raw construction materials LCI. They highlight a lack of detailed life cycle assessment of hydrogen liquefaction processes is available, and thus, are unable to compare their results with any other process. The study also states that refrigerants are widely known for having significant impacts on the environment, and are thus, included. Results show a relatively insignificant amount of kg CO_2 eq. impacts because of construction materials, however the provided material list seems somewhat unsubstantiated, including only three material types, of which all three have the exact same amount used.

Table 1.5: Alternative material weights.

Material	**Weight [ton]**
Aluminum	13.89
Nickel	13.89
Titanium	13.89

Noh et al. [41] conducted an LCA on various cradle-to-gate hydrogen pathways linked to wind on sea energy. For all scenarios, hydrogen is produced offshore on a platform. It then compares the environmental performance of various forms of hydrogen in the context of ship transport. The forms of hydrogen that are compared are CGH_2, LH_2, liquid organic hydrogen carriers (LOHC), and ammonia (NH_3). It includes the environmental impacts of these technologies at transport distances ranging from 100 km to 10,000 km. The study employed the CML-IA baseline V4.7 method [38] using the SimaPro 9.1.1.1 software [40]. The study evaluated the liquefaction step as consuming 15.0 kWh/kg. It concludes that LH_2 becomes the most environmentally efficient pathway at distances above roughly 1000 km in terms of GWP. It scores similarily to NH_3 in this metric but the study notes that BOG rates may decrease through technological innovations and this will increase the efficiency further . The study highlights the high amount of energy lost in the entire hydrogen value chain. Starting at 41-57% for 100 km distance travelled, increasing along with travel distance. A question remains about the assumptions in this paper. The notion that offshore hydrogen liquefaction has the same environmental impacts as land based liquefaction is not substantiated.
Incer-Valverde et al. [42] presented a liquefaction cycle using helium as refrigerant. Its SEC is calculated at 8.06 kWh/kg. The environmental impact rates for each component type are calculated using the associated exergy destruction, compressors being the most inefficient. This data could help future research in the environmental and economic cost optimization for the liquefaction cycle. It neglects the construction, maintenance, and leakage environment costs. The work uses the Eco-Indicator 99 [23] for LCIA.
Choe et al. [43] performed an LCA on green hydrogen production, with direct integration of a liquefaction unit. It analysed the validity of this pathway with respect to energy and cost intensiveness, as well as an environmental analysis. Their liquefaction cycle is calculated at an SEC of 5.418 kWh/kg, which is a low value when compared to others, indicating an efficiently designed cycle. One factor in this result is the optimized MR. The SEC value is used in all of the nine evaluated scenarios, and is calculated in Aspen Hysys [44]. This is a case where the hydrogen liquefier itself is actually included in the analysis. In the supplementary materials, they provide a table for the materials used for the construction of a 10 TPD hydrogen liquefier. This data is reproduced in Table 6. The five material types for this liquefier are congruent with the material types of the liquefier materials given in Table 1, however, this work estimates a total amount of materials used for a 40 TPD liquefier that is roughly 3 times lower than the 50 TPD liquefier of Stolzenburg et al. [29], which is a clear discrepancy. They cite a source from IRENA [45] for these values, however this source does not appear to contain the produced data. The method for impact analysis used was ReCiPe2016 [22]. However, from the produced figures and conclusions, it is unclear what the impact of these construction materials has on the overall liquefaction environmental costs.

Table 1.6: Materials used for constructing a 10 TPD hydrogen liquefier according to Choe et al. [43].

Material	**Weight [ton]**
Carbon Steel	123
Stainless Steel	193
Copper	49
Aluminum	50
Concrete	15,111

Additionally, a review study on the methodological choices of LCA literature was done. Valente et al. [46] provides unique insight on a meta level. Most of the studies included cradle-to-gate boundaries, with cradle-to-grave boundaries being the preferred choice in the case of mobility-oriented works. Common global warming impact and energy consumption methodologies used were the Intergovernmental Panel on Climate Change (IPCC) method [47], and the Verein Deutscher Ingenieure (VDI) [48]. Other categories were most often evaluated using CML-IA [38]. In general, however, all kinds of methods are used across the board, like Eco-indicator 99 [23], ReCiPe [22], and Greet [33], which were also relatively prominent. The study also states that number of works published on hydrogen LCA has been steadily increasing. Most reviewed works used scientific literature or LCA databases as information sources, and very few studies used real data. On a sidenote, there is an official guideline document on LCA for hydrogen production called HyGuide [49], which is often cited in the literature. It states nothing about how to treat the environmental impacts of hydrogen liquefaction [50], [51].

1.4 Knowledge gaps in liquid hydrogen research

In this literature review, we find only a total of three works with a given list of materials for plant construction, Choe et al [43], Stolzenburg et al. [29], and Kim et al. [16]. Only Kim et al. contains environmental impacts assessment of refrigerants, albeit in a seemingly faulty manner, as no leakage analysis is applied. The reviewed works do not consider the same materials, do not use the same units, and thus comparing them does not seem very sensible. Apart from these particular studies, the way that hydrogen liquefaction is treated in most full-chain cradle-to-grave LCAs, can be boiled down to pulling out a single paper, taking out the SEC of the liquefaction cycle (shown in Figure 1.4), and calculating the environmental impacts purely based on the operating energy costs from this number. For this, there is no clear justification. Despite some limited efforts made on MRs and liquefier construction, it is still not clear whether the entire life cycle of the hydrogen liquefier itself will significantly impact the used figures. **This thesis aims to address this, and provide useful and original data on the validity of previous research, and guidelines/recommendation for future work.** It is assumed in most of the literature that the environmental impacts of hydrogen liquefaction can be calculated using only the operating energy. **This thesis will test that assumption, and provide recommendations and guidelines for if and when this assumption fails to hold up.** An interesting question is whether an analysis of liquefier construction materials can be neglected. The literature provides bits of evidence that the environmental impacts of plant construction materials is negligible compared to electricity and refrigerant impacts. As for methodology, for LCIA, the Eco-indicator, CML and ReCiPe methods were most prevalent in their various iterations over time. The Ecoinvent database was the most popular, with some exceptions, namely the use of original data or other lesser-known sources.

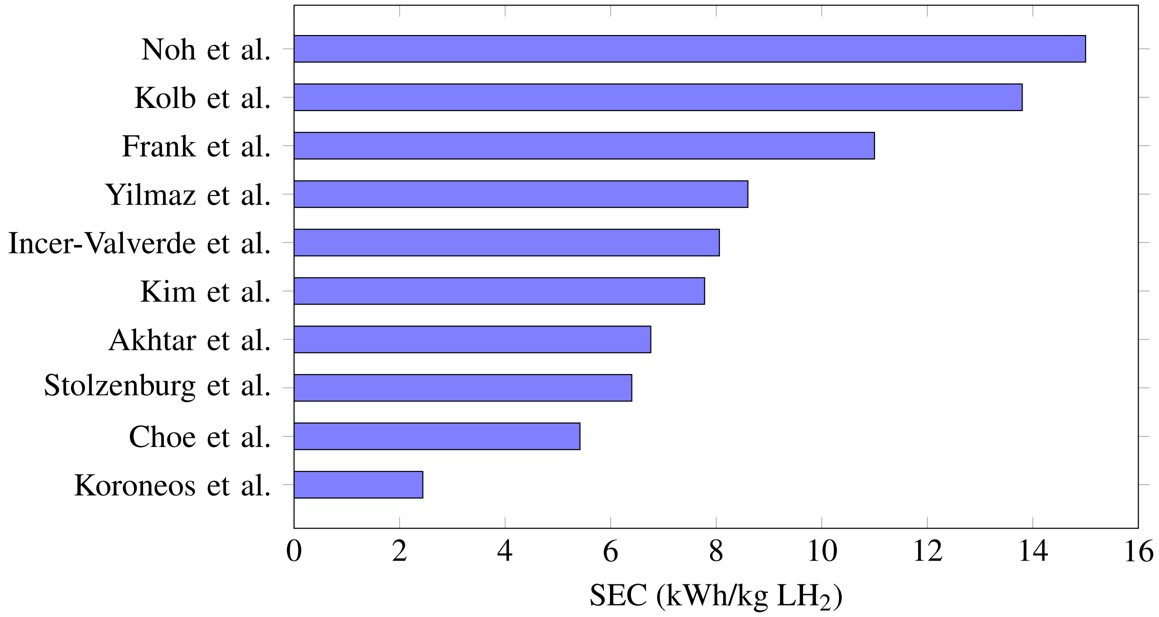

Figure 1.4: SEC used for hydrogen liquefaction LCA by reviewed works, ranked by value. From top to bottom: Noh et al. [52], Kolb et al. [37], Frank et al. [32], Yilmaz et al. [36], Incer-Valverde et al. [42], Kim et al. [53], Akhtar et al. [15], Stolzenburg et al. [29], Choe et al. [43], Koroneos et al. [24].

1.5 Goal and scope of the thesis

According to the literature, the environmental impacts of hydrogen liquefaction have not yet been properly determined. Hydrogen liquefaction is a thermodynamic process that occurs in a hydrogen liquefier. A hydrogen liquefier is a large, complex industrial plant, and estimating the environmental costs of such a facility requires specific, in-depth technical knowledge. Potential relevant factors include total power consumption, refrigerant leakage, and raw construction materials. In previous research, these factors are rarely included. It is vital to the understanding of hydrogen as a potential clean fuel that all of these metrics are assessed, so that researchers and policymakers can accurately judge its environmental viability. Various types of hydrogen liquefiers exist, both operational and theoretical. These facilities must be analyzed, as well as various aspects such as sizing, and including the potential for LNG regasification integration. The question posed in this thesis is as follows: **What are the environmental impacts of various types of hydrogen liquefiers?**

Goals

The primary goals of this work are understanding the environmental impacts of various hydrogen liquefier types, focusing on aspects such as impact sources, plant sizes, types, and LNG regasification integration. A secondary goal is to provide clear guidelines and models for implementing hydrogen liquefaction in future LCA research.

Boundaries of LCA

This thesis will first make a gate-to-gate assessment of the conversion of hydrogen gas to LH_2 through a commercial hydrogen liquefaction plant. Gate-to-gate refers to the boundaries of this LCA. It means that we will first analyze only the liquefaction, rather than taking the whole hydrogen life cycle into account, this is shown in Figure 1.5. First, A base case hydrogen liquefaction plant is established. This baseline model is then used to compare the environmental impacts of different plant sizes, refrigerant mixtures, and liquefaction cycle efficiencies. These comparison scenarios are based on liquefier designs found in the literature. The goal of these results will be to aid LCA researchers in applying hydrogen liquefaction into their research. Next, a cradle-to-grave analysis is done. Cradle-to-grave hydrogen LCA will provide realistic scenarios that utilize LH_2 where it would be preferable logistically. It will show the environmental impacts of different pathways of hydrogen utilization. Cradle-to-grave LCA defines system boundaries from the moment when the hydrogen molecule is created, until it turns into water following its extraction of useful energy. These scenarios are described in section 3.1. For decision makers, cradle-to-grave research can aid in the selection of sustainability projects and policies.

The primary reason that two approaches are combined in this thesis is that high-quality data on liquefier impact is lacking, or not publicly available. Figure 1.5 shows a generalized pathway of the life cycle of hydrogen, in which hydrogen liquefaction is utilized. It includes ship transport, as liquefying hydrogen would only make logistical sense if long distance transport to an end-user is required. For shorter distances or localized distribution, less energy-intensive hydrogen forms would be more cost-effective and environmentally efficient.

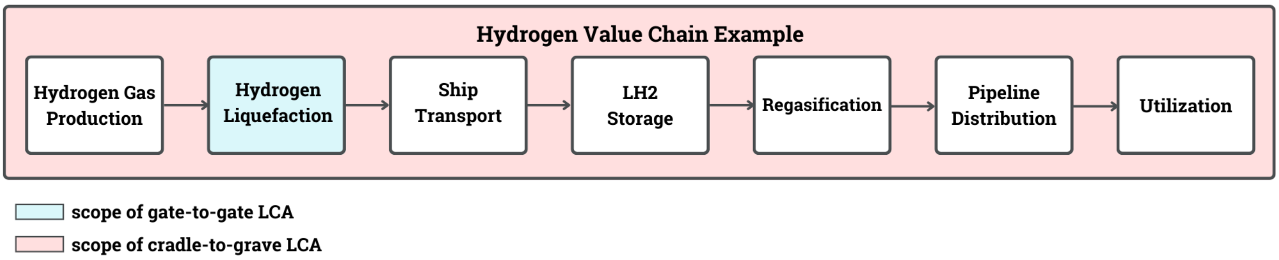

Figure 1.5: Boundaries of LCA performed in this Thesis

Functional unit definition

This thesis will include both a *gate-to-gate* LCA on hydrogen liquefaction, as well as a *cradle-to-grave* LCA. A **functional unit** is selected for both phases. This functional unit will serve as the reference point from which the environmental impacts are analyzed and compared. The functional unit will be the production of **one kilogram of liquefied hydrogen (1 kg LH_2)**. This normalization will allow the comparison of liquefiers with varying production capacities.

2. Methodology

This chapter outlines the methodology for calculating the environmental impacts of different hydrogen liquefier configurations. First, environmental impact source categories (inputs) and resulting ecocost categories (outputs) are defined. Then, various scenarios are produced to showcase the effect of different variables of the hydrogen liquefier on its environmental impacts. Afterwards, details about modelling approaches and assumptions are presented and argumented. The limitations imposed by these choices and assumptions are discussed toward the end of the chapter. This chapter means to provide the necessary details to ensure the reproducibility of this work.

2.1 LCIA method

Ecocost [54] is an LCIA method that was developed by Delft University of Technology (TUDelft) [55]. It takes a different approach than previous LCIA methods and seeks to adress inherent problems. Ecocost is the chosen LCIA method of this thesis. To understand the innovations of the Ecocost method, we compare it to other established methods in the field. ReCiPe [22] is a well-rounded LCIA method developed by Leiden University. It addresses most aspects of environmental harm. ReCiPe attributes damage values in seventeen categories, which can then be interpreted on two levels of harmful effects. Namely, damage pathways and endpoint areas of protection. The inherent problem of this approach is that comparing damage values is inherently subjective and joining them together to a single score requires arbitrary weighting. Furthermore, it is currently extremely difficult to get accurate descriptions of environmental damage in this way, as it is simply too hard to define the damage caused by each substance. This means the inaccuracy of the results is very high. Ecocost takes a different approach. Instead of measuring the damage caused by pollution, it attributes monetary costs associated with pollution prevention. This allows for objective category comparison, integration of LCA into policy considerations such as tradeable emission rights and provides an innovation-based perspective for sustainability goals.
Both Idemat [21] and Ecoinvent [20] databases are used in this thesis. Ecoinvent is a large database but lacks data on certain advanced materials that TUDelft LCA researchers require in their research, as such they have constructed their own database called Idemat. In this Thesis, we will use a combination of the two databases. This is problematic, because different databases have different methodologies, creating systematically wrong comparative data. Despite these limitations, mixing databases in this manner was judged to be acceptable out of necessity. SimaPro [40] is software that integrates these databases and methods and is used in this thesis.

2.2 Environmental impact sources and result categories

A hydrogen liquefier has three significant sources of environmental impacts:

- **Electricity**: The energy required to run the liquefaction plant, contributing to impacts based on the source of electricity.

- **Raw construction materials**: Materials like steel and concrete used in building the liquefier, contributing to impacts through their production and resource extraction.
- **Refrigerants**: Cooling agents needed for hydrogen liquefaction, which contribute impacts through their production and emission into the atmosphere when they leak.

Depending on the size and type of the liquefier, the relative contribution can vary. Options like power sources make a large impact. Or LNG regasification integration, which requires additional logistics but decreases the required electricity. Figure 2.1 shows how these categories fit into the LCA framework. It highlights that these are the sources of environmental impacts that flow into impact result categories. In this way, hotspots of environmental impacts can be identified when the results are calculated. Four resulting impact categories are described by the EcoCost method:

- **Climate change**: Harm caused by the release of greenhouse gases that contribute to global warming.
- **Human toxicity**: Adverse health effects on humans due to exposure to harmful substances, such as heavy metals released during material production, which can cause respiratory and other health issues.
- **Ecotoxicity**: Damage to ecosystems due to the toxic effects of pollutants, such as chemical leaks that can contaminate water sources and affect aquatic life.
- **Resource scarcity**: Environmental costs resulting from the depletion of finite natural resources, such as the use of uranium in nuclear power production.

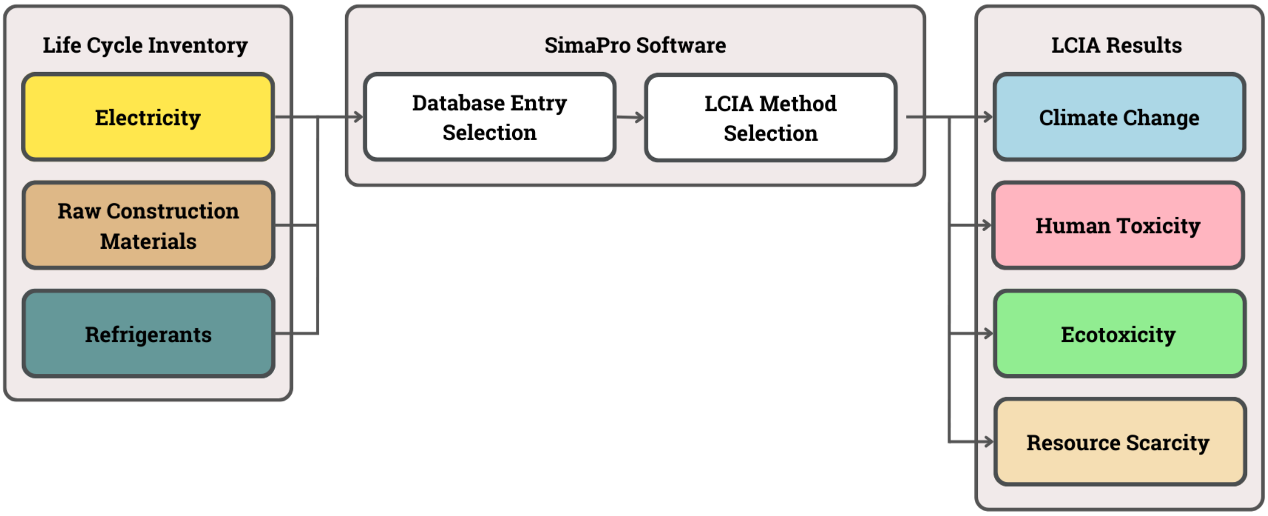

Figure 2.1: Visualisation of how environmental impact sources flow into impact result categories.

2.3 Refrigerants and refrigerant mixtures

Refrigerants and refrigerant mixtures are cooling fluids with optimized properties. During the liquefaction process, these refrigerants are compressed, expanded, and otherwise manipulated with large machinery which produces the cooling effect. The refrigerant mixture in this complicated process is trapped in a closed loop. A closed loop refers to the fact that the cooling system is filled up with a “charge” of refrigerant, after which it only needs refills due to leakage. Many Refrigerant compounds are widely known to be harmful to the environment. Therefore, it is vital that we analyze the impacts of various refrigerant mixtures that can potentially be applied to hydrogen liquefaction. The environmental impacts of these mixtures can be calculated if we can estimate the total refrigerant losses during the lifetime of the liquefier. These losses are the initial filling up of the system, the leakage rate during its lifetime, and the losses during the recycling.

2.3.1 Estimation of refrigerant emissions through leakage and end-of-life losses

Refrigerant leakage rates in commercial liquefaction plants are a very specific type of data, which is not widely available. A report from Efficiency Maine was published on refrigerant leakage assessment in 2022 [56]. The report states that it is very difficult and expensive to study refrigerant leakage, which explains why there is very little primary research on refrigerant leakage rates. In this thesis, an attempt was made to come to reasonable estimates for industrial refrigerant leakage rates based on public sources such as papers, reports, and industry regulations. The focus of these public sources is often commercial refrigeration, which is the continuous cooling of perishable goods such as food. Leakage rate data of commercial refrigeration is often mixed with industrial refrigeration, despite their large differences [57]. According to the Efficiency Maine report, refrigerant leakage rates for commercial & industrial applications range from 2.42% to 6.75% of total charge annually [56].

Another source for leakage rates is the regulation for refrigerant leakage in Industrial Process Refrigeration (IPR) set by the Environmental Protection Agency (EPA), under section 608 of the Clean Air Act mandate [58]. According to these guidelines, IPR systems using at least 50 pounds of refrigerants should have annual leakage rates of no more than 35% of the total amount of refrigerant in the system under normal operating conditions. Furthermore, it states that refrigerants are not allowed to be vented during the disposal of appliances.

An additional source on refrigerant leakage rates was produced by Accuvio [59]. They collected data on average refrigerant leakage rates across different industries in 2017. Accuvio is a sustainability and environmental, social, and governance (ESG) reporting software. Their software is designed to help organizations manage their sustainability data. Their report cites leakage rates from two separate governmental organizations. The first is a UK agency, namely the Department for Environment, Food and Rural Affairs (DEFRA) [60]. According to their reporting, domestic industrial refrigeration leakage rates are on average 8.00%. For individual components, heat pumps are attributed 6.00% and condensers are attributed 10.00%. The other cited party was the Intergovernmental Panel on Climate Change (IPCC) [47]. They attribute a rate of 7.00%–25.00% for ”Industrial Refrigeration including Food Processing and Cold Storage” but mention this should be used as a preliminary estimate [59]. As a final public source, there is a news article that states that German refrigeration leakage rates have declined from 2.36% in 2019 to 1.12% in 2022 [61]. The original source of this data was not found.

Besides the yearly refrigerant leakage rates, there is also the question of startup and end-of-life losses. While filling up an industrial system with refrigerant, a large initial loss of refrigerant may occur. Additionally, recycling at the end-of-life of the system may incur a loss as well. Leakage data regarding the initial setup and decommissioning of refrigeration systems are discussed in a guideline document produced by the IPCC. According to the 2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories, end-of-life refrigerant recovery efficiencies are between 0% and 90%, and initial charge emissions are 0.5%–3.0% [62]. Since these figures are concerned with the entirety of refrigeration, some amendments need to be made to these figures to apply them to hydrogen liquefiers. It is unlikely that end-of-life refrigerant recovery in industrial applications would be as low as 0%, because regulations discussed earlier in this chapter forbid the venting of refrigerants during the disposal of appliances. This means that we can safely assume that any operated liquefier complying with regulations should realistically be in the upper end of this distribution. For the initial loss, we assume the higher end of the spectrum, as this process may

involve a long time of exposed refrigerant as the system fills up.
To conclude, an estimation of refrigerant leakage rates for hydrogen liquefiers is given based on a variety of sources, some of which are a little shaky. This was done out of necessity due to the lack of high-quality data. Estimations of leakage rates vary widely when commercial and industrial refrigeration are combined. Therefore, we adopt three levels of leakage based on the quality and frequency of leakage prevention. The figures are given as a percentage of total charge that is leaked annually:

- 2% – High level of leakage prevention, consistent checks and maintenance
- 8% – Based roughly on the average leakage rates in UK
- **30% – Compliant but careless industry, reaching the legal boundary**

The 30% case is highlighted because this figure was used for all scenarios, as even this highest figure still contributes insignificant environmental impacts, as can be seen in Figure 3.2

The estimates for the initial setup losses and decommission losses used in this thesis are as follows:

- Startup loss $\sim$ 2.5% of total charge
- End-of-life loss $\sim$ 7.5% of total charge

Total volume and leakage estimation

The total leakage over the lifetime of the equipment is calculated as follows:

- Step 1: Calculate the total volume of all equipment:

  $$V_{\text{total}} = \sum_{i=1}^{n} V_{\text{equipment, } i} \tag{2.1}$$

  where $V_{\text{equipment, } i}$ is the volume of each individual piece of equipment, and n is the total number of equipment components.

- Step 2: Multiply the total volume by the leakage rate to calculate the leakage volume:

  $$V_{\text{leakage}} = V_{\text{total}} \times r_{\text{leakage}} \tag{2.2}$$

  where V_{leakage} is the total volume of refrigerant leaked per year, and r_{leakage} is the yearly leakage rate.

- Step 3: Calculate the total annualized leakage volume over the lifetime of the equipment:

  $$V_{\text{total, annualized}} = \frac{V_{\text{yearly}} \times t_{\text{lifetime}} + V_{\text{setup}} + V_{\text{EOL}}}{t_{\text{lifetime}}} \tag{2.3}$$

  where V_{yearly} is the yearly leakage volume, t_{lifetime} is the total lifetime of the liquefier in years, V_{setup} is the volume of refrigerant leaked during setup, and V_{EOL} is the volume leaked at the end of life.

- Step 4: Multiply the annualized total volume by the average density to calculate the total leaked mass:

  $$m_{\text{total}} = V_{\text{total, annualized}} \times \rho_{\text{avg}} \tag{2.4}$$

  where $V_{\text{total, annualized}}$ is the annualized total volume of refrigerant leaked, and ρ_{avg} is the average density of the refrigerant mixture, which is estimated in Figure 2.3.1.

Refrigerant mixtures for hydrogen liquefaction

Most hydrogen liquefier designs feature two separate phases of cooling, precooling and cryogenic cooling. The simplified reason for requiring two separate refrigerants is that a single optimized refrigerant mixture would freeze at extremely low temperatures. The solution is to make an optimized refrigerant for the higher temperatures, and a freeze resistant refrigerant mixture for the lower temperatures. Figure 2.2 reiterates this distinction between two stages.

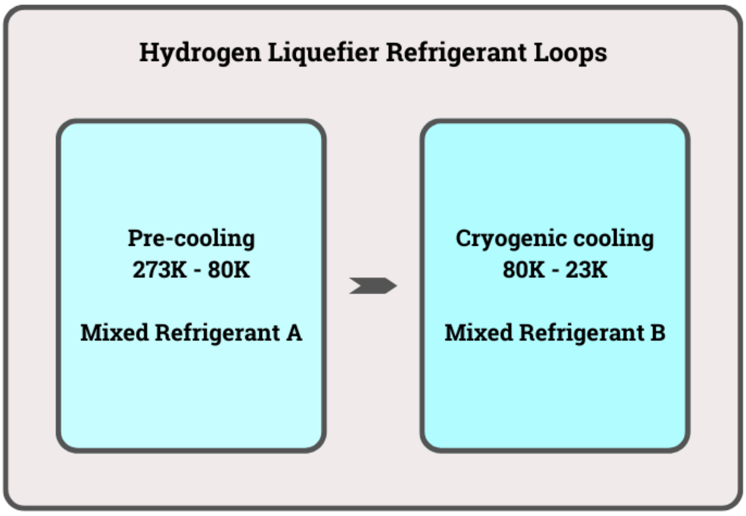

Figure 2.2: Visualization of the two closed loop refrigerant systems present in most modern hydrogen liquefier designs.

The precooling stage is subject to a lot of different possibilities for optimization, and many novel cycles with unique refrigerant mixtures have been developed in the literature. The precooling refrigerant mixtures that were analysed in this thesis are given in table Table 3.4. The cryogenic phase is less subject to discussion and is dominated by the limitations set by the extremely low temperatures. For this stage, hydrogen mixed with helium, neon or standalone can be used as a refrigerant because these substances are the only ones that do not freeze at 23 Kelvin. The cryogenic cooling refrigerant mixtures that were analysed in this thesis are given in table Table 3.5.

Total refrigerant charge estimation

To environmental impacts of refrigerant leakage in hydrogen liquefaction can be calculated if we can estimate how much refrigerant is in the system at normal operating conditions. The total amount of refrigerant in the system is referred to as the total charge. The total charge estimation was done for the four sizing scenarios using Aspen Economic Analyser software data as an input. The required parameters for all the equipment pieces used in the liquefaction cycle are provided by the program. The volumes for many equipment types could be directly calculated. Namely, for heat exchangers, flash collumns and cryoboxes. These volumes were used directly as liquid volume estimation. For adsorbers, the liquid volume was given directly in the data. For the Turbines, Table 4.8 of Tamarona et al. [63] was used. For compressors, it was assumed that the liquid volume was 50% of the total volume. A density of stainless steel of 8000 kg/m^3 was used for this calculation.
The data also excluded the valves and piping, so 50% was added to the total volume to account for this as a preliminary estimation. This 50% addition is assumed to be an overestimation but it is chosen arbitrarily, since no applicable data was found to properly determine the amount of piping present in hydrogen liquefiers. Next, the average density of the refrigerant mixture was calculated, after which the leakage dynamics were applied assuming total liquefier lifetime of 30 years. The preliminary piping and valves estimation of 50% addition in total volume was unchanged in the final

results. The reason is that the total environmental impacts of refrigerant leakage turned out to be insignificant as can be seen in Figure 3.2, so it was deemed unnecessary to improve the accuracy further and implement detailed estimation approaches.

Average density of MR estimation

Determining the total weight of refrigerant inside a liquefier is challenging due to the varying states of the refrigerant at different stages of the liquefaction cycle. To address this, the average density of various refrigerant mixtures was estimated as follows. First, the composition of the refrigerant mixtures, along with the temperatures and pressures provided in the stream data from Tamarona et al. [63] were entered into the Refprop software tool by NIST [64] to generate a table of local densities for each stream. However, in some streams, the refrigerant mixture is partly liquid, resulting in extremely high local densities, which could lead to an overestimation of the average due to the presence of these outliers. To mitigate this, a second step was taken. An average temperature and pressure were calculated separately for both the precooling phase and the cryogenic cooling phase by averaging the temperatures and pressures from the stream data. For the precooling phase, this resulted in 244.8 K and 22.71 bar, and for the cryogenic cooling phase, 95.31 K and 10.82 bar. These average values were then used for all liquefaction cycles analysed in this work. The densities for each refrigerant mixture were then calculated using these averaged temperatures and pressures. Finally, The two resulting average densities were averaged again to obtain the final average density, which was used in subsequent calculations. As a note, some studies directly included the compositions of their refrigerant mixtures, while others included other data that was sufficient to calculate it. Other studies, unfortunately did not have enough information in them to be included, notably the IDEALHY project, which denoted only the type of components in their mixed refrigerant, but not the composition.

- Step 1: For each phase (precooling or cryogenic), calculate the average density by averaging the densities of individual streams:

$$\rho_{\text{avg, streams}} = \frac{\sum \rho_{\text{stream}}}{n_{\text{streams}}} \tag{2.5}$$

  where ρ_{stream} is the local density for each stream.

- Step 2: Average the temperature and pressure across all streams for the phase, then calculate the density at these averaged conditions:

$$T_{\text{avg}} = \frac{\sum T_{\text{streams}}}{n_{\text{streams}}}, \quad P_{\text{avg}} = \frac{\sum P_{\text{streams}}}{n_{\text{streams}}} \tag{2.6}$$

$$\rho_{\text{avg, conditions}} = \rho(T_{\text{avg}}, P_{\text{avg}}) \tag{2.7}$$

- Step 3: Take the average of the two densities obtained from Step 1 and Step 2 to get the final average density for the phase:

$$\rho_{\text{final}} = \frac{\rho_{\text{avg, streams}} + \rho_{\text{avg, conditions}}}{2} \tag{2.8}$$

We repeat the same process for both the precooling and cryogenic phases separately.

Database choice

The Environmental impacts of refrigerant components were modelled in two stages. The production of the compound, and its emission to the atmosphere. For most of the refrigerant components that make up the hydrogen liquefaction MRs, this data was available in both the Ecoinvent and Idemat databases. For some components, however, only one source was available. The database choice preference was Idemat where possible. The final database selection for each compound is shown in Table 2.1. Note that the choice of database is only relevant for the production impact, as the data for emission to the atmosphere does not require a database entry. Additionally, note that hydrogen was chosen to be produced using SMR, since this is as of today still by far the most common method of production, and green hydrogen isn't yet included in these databases as the technology isn't widely implemented commercially as of 2024. Additionally, the production of R14 was not available from either database, so instead the closest component R23 was chosen. The impact of this choice is extremely low, since almost all of the negative impact from R14 comes from its emission, which was included correctly.

Table 2.1: Components and corresponding database entries.

Component	Database	Entry
R14	EI	Trifluoromethane GLO— market for trifluoromethane — Cut-off, U
Methane	IDEMAT	A.070.09.101.230701 CNG
Propene	IDEMAT	A.030.14.123.230803 Propylene
Helium	EI	Helium GLO— market for helium — Cut-off, U
Ethylene	IDEMAT	A.030.14.115.230803 Ethylene
Hydrogen	IDEMAT	A.030.08.108.230701 Hydrogen, methane to hydrogen (SMR)
n-Pentane	IDEMAT	Pentane corrected from plastics europe copy
i-Pentane	IDEMAT	Pentane corrected from plastics europe copy
i-Butane	EI	Isobutane GLO— market for isobutane — Cut-off, U
n-Butane	IDEMAT	A.030.14.108.230701 Butane
Ethane	EI	Ethane GLO— market for ethane — Cut-off, U
Propane	IDEMAT	A.030.14.122.230701 Propane
Argon	IDEMAT	A.030.08.101.230701 Argon
Nitrogen	EI	Nitrogen, liquid RoW— market for nitrogen, liquid — Cut-off, U

2.4 Raw construction materials

Calculating the total mass of all raw construction materials for hydrogen liquefiers was done by adding up all material weights:

$$m_{\text{aluminum, total}} = \sum_{i=1}^{n} m_{\text{aluminum}, i} \tag{2.9}$$

where $m_{\text{aluminum}, i}$ is the mass of each individual construction material, and n is the total number of materials used in the equipment. This same scheme was followed for each raw construction material. The materials required for the construction of a hydrogen liquefier were estimated for the three sizing scenarios including the base case adapted from Tamarona et al [63] and the 86 TPD liquefier of Kerkar et al. [65]. The Aspen Economic Analyser files were provided by the authors of these works. These files were systematically scanned for their data on equipment weight, material and concrete requirements. Some additional multiplication factors were implemented as to not underestimate the required materials. Namely, for the concrete, a factor of 1.2 for wasted concrete and 1.4 as an extra margin were used. To account for the piping, the equipment component weights were added up, and 5% of this total weight was added in the form of piping. These estimations are educated guesses. It can be assumed that specialized, high-tech equipment used in hydrogen

liquefaction is made of optimized steel alloys. Interestingly, steel alloy choice does not affect the environmental impacts significantly.
From an environmental impact perspective, the production of steel is categorized in three essential types of steel products. Namely hot roll coils, galvanized steel and steel sections. Where steel sections should be understood to be things like steel beams or pipes. These three groups of steel vary in impacts from each other, but not so much internally, and thus the specialized alloys used in liquefier equipment can be put into the hot roll coils group. The construction materials LCI for the base case scenario as well as the database entry selection for each material type is given in Table 2.1. Note that copper has two entries, these were taken as a 50/50 split between the two for each kg of copper. The stainless-steel selection is a corrosion resistant stainless steel. It's not sure that this is the exact material used in these components. However, other stainless steels have similar environmental impact characteristics, so the choice makes only a small difference and likely won't produce different results. A list of all materials used in the base case of this research is shown in Table 2.2. More details about the base case liquefier analyzed in this thesis is discussed in section 3.1.

Table 2.2: List of materials for the construction of a 125 TPD hydrogen liquefier with corresponding weight and database entries from the IDEMAT database.

Material	**Weight (kg)**	**Entry**
Concrete	4,307,402	A.040.07.101.230701 Concrete
Carbon Steel	499,946	A.100.02.101.230701 Steel hot rolled sheet USA
Stainless Steel	120,400	A.100.06.124.230701 X20Cr13 (420) 44% inox scrap (World)
Aluminium Alloy	157,800	A.100.24.103.231104 Aluminium trade mix (76% prim 24% sec)
Copper	27,737	A.100.30.098.230701 Copper (European Copper Instituut)
	27,737	A.100.24.110.230713 Copper wire, plate, pipe, trade mix (70% prim 30% sec)
Pipes	41,765	A.100.02.104.230701 Steel beams, pipes, sheet (from trade mix 44% recycled)

2.5 Power consumption

The power consumption of a hydrogen liquefier has two relevant components. The liquefaction process power, and the auxiliary power. They add up to the liquefaction plant power. The liquefaction process power can be calculated and modelled by engineers using various models and software. This figure is reported as the specific energy consumption (SEC) in the various liquefier designs in the literature. On the other hand, the auxiliary power is much harder to determine, as it involves all other plant systems such as vacuum pumps, control systems, computers, safety devices and other machinery that is not directly used in the thermodynamic liquefaction cycle. It should be noted that this additional power consumption is not included in the SEC figures that are reported in the literature, and thus, the environmental impacts are underestimated by using only this figure. Since it is quite difficult to determine the auxiliary power consumption of a hydrogen liquefaction plant without direct access to commercial liquefiers, little data has been produced on this aspect of hydrogen liquefaction.

2.5.1 Methodology for auxiliary power consumption

The only work that provides an assessment of the auxiliary power of a hydrogen liquefier is Stolzenburg et al. [29]. In this work, the auxiliary power consumption for a 40 TPD and a 50 TPD hydrogen liquefier are calculated. Since this is the only data available currently, it is used as a guiding factor for our LCA modeling. Based on these two data points, an exponential function of the form $a \cdot e^{-bx}$ was fitted, where a and b are fitting parameters and x is the liquefier capacity in TPD. This function was applied to all liquefiers to produce an adjusted SEC. The effective increase in power consumption is shown in Figure 2.3. A logarithmic scale is chosen to show that for increasing scales of LH_2 production, the auxiliary power as a percentage of total power consumed will approach zero. The two data points to which this function is fitted are also shown. For the 40 TPD liquefier, the auxiliary power was a 5.77% increase of the total, and for the 50 TPD liquefier, this increase was 5.46%. Note that two data points are insufficient for a conclusion of high certainty, and more experimental data is needed to improve the accuracy of this function.

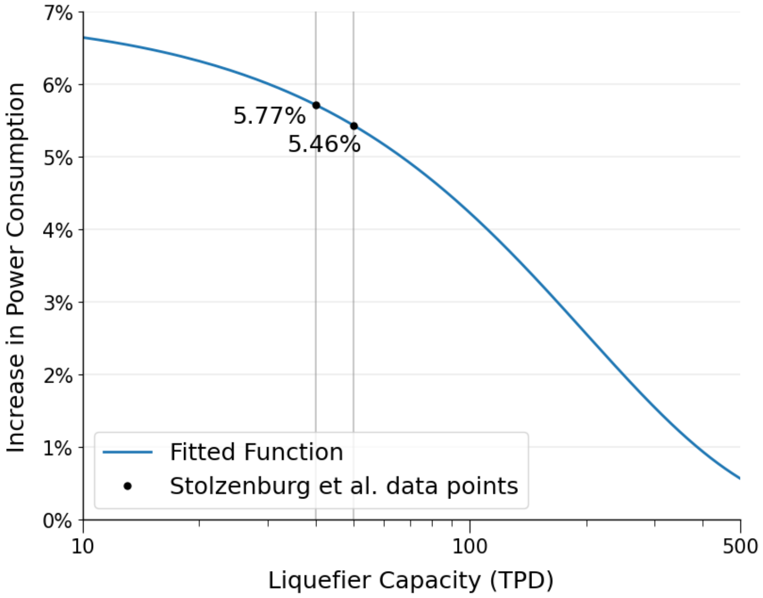

Figure 2.3: Effective increase in auxiliary power consumption as a function of liquefier capacity. The two data points for 40 TPD and 50 TPD from Stolzenburg et al. are shown [29].

To calculate this increase, we use the following function:

$$\text{Sizing Factor} = 1 + 0.0699 \cdot e^{-0.00502 \cdot \text{Capacity}} \tag{2.10}$$

This factor is multiplied by the liquefaction cycle SEC to produce the total plant SEC. In Figure2.3, this factor is presented as a percentage, with the increase in power consumption shown from the baseline of 100%.

2.6 LNG regasification integration

The integration of LNG regasification has the potential to utilize cold exergy that is currently being wasted by commercial processes. The power consumption reduction has been calculated alongside optimized theoretical liquefaction models in two of the reviewed studies. The reduction in power consumption from LNG integration was calculated in Stolzenburg et al. at 9.32% for a 50 TPD liquefier [29]. Kim et al. calculated a reduction of 10.70% for a 300 TPD liquefier [53]. We assume that this reduction effect is size-independent, as it is related to the thermodynamic cycle itself, and not the plant. The LNG is used to significantly reduce the cooling requirements of the precooling phase. Since this energy is normally wasted, it is seen as ”free”. As a simplification, the average reduction in power consumption is assumed to be 10%.
The LNG factor is defined as:

$$\text{LNG factor} = \begin{cases} 1, & \text{if no LNG regasification is used,} \\ 0.9, & \text{if LNG regasification is integrated.} \end{cases} \tag{2.11}$$

The adjusted SEC is calculated as:

$$\text{Adjusted SEC} = \text{SEC} \times \text{Sizing Factor} \times \text{LNG Factor} \tag{2.12}$$

The table below presents the SEC values for different scenarios, both with and without LNG integration:

Table 2.3: Adjusted SEC calculation for different scenarios.

SEC	**Capacity** [kWh/kg LH_2]	**Sizing** [TPD]	**LNG Factor**	**Adjusted SEC** [kWh/kg LH_2]
6.67	125	1.0373	1	6.91
6.67	500	1.0057	0.9	6.03

As shown in Table 2.3, the integration of LNG regasification consistently reduces the SEC values, highlighting the efficiency gains obtained by utilizing the available cold exergy in LNG regasification.

2.7 Sustainable energy production fluctuations

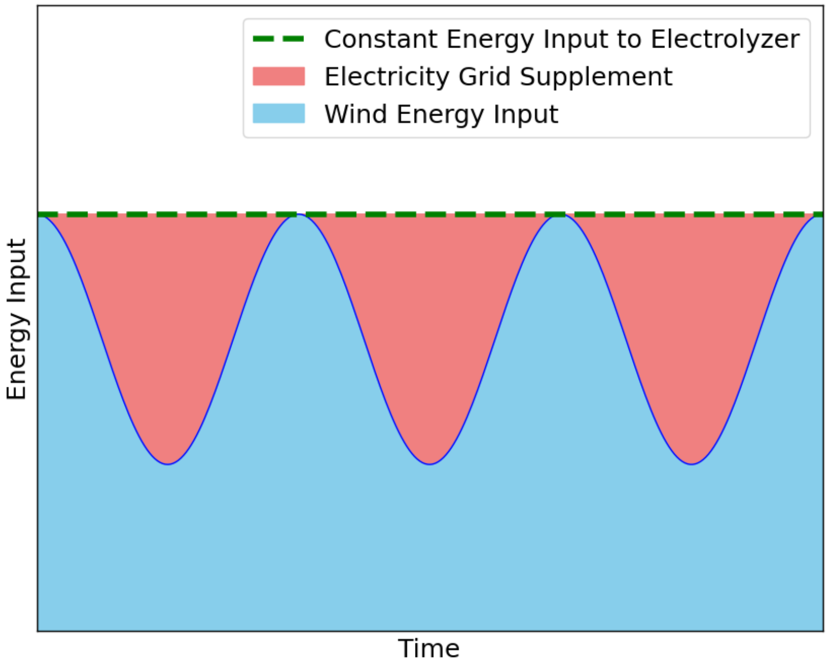

Figure 2.4: Simplified electrolyzer energy input: variable wind energy + grid supplement.

A relevant problem exists within the sustainable hydrogen value chain, which should not be neglected. Namely, the fluctuation of energy production rates in solar and wind energy. Solar and wind energy are intermittent sources of power, as the sun doesn't always shine, and the wind doesn't always blow. While a hydrogen liquefier and hydrogen electrolyzer both require a constant flow of electricity during normal operation, this cannot be provided by wind energy or solar energy alone. This concept is illustrated in Figure 2.4. In this figure, the dashed green line represents the required constant energy input of an electrolyzer. Wind energy output is represented in the blue area, which isn't always sufficient to supply the required amount. A supplement of grid energy is sometimes needed to constantly supply the electrolyzer input, represented by the red area. Hence, in some scenarios, 40% of the total energy supply is considered as grid energy. 40% is chosen arbitrarily as power capacity management is complex and the results of these calculations should be seen as an illustration of this fluctuation challenge. The example is given for wind energy and an electrolyzer, but the same issue of power fluctuation applies to solar energy, and a constant power requirement also applies to hydrogen liquefiers.

3. Results and Discussion

3.1 LCA scenario development

Base Case Hydrogen Liquefier – State-of-the-Art Mixed Refrigerant Liquefier

First, we define a hydrogen liquefier that will be the base case in all of the scenarios developed in this thesis. In each seperate section of these results we vary one of the parameters that define the hydrogen liquefier. Our base case is adapted from Tamarona et al. [63] and is consistent with the screening process in Al-Ghafri. et al [13]. Passing this screening process means that our base case represents a state-of-the-art hydrogen liquefier that includes efficient, novel technologies that are feasible today and are also suitable for upscaling.
This modern liquefier has a production capacity of 125 TPD and an SEC of 6.67 kWh/kg LH_2, which is in line with an efficient but realistic SEC when compared to the figures found Figure 1.4. The system uses a mixed refrigerant for an efficient precooling stage and hydrogen as the refrigerant in the cryogenic cooling stage. The lifetime of the liquefier is assumed to be 30 years. The composition of the mixed refrigerant is shown in Table 3.4 and Table 3.5, and the estimated raw construction materials are listed in Table 2.2. Additionally, the electricity used for powering the base case liquefier is chosen to be sourced from offshore windmills. Other sources of electricity are also analysed in this work, shown in Figure 3.5.

3.2 Hydrogen liquefier types comparison

Many older hydrogen liquefiers, such as the Legacy Liquefier in Leuna, Germany, are still in operation today [11]. We compare these legacy technologies with more modern alternatives. Our base case liquefier is compared to a legacy liquefier operational in Leuna, Germany. This Legacy liquefier uses nitrogen from air separation for precooling, an outdated method when compared to an optimized mixed refrigerant, and has a production capacity of only 5 TPD [13]. It also lacks LNG regasification, a feature missing in the base case as well. A third scenario is implemented that does integrate LNG regasification exergy. This integration further reduces electricity consumption, but requires some logistical difficulties to be overcome. This LNG enhanced liquefier is identical to the base case except for the LNG integration, for which the calculation is shown in section 2.6.

Table 3.1: Hydrogen liquefier type variation scenarios.

Scenario Name	Liquefier Size (TPD)	Adjusted SEC (kWh/kg LH_2)	Precooling Refrigerant	LNG
Legacy Liquefier	5	12.71	Nitrogen	No
State-of-the-Art Liquefier	125	6.67	Mixed refrigerant	No
LNG Enhanced Liquefier	125	6.22	Mixed refrigerant + LNG	Yes

In Figure 3.1 a detailed depiction of the three liquefier types is presented. It emphasizes the gate-to-gate scope of this LCA section, which focuses solely on the liquefaction phase within the broader value chain. The logistical challenges of LNG integration are also represented, as a third-party LNG terminal will need to operate simultaneously with the liquefier for this to be implemented.

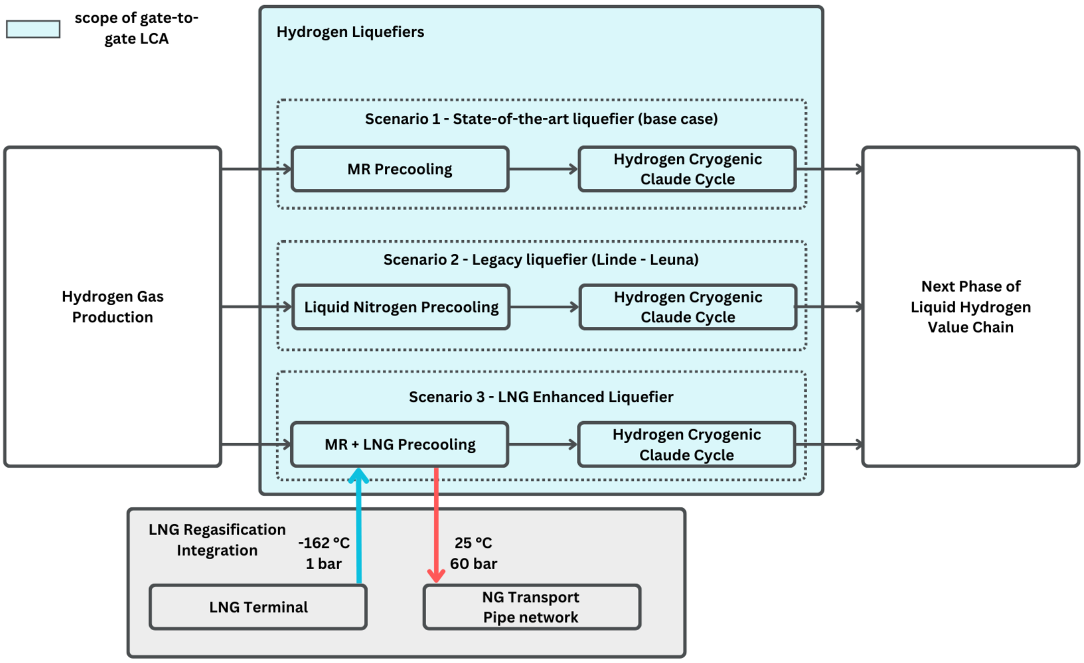

Figure 3.1: Visualisation of hydrogen liquefier type variation scenarios located in the hydrogen value chain.

The results are shown in Figure 3.2. The figure shows both source and result categories of environmental impacts. Electricity is by far the largest source of environmental costs, despite wind energy being the power source. Wind energy has a low ecocost when compared to other power sources, thus these results show the dominance of electricity as a source of ecocost. The legacy liquefier by Linde has a lower production capacity and lower efficiency, which explains why the power consumption has a significantly higher cost compared to the other scenarios. As for the refrigerants, it can be said that the legacy liquefier, which uses liquid nitrogen as a refrigerant, is more environmentally friendly than the MRs used by the modern liquefiers. However, the total amount of costs caused by refrigerant leakage is very small. For the LNG scenario, the most efficient system, the percentage of refrigerant contribution is the highest. Despite relatively large refrigerant leakage costs, it's still only 2.02% of total ecocosts. Refrigerant emission impacts can thus be seen as negligible, as long as no CFCs, PFCs, HCFCs and other halogenated hydrocarbons are used in the mixed refrigerant compositions. The inclusion of one or more of these compounds drastically affects the refrigerant leakage impact, as can be seen in Figure 3.6 by the entry of R14. Note that the highest estimate scenario of refrigerant leakage, 30% of total charge annually, was chosen for these scenarios. Aditionally, the construction materials were calculated based of off the base case, since no data was found on the legacy liquefier by Linde.

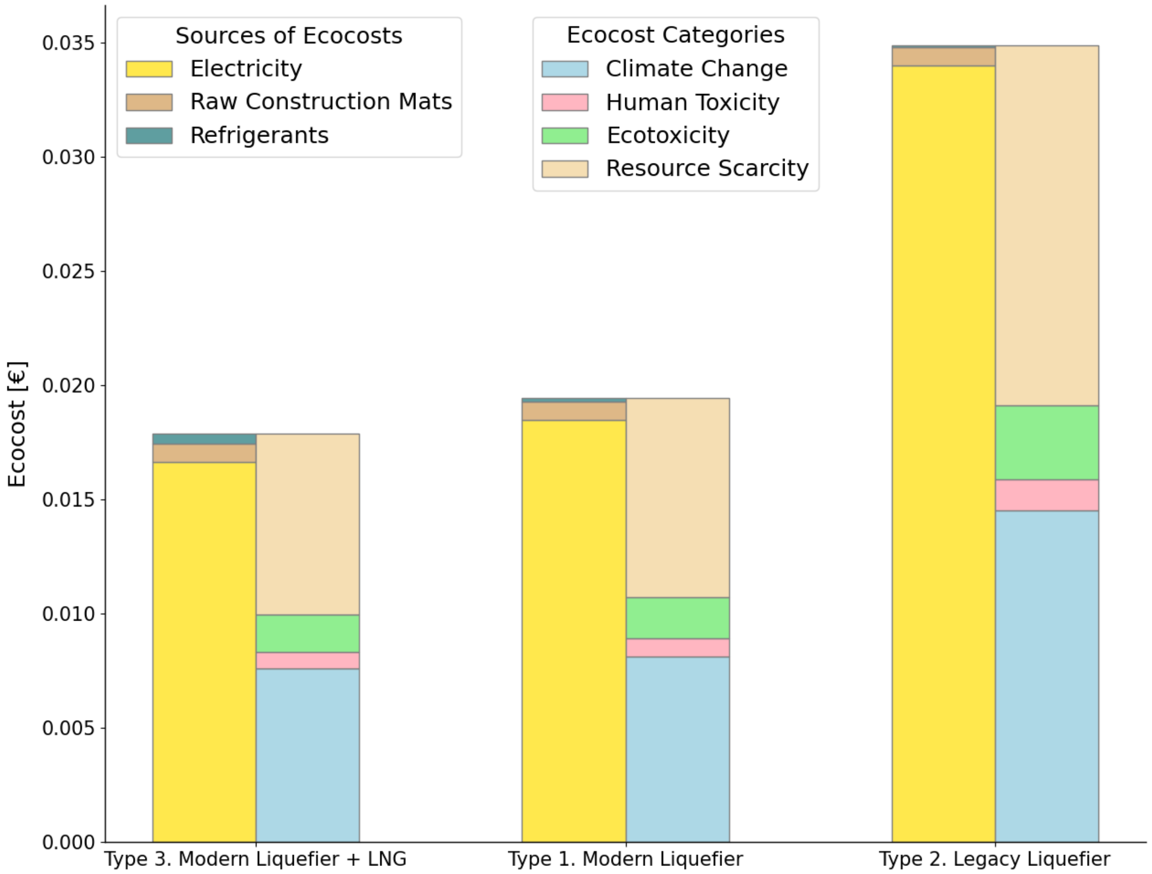

Figure 3.2: Environmental impact sources and resulting harm categories for liquefying 1 kg of hydrogen gas.

3.3 Liquefier construction materials

The raw materials that are required for the construction of a hydrogen liquefier are estimated in this work. These estimations, as well a other estimates that are pulled from the literature are compared on their environmental performance. The power consumption and refrigerant leakage are assumed equal to the base case for all liquefiers in this analysis. This means that only construction material impacts are varied and normalized to 1 kg of LH_2 production. The construction materials for Choe et al. [43] and Stolzenburg et al. [29] were taken directly from the supplementary materials. The construction materials for the base case and Kerkar et al. [65] were estimated in this work, for which the calculations are explained in section 2.4. The totals are shown in Table 3.2. There is a large gap in the estimation of concrete with respect to this work and the other two sources. A likely explanation is a difference in methodology. The other works did not explain their methodology, however. Another discrepancy is the missing of both copper and stainless steel from the Kerkar et al. entry. The reason for this is the lack of implementation of coldboxes, adsorbers and various other equipment pieces in the APEA files, which were used to produce these figures.

Table 3.2: Material requirements for various sources. The first part shows capacities, concrete, and piping requirements, while the second part lists metal materials.

Source	Capacity (TPD)	Concrete (kg)	Piping (kg)
Kerkar et al.	86	2,937,183	22,869
Tamarona et al.	125	4,307,402	41,765
Choe et al.	10	15,111,000	-
Stolzenburg et al.	50	46,620,000	-

Source	Carbon Steel (kg)	Aluminium (kg)	Copper (kg)	Stainless Steel (kg)
Kerkar et al.	428,700	30,180	-	-
Tamarona et al.	499,946	157,800	55,473	120,400
Choe et al.	123,000	50,000	49,000	193,000
Stolzenburg et al.	380,000	140,000	150,000	595,000

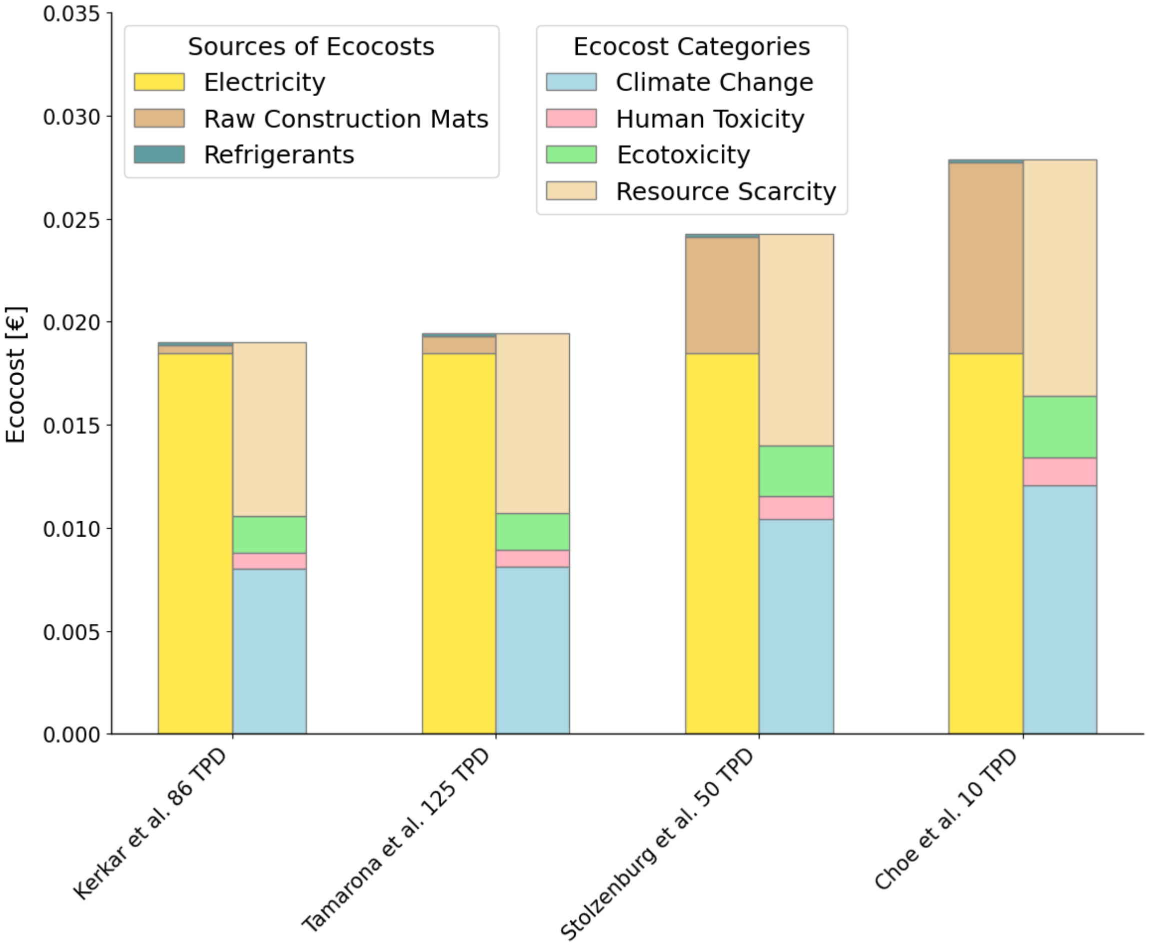

Figure 3.3: Eco-cost breakdown of liquefying 1kg hydrogen: construction material comparison by literary source.

Accurately determining the environmental impacts of large-scale industrial liquefier construction remains a challenge. As shown in Figure 3.3, the impacts of raw construction materials varies significantly across the available studies. While the size difference of the liquefiers likely plays a role due to normalization effects, scale alone cannot explain the 6.9-fold difference in environmental impacts between the 50 TPD and 125 TPD liquefier construction material impacts. The discrepancies most likely stem from differences in methodology that is used by the various studies to calculate construction material life cycle inventories (LCI). Further research is needed to accurately determine the environmental impacts of hydrogen liquefier construction materials. However, as shown in Figure 3.5, the construction material impact metrics lose significance when compared to the impact of power source selection. In a future where hydrogen liquefier power is sustainably sourced everywhere, the accurate calculation of construction material impacts may become relevant to researchers. Hopefully, by then, high quality data from commercial liquefiers will be available for public research.

3.4 Liquefier sizing scenarios

The impact of scaling up the liquefier system was analyzed. The base case liquefier, which was adapted from Tamarona et al. [63], has a capacity of 125 TPD. There are two upscales of this system present with capacities of 250 TPD and 500 TPD, respectively. Several configurations of these liquefiers are available; however, only the base designs were analyzed. Aspen Economic Analyzer files were used for this evaluation, which were provided by the original author following direct communication. The SEC of the upscales as adjusted according to subsection 2.5.1. This resulted in an adjusted SEC of 6.91, 6.80, and 6.67 $\mathrm{kWh\,kg^{-1}\,LH_2}$, respectively.

Table 3.3: Raw construction materials required for various upscales of a hydrogen liquefier.

Material	**125 TPD (kg)**	**250 TPD (kg)**	**500 TPD (kg)**
Concrete	4,307,402	7,240,516	14,107,621
Piping	41,765	79,470	186,835
Carbon Steel	499,946	904,418	2,184,640
Aluminium	157,800	373,150	757,950
Copper	55,473	102,234	381,645
Stainless Steel	120,400	206,500	400,900

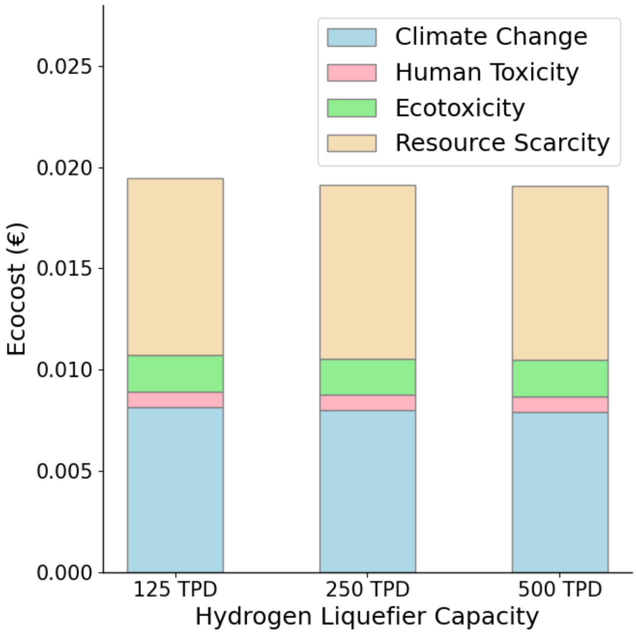

Figure 3.4: Eco-cost breakdown of the production of 1kg LH_2 for various upscales of base case hydrogen liquefier.

In figure Figure 3.4 the environmental effects of upscaling a hydrogen liquefier are shown. The difference in the scenarios is small, and barely visible to the naked eye. This doesn't mean that creating larger more efficient liquefaction cycles is useless. As technology progresses, larger facilities will prove to lend themselves for more efficient and specialized equipment which will lead to the decrease of the SEC. It reiterates the importance of achieving ever more efficient technologies, and the importance of SEC optimization research.

3.5 Liquefier power source scenarios

Hydrogen liquefaction uses vast amounts of electricity. The source of this electricity is therefore of primary concern to the environmental impact of the liquefaction process. In the scenarios in this section we take the base case liquefier but vary its power source. The analysis included sustainable power sources, fossil-based sources and electricity grid mixes which are a composition of power sources that is based on the current state of humanity.

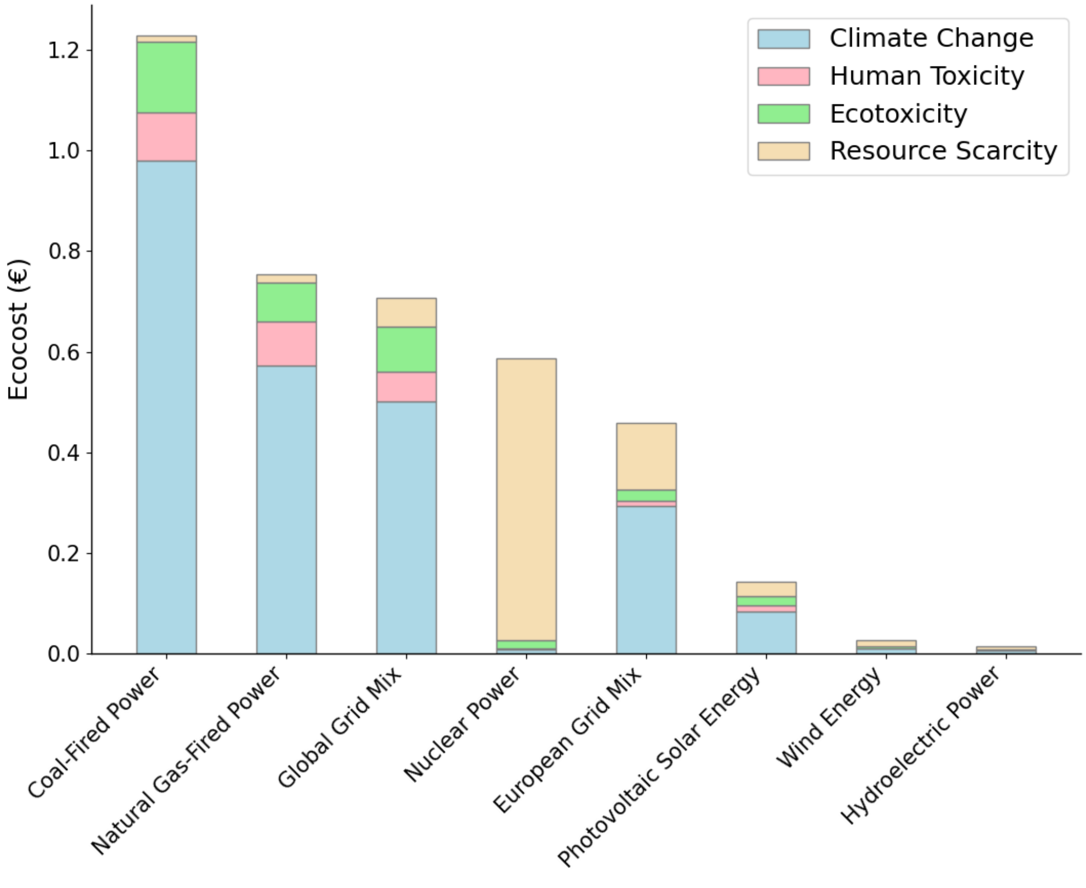

Figure 3.5: Eco-cost breakdown of the production of 1kg LH_2 for a hydrogen liquefier varied by power source.

Figure 3.5 shows the results of this analysis. the graph highlights that hydrogen liquefaction's environmental impacts are primarily driven by electricity consumption. The base case liquefier, represented by wind energy, appears minimal on the graph, significantly overshadowed by alternative, less sustainable energy sources. As such, the information in this graph exceeds the boundaries of hydrogen liquefaction itself and approaches an ecocost breakdown of power production. When compared to Figure 3.2 it can be seen that refrigerants and raw construction materials are made even further insignificant when environmentally inefficient power sources are considered. This graph reiterates the environmental impact profile of the most common power generation methods employed by humanity. In terms of Climate Change, coal and natural gas are the cause of extreme environmental harm. Since these two still make up a very large proportion of both the global and European electricity grid, their impact profile is very similar. Nuclear power also stands out, with its climate change potential rivaling that of the most sustainable options out there, but the limited global supply of uranium standing in the way, among other reasons [54]. PV energy is also a standout. Despite being far more environmentally friendly, it can be viewed as an inferior form of sustainable power generation when compared to its counterparts as of 2024. This conclusion is not widely known, but it has been repeatedly shown in the literature in recent years. The primary factor for this discrepancy is the mining and processing of the required materials for solar panels [66]. Further innovation in PV technology may improve its performance in the future.

3.6 Mixed refrigerants

Chemical compounds ecocosts compared

Figure 3.6 shows the environmental impacts of the leakage of various chemical compounds. This list of compounds was made by including the components used in the refrigerant mixtures found in the literature. The most prominent result shown in this graph is the inclusion of R14, a PFC (perfluorocarbon). This chemical compound is one on the list of many chemicals that are very useful as cooling fluids, but are also extremely harmful to the climate and/or ozone layer. It scores several orders of magnitude higher in ecocost than all other compounds, which is why an additional insert graph is included to represent its high ecocost. The next thing to note is the discrepancy of harmful effects between the various hydrocarbons. Methane stands alone with very high impacts to the climate, whereas the other hydrocarbons owe the bulk of their impact costs to the human toxicity category. In the updated climate potential impacts of the IPCC [47] it is shown that methane specifically has very high GWP, as it is very efficient at trapping heat in the atmosphere. Other hydrocarbons, have different breakdown mechanisms. These tend to break down into VOCs (volatile organic compounds), which produce various direct harmful effects to human and animal bodies [67]. Helium should also be mentioned, as it might seem strange that an inert gas would score so high on its environmental impact. The production of helium is to blame in this case, which is a product of cryogenic distillation of gas mixtures. This process is similar to liquefaction and is energy intensive. Nitrogen also is produced in this way, however since it contributes 78% of the air we breathe, this is far less costly. Neon is absent from the data, as it was not found in the database. A final note on these results is that they are based on 1kg of substance. Since these compounds are normally gases, their densities are quite different. 1kg of hydrogen will contain far more molecules than 1 kg of propane, for example. This is also why the heavier hydrocarbons represent lower amounts of environmental impact, which may seem counter-intuitive at first

In Figure 3.7 the ecocosts of various refrigerant mixtures are shown in their operating conditions. These mixtures were all sourced from the literature. Most of the mixtures are precooling mixtures, their composition shown in Table 3.4. the refrigerant mixtures for cryogenic cooling are presented with a CC indication, and compositions are shown in Table 3.5. Its very important to make this distinction, as the operating pressure and temperature of the precooling mixtures and cryogenic mixtures are different. The precooling mixture present in Ansarinasab et al. [30] contains R14, a very harmful compound. It is very prominent in Figure 3.7. The impact of R14 is so extreme that its climate change impact is several magnitudes exceeding the other mixtures. Apart from this, some other interesting conclusions can be made from these results. Namely, the cryogenic refrigerants are all very environmentally efficient, which is due to their large inclusion of hydrogen, which does not provide any harm to the environment when emitted into the atmosphere. This is a very positive outcome for hydrogen liquefaction technology, since the cryogenic cooling refrigerant closed loop is far larger than the precooling loop. For the base case, the volume of the cryogenic cooling refrigerant charge volume was estimated to be 9.37 times the size of the precooling refrigerant charge volume. Here, the low density of hydrogen plays a large role in keeping down ecocosts. From Figure 3.6 it would seem that hydrocarbons are competitive with hydrogen, however the switch to volumetric based results instead of mass based results corrects this perception. The densities of each mixture at operating conditions are shown in Table 3.6 The precooling refrigerant mixtures also

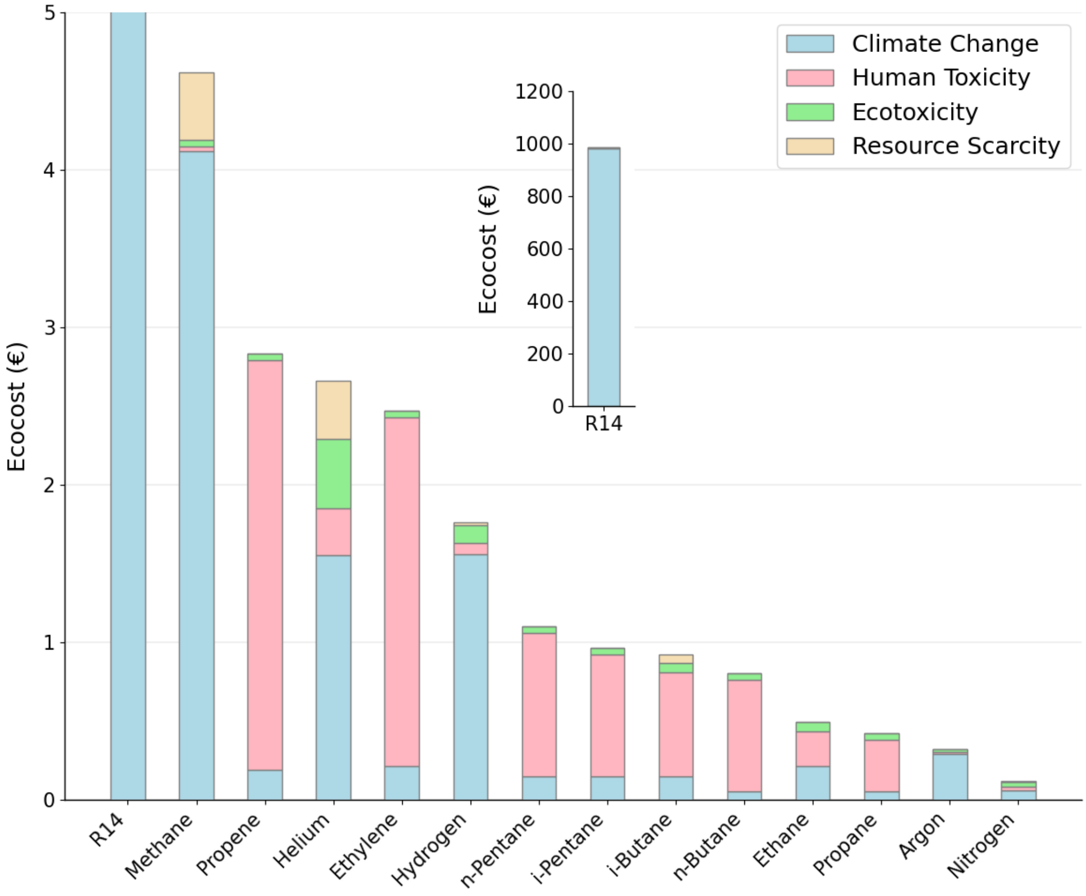

Figure 3.6: Eco-cost breakdown of leaking 1kg of various mixed refrigerant components.

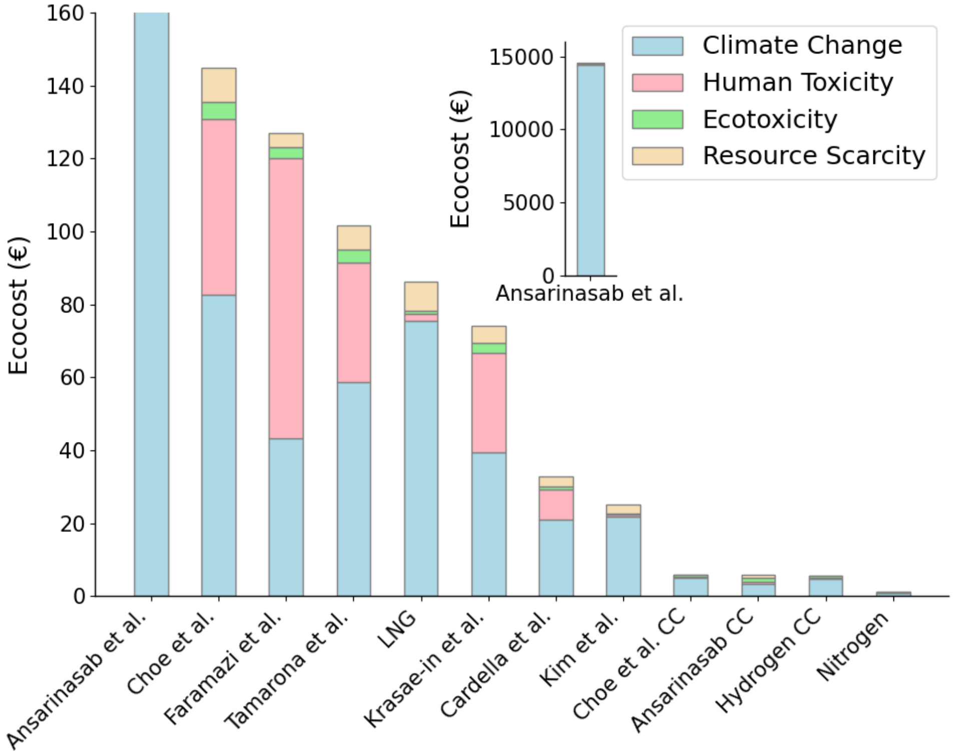

Figure 3.7: Eco-cost breakdown of leaking 1 m^3 of various refrigerant mixtures at operating conditions. For precooling mixtures $p = 22.71\,bar$ and $T = 224.8\,K$. For cryogenic cooling mixtures $p = 12.21\,bar$ and $T = 95.31\,K$. cryogenic cooling mixture are denoted with 'CC'.

show some significant variations in their environmental efficiencies. Where nitrogen is still a winner from this perspective, the more energy efficient hydrocarbon mixtures have some interesting variations in terms of climate change vs. human toxicity effects. Roughly speaking, a higher percentage of smaller hydrocarbons such as methane and ethane result in higher climate change effects, and the larger hydrocarbons produce more human toxicity. Still, these results are all based off the assumption that their operating state is the same for all compounds, which can not be true in reality. Still, the result of Figure 3.2 holds. Namely, that refrigerant leakage in hydrogen liquefiers has an insignificant environmental impact, for all precooling and cryogenic cooling mixtures, as long as no halogenated hydrocarbons are used. A small note should be added. Neon was not included in the calculations in these results, since it was not available in either database used. This only affects the Ansarinasab et al. CC mixture.

Table 3.4: Mass fractions of precooling mixed refrigerant components for various references.

Components	Ansarinasab et al.	Choe et al.	Tamarona et al.	LNG	Krasae-in et al.
R14	0.172631				
Methane	0.066858	0.17268	0.156	0.8254	0.118
Ethylene	0.11004				
Hydrogen	0.000495				0.002
n-Pentane	0.265354			0.0004	
i-Pentane					
i-Butane		0.03143			0.5438
n-Butane	0.028501	0.53710	0.471	0.0294	0.465
Ethane	0.051609	0.23827	0.247	0.0904	0.259
Propane	0.194629	0.01185	0.041	0.0535	
Nitrogen	0.109883	0.00868	0.085	0.0009	0.155
Neon					

Components	Cardella et al.	Kim et al.	Faramazi et al.	Nitrogen	Kerkar et al.
R14					
Methane	0.2001	0.1898	0.098016		0.0759
Ethylene			0.280261		0.0963
Hydrogen	0.0231				
n-Pentane		0.0001			0.2477
i-Pentane			0.298802		
i-Butane					
n-Butane		0.0068			0.1179
Ethane		0.0208			0.0772
Propane		0.0123	0.215842		0.2420
Nitrogen	0.2330	0.7702	0.107078	1	0.1250
Neon					0.0180

Table 3.5: Mass fractions of cryogenic cooling mixed refrigerant components for various references.

Components	Hydrogen	Ansarinasab et al.	Choe et al.
Hydrogen	1	0.022567	0.93333
Helium		0.605201	0.06667
Neon		0.372232	

Table 3.6: Density of refrigerant mixtures at operating conditions. For precooling mixtures p = 22.71 bar and T = 224.8 K. For cryogenic cooling mixtures p = 12.21 bar and T = 95.31 K. cryogenic cooling mixture are denoted with 'CC'.

Refrigerant Mixture	**Density [kg/m^3]**
Choe et al.	104.3
Tamarona et al.	103.31
Kerkar et al.	101.57
Ansarinasab et al.	85
Faramazi et al.	82.42
Krasae-in et al.	68.88
Nitrogen	31.842
Kim et al.	26.9545
LNG	22.067
Cardella et al.	22.067
Ansarinasab et al. CC	3.5
Choe et al. CC	3.2458
Hydrogen CC	3.1433

3.7 Hydrogen value chain scenarios

This section describes realistic value chain scenarios that involve LH_2. Whereas the previous sections describing scenarios all dealt with **gate-to-gate** LCA, This section deals with scenarios that are **cradle-to-grave**. The difference between these types of LCA was explained in section 1.5. The hydrogen liquefier used in all the scenarios in this section is the base case liquefier from section 3.1. That means that we do not compare liquefiers in this sections. Instead, we compare its various value chain logistics. The scenarios analyzed in this section are shown in Table 3.7. These scenarios represent real-world potential opportunities for the utilization of hydrogen as a carrier of sustainable energy. Countries are chosen with a local sustainable power source that can be used to produce hydrogen, which is then subsequently liquefied and transported over sea to the Port of Rotterdam in the Netherlands. Namely, hydro-electric power from Norway, nuclear energy from Canada and solar power from Chile. These energy carrier solutions are also compared to a scenario which doesn't require sea transport, where hydrogen is produced in the Netherlands locally using wind energy. Liquefaction is included in all scenarios because this may be required for storage.

The Netherlands, with many wind parks and plans to expand their capacity, and the major seaport of Rotterdam, is an ideal location for both LH_2 delivery and sustainable hydrogen production. Norway was chosen as a sensible location for hydroelectric power, due to the 4000+ dams in the country. Chile was chosen for PV arrays. Given the ample sunlight opportunities in Chile, this may offer economically attractive business cases for solar arrays if premiums are payed for sustainable energy in the European Union. Canada was chosen for nuclear energy because of three characteristics, namely existing nuclear facilities, local uranium deposits in the earth, and abundance of space. The shipping routes are shown in Figure 3.8. End usage of hydrogen in fuel cells is not included in scenario analysis because hydrogen fuel cells only emit water into the atmosphere.

In scenarios 2 and 7, a mix of grid electricity and renewable energy sources is used to simulate the variability of renewable energy supply of these sources as discussed in section 2.7. Additionally, scenario 3 is included, which uses grid electricity exclusively, highlighting the potential for 'greenwashing'—where companies could misleadingly market hydrogen as sustainable while relying on non-renewable power. This serves as a worst-case scenario, illustrating the environmental implications of false sustainability claims.

Table 3.7: Hydrogen value chain scenarios overview.

Scenario	Power Source	Production Location	Liquefaction	Transport Distance (km)	BOG Losses (%)
1	Wind farms	Netherlands	No	n.a.	n.a.
2	40% Wind, 60% Grid	Netherlands	Yes	n.a.	n.a.
3	100% Grid	Netherlands	Yes	n.a.	n.a.
4	Hydroelectric dams	Norway	Yes	1000	0.48
5	Nuclear power plants	Canada	Yes	3000	1.39
6	Solar panels	Chile	Yes	15000	7.09
7	40% Solar, 60% Grid	Chile	Yes	15000	7.09

Figure 3.8: Hydrogen value chain scenario sea transport routes. From short distance to long distance: Norway - Netherlands $\sim$ 1000 *km, Canada - Netherlands* $\sim$ 3000 *km, Chile - Netherlands* $\sim$ 15000 *km.*

A PEM Electrolyzer will be used for all hydrogen production in this cradle-to-grave LCA. The SEC of hydrogen production of PEM electrolysis is 56.3 kWh/kg according to Fragiacomo et al. [68]. Only the power cost was included in the environmental impact modelling for the PEM electrolyzer. To model the sea transport of LH_2, we use the highly similar LNG carrier. LNG carriers typically travel at speeds ranging from 15 to 20 knots (1 knot = 1.852 km/h). We assume the average speed for LNG carriers is 19 knots, which is equivalent to 35.2 km/h.

BOG effects are not yet accounted for using this approach. Al-Ghafri et al. reports storage tank daily boil-off rates for LH_2 transport of $< 0.4\%$. In the near future, it is likely that these losses will be reduced using new innovations like improved insulation and tanker engines that can use the BOG as fuel. For this analysis, the BOG is assumed to be vented, and the total losses are shown in Table 3.8. Additionally, LNG has a different density compared to LH_2. This was also compensated in the calculations. At sea transport conditions, LNG is 6.35 times denser than LH_2, which means more ships are needed to transport the same weight of LH_2.

Table 3.8: Travel time and boil-off gas (BOG) loss calculation for sea transport between different countries and the Netherlands. The travel time (in days) is based on a constant speed of 35.2 km/h, and the total BOG loss (%) is calculated using a constant daily loss rate of 0.4% [13].

Route	**Distance (km)**	**Travel Time (days)**	**Total BOG Loss (%)**
Norway – Netherlands	1000	1.18	0.48
Canada – Netherlands	3000	3.55	1.39
Chile – Netherlands	15000	17.75	7.09

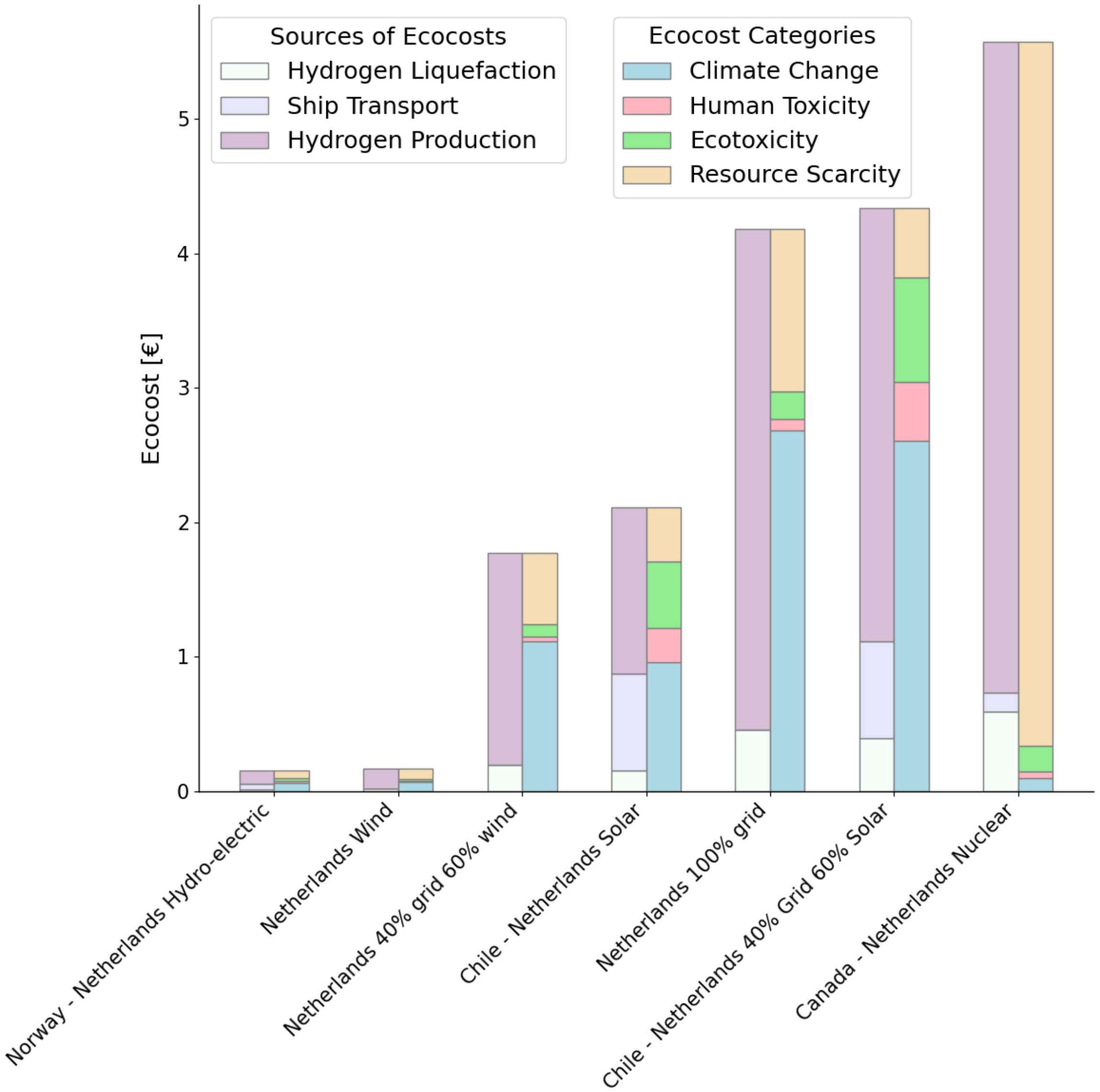

Figure 3.9: Eco-costs of delivering 1kg hydrogen to the port of Rotterdam by different production locations & energy sources.

The cradle-to-grave LH_2 value chain scenario results are shown in Figure 3.9. It can be seen from this data, that the total value chain has a totally tranformative impact on the sustainability of hydrogen technologies. Where optimistic cases such as local dutch wind energy and hydro-electric energy from Norway are very environmentally efficient, far away solar arrays in Chile with long transport distances score very poorly. The fact that using 40% local grid energy is competitive with the optimistic Chile scenario, where 100% of the energy is supplied by solar panels (impractical), is alarming. Solar panels creating hydrogen seems at face value like very sustainable choice. However, if PV technologies are not improved drastically, this scenario cannot be seen as green. Even more

alarming, is the fact that the 40% grid scenario in Chile is outcompeted by 100% grid hydrogen. The scenario where hydrogen is fully produced by grid electricity, is far less environmentally efficient than even fossil fuel based options. Of course, European laws forbid this type of hydrogen production. However, without sufficient oversight, grid-based hydrogen may supplement sustainable hydrogen in large quantities. This non-sustainable hydrogen may come cheaply to European markets in the future, once the hydrogen economy is built to scale. It cannot be underemphasized that such a scenario would be environmentally counterproductive. Clear and realistic rules are needed for the hydrogen electrolysis process. Strict oversight must be implemented, lest any theoretical environmental gains will remain theoretical only.
The extreme case of nuclear energy must also be considered. The ecocost of its climate change impact alone is very low. This means that, for climate change reduction, it must be considered viable. However, the potential for nuclear catastrophe is something to consider. Nuclear power plants are a political choice, and if climate change reduction is prioritized, nuclear plants can be considered. Innovations to safety of uranium based nuclear facilities, or alternative nuclear fuel types which are inherently safe, could vastly improve the performance on this front. Such an innovation would potentially solve all sustainable power requirements for humanity and end man made climate change.

3.8 Limitations

Change of impacts

As of 2024, climate change is hot topic and a widely known phenomenon. Significant uncertainties persist in environmental harm modeling. For example, the CO_2-eq of butane is largely reported to be 4, which is old and outdated information. According to the sixth assessment report from the IPCC [69] new research determines the CO_2-eq of butane to be 0.006 instead of 4, which makes a major impact on LCA research including this work. Further revisions and updates in environmental harm modeling may shift future and past results of LCA research.

LNG integration

The heat integration of LNG into a hydrogen liquefier involves extra logistical steps, which are very hard to model. If we assume the most thermodynamically efficient method of LNG integration according to Kim et al. [53], LNG is used alongside the precooling refrigerant inside the coldbox. In this research, the following simplifications were made. The loss of LNG due to leakage was assumed to be equal in weight to the amount of mixed refrigerant that would be lost if there were no LNG integration. This LNG loss was added to the base mixed refrigerant loss that would occur in the non-LNG case. The result then, should overestimate this refrigerant leakage, which was deemed adequate because the effects of refrigerants were themselves insignificant. The required extra piping, manpower and other requirements for LNG integration were not modelled in this work, since no such information was found. The legal and practical side of LNG integration is a far tighter bottleneck for implementation.

Legacy liquefier assumptions

Some assumptions were necessary for the Line liquefier in Leuna used in the type scenarios. It is assumed that the same weight of nitrogen refrigerant is used per kg of LH_2 as is used by refrigerant mixture from the base case. The density of nitrogen is roughly 3 times lower than the hydrocarbon based precooling mixture used in the base case. This means that using the same weight of nitrogen would likely overestimate the amount of nitrogen leaked. However, as we'll see in the results, refrigerants have a negligible effect on environmental impacts and this barely makes any difference.

The raw construction materials of the old liquefier were also assumed to be the same as the base case, as there is no data available on this.

Power transport distance

An issue in assessing the environmental impacts of electrical power is the assessment of electricity cable transport. For the full value chain LCA scenarios, the electricity travel distance and type of power cables (onshore vs offshore) needs to be determined for each case. Initial work was done on offshore wind energy, where the largest environmental effects from cabling was anticipated, as underwater electricity cables are challenging to place and maintain. However, after accounting for the additional cabling requirements that were not already included in the databases, the extra cables made no significant change to environmental impacts. Below you will find this initial exploration of including extra electricity cables in the LCA. The electricity from offshore windmills in IDEMAT are assumed to have a total efficiency of 47% and lifetime of 20 years. They are modelled as 5MW capacity windmills with each an added 1km of sea cables. Typically, wind turbines are spaced 1km apart, which implies that the cables connecting each windmill are already included in the database entry, but the cable from the wind farm to the shore is excluded. So next we calculate the length of this sea cable and examine its environmental impacts. Since we're using the Port of Rotterdam as a location, a sea cable to one of the many local wind park would stretch roughly 25 km from the wind farm to the shore and would need to be only fractionally attributed to the liquefier since it uses only part of the total power production capacity of the wind park. To use and example, Wind Park Lichterduinen has a capacity of 760 MW, assuming the efficiency of 47%, and an adjusted liquefier power usage of roughly 3.6 MW of the base case liquefier scenario we can calculate that 4.74% of the total capacity of the wind farm will be used by the liquefier. This exact percentage must then be attributed to the liquefiers share of the electricity cable construction and maintenance. Of a 25 km European high voltage sea cable, the liquefier should be attributed $\sim 1184m$ of total cable length over its entire lifetime of 30 years. These impacts are so low that they really reiterate the positive environmental effects of sea-based wind parks, despite the hassle of underwater power cables. A similar calculation was done for Norway. Where a 75 km high voltage European cable is connected from a Norwegian seaport to one of the 4000+ hydroelectric facilities that Norway has, and these results are similarly insignificant.

LH_2 losses during storage

Daily storage losses are reported to be as low as $< 0.1\%$ per day [13]. However, this hydrogen is unlikely to be vented; rather, it is more economically efficient to re-liquefy it. These extra power costs are assumed negligible in this thesis. Economies of scale will also most likely significantly reduce storage losses.

Feed pressure

When Hydrogen gas enters the liquefaction plant, it is usually assumed to be around 20 bar. One of the first processes in the liquefaction plant is the compression of this hydrogen gas feed to about 80 bar, since this is required for efficient cooling. This step can be skipped in cases where hydrogen gas feed pressure is already 80 bar, which is realistic in the future as electrolysers are expected to produce such high-pressure hydrogen gas products. The effect is a reduction in power consumption of 0.74 kWh/kg LH_2 [29] in SEC for all hydrogen liquefiers alike. Although this effect would contribute to a significant reduction in power consumption, it is not further implemented in this research, since it is more of a property of the hydrogen production, and not the liquefaction. High-pressure hydrogen is by itself a gas product of higher energy and will also potentially reduce the power consumption requirements for other processes. Thus, the reduction in environmental impacts should be attributed to the Electrolyzer.

4. Conclusions

The environmental impacts of hydrogen liquefaction

The source of the environmental impacts of hydrogen liquefaction were found to be dominated by the power consumption. Even when the power source was wind energy, which is highly sustainable, 94.97% of ecocost of hydrogen liquefaction originated from power consumption. The resulting environmental impacts of hydrogen liquefiers are thus essentially the same as the environmental impacts of its power source. The result that power input is so dominant implies that the simple method used to model hydrogen liquefaction in previous studies seems mostly sufficient. Taking the SEC of a theoretical liquefaction cycle, and calculating environmental impacts using solely this metric is sufficiently close to the reality in most cases. Three categories of environmental impact sources were identified, power consumption, refrigerant leakage and raw construction materials. Refrigerant leakage was shown to be insignificant for environmental impacts in virtually all cases, the highest contribution was found to be 1.59% for a liquefier that includes LNG integration. The low impact of refrigerant leakage is due to the fact that efficient refrigerant mixtures for hydrogen liquefiers do not contain harmful halogenated compounds, but use hydrocarbons or inert gases instead. These refrigerants mixtures are orders of magnitudes more environmentally efficient and provide little harm to humanity when emitted into the atmosphere at legally permitted rates. There is one exception, where R14 was used in a refrigerant mixture. This refrigerant mixture was calculated to have a 14,311.84% increased ecocost when compared to the base case mixture on volumetric basis at operating conditions, which highlights the dangers of halogenated refrigeration compounds. Liquefaction plant construction material impacts were shown to only have a significant impact when considering very environmentally efficient electricity inputs such as wind energy or hydropower. the construction materials accounted for only 2.80% in the base case where wind energy was used. This result, however, is not conform other research data, which estimates a larger quantity of materials, primarily concrete. One other source resulted in an increase of construction material impact by 490.4%, or 23.19% of total impacts. Auxiliary power requirements for hydrogen liquefaction were also analyzed. For liquefiers of significant production capacity, auxiliary power requirements can be assumed to approach zero. A simple formula was constructed in Figure 2.3 to estimate this auxiliary power requirement based on liquefier size. However, this formula is based on too little data to be conclusively adopted, and additional data is needed to update it. Data was gathered and models were created in this work that should offer LCA researchers sufficient information to model hydrogen liquefaction with reasonable certainty until the technology is further in its adoption curve.

Hydrogen Value Chain

The environmental impacts of LH_2 value chain scenarios were analyzed. The scenarios represent realistic supply chain routes of LH_2, comparing various transport distances as well as energy sources, choosing the port of Rotterdam in the Netherlands as destination. Hydrogen production, liquefaction and transport from Norway using hydro-electric power was found to have virtually equal environmental impact as local production through wind energy. Hydrogen production, liquefaction and transport from Canada using nuclear energy was found to have the highest impact of all scenarios when including all ecocost categories. However, when considering only the climate change impact nuclear based hydrogen from Canada scores third best with only a 39.6% increase in climate change impact compared to the local hydrogen production through wind energy in the Netherlands with no transport. The impact of the liquefaction of hydrogen ranges from 12.29% to 13.13% of its production impacts. Ship transport impacts are directly related to the distance travelled, with its highest relative impact being 34.23% of total impacts in the Chile solar scenario. Energy from the grid was used to supplement sustainable energy or even replace it completely in some scenarios to simulate energy production fluctuation and greenwashing. The performance of the Dutch grid supplement scenario, where 40% of energy inputs are non-sustainable, are comparable to the performance of long distance solar based hydrogen from Chile. This is alarming since solar power is seen as a sustainable power source. compared to the wind energy base case the environmental impacts of solar based LH_2 from Chile is 12.43 times higher for the total impacts, and 8.21 times higher when comparing only the climate change impacts. The results provide a counter argument to the notion that solar based hydrogen is a sustainable alternative to fossil fuels in its current state. The greenwashing scenario was calculated to have the highest climate change impact. Despite having no need for any transport, the impact on climate change of hydrogen production and liquefaction using European grid electricity is 38.07 times higher than using wind energy, which reiterates the importance for proper oversight of sustainable hydrogen production and liquefaction companies.

5. Recommendations for future research

One of the main goals of this thesis was to create a model for future LCA researchers to apply to their work. This work provides the tools for this purpose. The following method is the simplest version, and any of the steps can be improved upon with more detailed analysis of the data in this work, or other novel and original data from future studies.

Simplified Method for Estimating Environmental Impacts of Hydrogen Liquefaction:

1. Choose SEC of liquefaction based on goal and scope of research. The base case in this thesis used a figure of 6.67 kWh/kg LH_2. This is the theoretical power consumption of the thermodynamic liquefaction cycle.
2. Use formula in Figure 2.3 to adjust SEC based on liquefier capacity to compensate auxiliary power consumption. For the base case, this adjusted SEC was calculated to be 6.91 kWh/kg LH_2. This adjustment includes the auxiliary power consumption. It accounts for all the power consumption of the liquefaction plant as a whole.
3. Add 2.5% to compensate for construction materials and refrigerant leakage (30 years liquefier lifetime). A more detailed analysis was used in this thesis, but an addition of 2.5% is a very rough approximation that is sufficient for simple analyses.
4. Use the calculated power consumption to calculate the environmental costs using a chosen power production source, such as wind or solar power.

Improving accuracy of hydrogen liquefaction LCA

In this research, public data of high quality was often hard to find for certain processes. The data that could be used to improve the accuracy of the results in this work are the following: Material compositions of specialized hydrogen liquefaction equipment, the total refrigerant weights/volumes, yearly refilling rates, and the real plant electricity usage vs theoretical. Given this data, an updated assessment could be made of hydrogen liquefaction.

Refrigerant leakage impact

The environmental impact of refrigerant leakage is negligible since no CFCs or HCFCs are used. Therefore, leakage controls can be relaxed compared to industry standards if it facilitates increased adoption of hydrogen liquefaction technologies, as long as sustainable power sources are used for hydrogen gas production and liquefaction.

Netherlands and Norway as a sustainable energy exporters

According to the findings of this work, only the most sustainable electricity production sources offer a sustainable energy transport future using LH_2. This means that both hydro-electric and wind energy offer promising use-cases of the LH_2 value chain. The Netherlands is a global front-runner in wind energy with many plans for future wind farms and Norway is a hotspot of mountains which are suitable for hydro-electric power generation. If local power production is in excess of power need, these nations may consider adapting LH_2 exports to net energy importing nations. For many sustainable energy sources, the issue of power fluctuation still remains a major challenge, as only running electrolysers and liquefiers part-time is unlikely to be financially feasible in the foreseeable future.

Fluctuation problem power capacity management analysis

The results in Figure 3.9 can be used to produce well-adjusted power management policy for sustainable hydrogen production via electrolysis. A requirement of 100% sustainable electricity from fluctuating energy sources could make hydrogen production economically nonviable. On the other hand, lax requirements would allow the production of environmentally unsustainable hydrogen though the supplementation of too much grid mix electricity. Future research should attempt to find a balance that both retains low-environmental impacts and achieves a constant energy supply. This may prove impossible, removing the potential for using wind and/or solar power for sustainable hydrogen production. Alternatively, variable Rate Electrolyzers could also help solve this issue.

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