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Sustainability analysis and simulation of a Polymer Electrolyte Membrane (PEM) electrolyser for green hydrogen production

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Summary:

In recent years, green hydrogen has emerged as one of the energy carriers for the future sustainable development of our society. Due to its non-emission of pollutants during its generation and use, hydrogen is seen as a perfect substitute for current fossil fuels. However, the main drawback of producing hydrogen via water electrolysis and using renewable energies to supply energy is its lack of economic competitiveness compared to existing energy carriers. Therefore, this thesis focuses on analysing the sustainability of a green hydrogen production plant, not only considering its environmental parameters but throughout its Life Cycle Assessment (LCA).

The Polymer Electrolyte Membrane (PEM) is selected as the most prominent method of green hydrogen production in the medium to long term. Subsequently, a small-scale production plant is simulated using Aspen HYSYS software, which provides key data for evaluating sustainability indicators. The selected indicators are based on the Gauging Reaction Effectiveness for the Environmental Sustainability of Chemistries with a Multi-Objective Process Evaluator (GREENSCOPE) methodology, and they are used to compare the simulated PEM plant with an Alkaline Water Electrolysis (AWE) plant, which is a more mature technology. Finally, to analyse the real feasibility of this process, a scaling-up process of the process simulation is carried out.

Preface

The present thesis was developed as part of an ERASMUS+ exchange program, which provided an invaluable opportunity to conduct research in Norway and gain exposure to a new culture and novel ways of working. I am deeply grateful to the *Universitetet i Sørøst-Norge (USN)* for their warm welcome and seamless integration into their academic community from day one. Furthermore, I would like to acknowledge the *Escola Tècnica Superior d'Enginyeria Industrial de Barcelona (ETSEIB)* from the *Universitat Politecnica de Catalunya (UPC)* for giving me the chance to participate in this exchange program, and for providing education over seven years that proved to be of significant value to the development of this work.

I would also like to extend my thanks to Associate Professor Gaurav Mirlekar, who served as the supervisor for this thesis, and whose unwavering commitment to excellence was instrumental in its successful completion. Not only did he provide guidance throughout the research process, but he also offered the opportunity to develop an article to be presented at the 64th SIMS Conference, thereby providing an avenue for immersion into the world of scientific research.

Finally, despite the physical distance between us, I would like to express my gratitude to my family and friends for their unwavering encouragement and support throughout this challenging and rewarding endeavour.

Porsgrunn, 15th of May 2023

Jordi Béjar Rabascall

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Nomenclature

Acronyms

ACM	Aspen Customer Modeler
ATR	Auto-Thermal Reforming
AWE	Alkaline Water Electrolysis
CCS	Carbon Capture and Storage
$C_{E,spec.}$	Specific Energy Costs
CEFIC	European Chemical Industry Council
CO_2	Carbon dioxide
CPS	Chemical Process Simulation
EPA	Environmental Protection Agency
FWC	Fractional Water Consumption
G20	Group of 20
GHG	Greenhouse Gases
GREENSCOPE	Gauging Reaction Effectiveness for the ENvironmental Sustainability of Chemistries with a Multi-Objective Process Evaluator
GWP	Global warming potential
H_2	Hydrogen gas
H_2O	Water
IEA	International Energy Agency
IRENA	International Renewable Energy Agency
KOH	Potassium hydroxide
LCA	Life Cycle Assessment
LHV	Lower Heating Value
MLI	Mass Loss Index
NaOH	Sodium hydroxide
O_2	Oxygen gas

PEM	Polymer Electrolyte Membrane
R-squared	Coefficient of determination
SIMS	Scandinavian Simulation Society
SMR	Steam Methane Reforming
SOE	Solid Oxide Electrolysis
US	United States
ηE	Resource Energy Efficiency

Units

ºC	Celsius degrees
$	Dolar
€	Euro
Ω	Ohm
A	Ampere
bar	Bar
h	hour
J	Joules
K	Kelvin
kg	Kilogram
kWh	Kilowatt-hour
m^2	Square centimetres
m^3	Cubic meters
Mt	Millions of tons
N	Newton
V	Voltage
W	Watt

1 Introduction

The groundwork for this thesis is based on establishing the background, motivation, objectives, methods, scope, scientific contribution, and structure of the report.

1.1 Background

In the coming years, the world will face the challenge of changing the current fossil fuel-based energy model, which is responsible for emitting most of the polluting greenhouse gases that accelerate global climate change (Perera and Nadeau, 2022). Due to its storage capacity, its occurrence in nature and its physical properties, hydrogen is considered by institutions, governments and researchers as one of the energy carriers of the near future. The main advantage of hydrogen is that its use and production in a more environmentally friendly way creates a carbon-neutral scenario that can help reduce current global warming problems. (Zhou *et al.*, 2022).

A perfect example of this change is the Norwegian government, which in 2020 set out a clear strategy to develop hydrogen as the future energy resource. This strategy has been developed not only with technological progress in mind but also with financial viability, security and ease of access. Furthermore, Norway's natural water resources make it one of the countries with the greatest potential for producing green hydrogen through water electrolysis. Currently, 92% of the country's electricity is generated from this resource (IEA, 2022a), which means that a large amount of water is available and the electricity needed for hydrogen production can come exclusively from renewable resources (Norwegian Ministry of Petroleum and Energy and Norwegian Ministry of Climate and Environment, 2020).

1.2 Motivation

Hydrogen has great potential as an important energy carrier in the future, so green hydrogen production methods, such as those based on PEM technology, should be prioritised over those based on non-renewable resources (Ayers *et al.*, 2021). However, steam methane reforming (SMR) is currently the prevailing method. The primary reason is that current green methods are not economically efficient, making them less competitive than those based on fossil fuels (Younas *et al.*, 2022). Therefore, research into the sustainable development of green hydrogen production methods is essential. Furthermore, advancements in simulation methods and scale-up processes can help in reducing costs during the design phases (Ramsey *et al.*, 2000).

1.3 Objective(s)

The objectives of this thesis are as follows:

- O1. Conduct a literature review to understand the current status of green hydrogen production methods and their sustainability.
- O2. Perform a simulation of a PEM electrolyser using Aspen HYSYS software.
- O3. Carry out a sustainability analysis using the GREENSCOPE approach.
- O4. Develop a scaling-up process and compare simulation performance with existing PEM electrolysers.

1.4 Methods

Once the objectives are defined and it is clear how far the research should go, an exhaustive literature review is conducted on hydrogen production methods and their sustainability. Among all the methods, Aspen HYSYS is selected as the best software for simulating the water electrolysis process based on PEM technology. Validation of the data, based on an existing simulation from the literature, is carried out using Python programming language. This process determines if the data is suitable for calculating the sustainability indicators, which are computed using the GREENSCOPE approach and Python as the software tool for their calculation and graphical representation. After completing the previous step, the best approach for the feasibility of the scaling-up process is sought, which is also compared with real data for validation. Finally, the appropriate conclusions are drawn to analyse whether the original objectives have been achieved and to give an overall view of the results obtained in the completion of this work.

1.5 Scope

The scope of this thesis is based on sustainable hydrogen production. Therefore, only the issues related to hydrogen production are analysed. In other words, neither the storage of the product nor the best technologies necessary for its further use are examined. Furthermore, the work focuses on electrolysers based on PEM technology, so the other technologies only serve as a comparative reference and are therefore not analysed in depth. It should also be taken into account that the study is based on simulations to obtain data. Therefore, no real plant was used for experiments and for obtaining real data.

1.6 Scientific contribution

Based on the work done in this thesis, a paper will be presented at the 64th International Conference of the Scandinavian Simulation Society (SIMS):

J. Béjar-Rabascall, G. Mirlekar, Sustainability analysis and simulation of a Polymer Electrolyte Membrane (PEM) electrolyser for green hydrogen production, Submitted in the proceedings of 64th International Conference of Scandinavian Simulation Society, SIMS 2023, Västerås, Sweden.

This paper contributes to making hydrogen a sustainable energy source to meet the growing demand for green energy. It introduces a quantitative assessment of sustainability through the entire Life Cycle Assessment and scales-up a simulation to reduce design and production costs of PEM electrolysers, which is a significant step forward in addressing the main drawback of high production costs.

1.7 Report structure information

The following is an explanation of the content of the chapters through which this master's thesis is structured:

1. Introduction (present chapter): Description of the foundations on which the thesis is based and the definition of the objectives and methodology on which this work is based.

2. Hydrogen fundamentals and production methods: A literature review to understand why hydrogen can be considered one of the vectors of the future and an overview of ways and technologies to produce green hydrogen.
3. Sustainability: A literature review on the principles of chemical sustainability and understanding of the GREENSCOPE methodology. In addition, an overview of the current state of sustainability of green hydrogen production is provided.
4. Process simulation: Description of the methodology used for the simulation of the hydrogen production plant of the PEM electrolyser and validation of the simulation. It also explains the discussion of the results of the different simulations performed.
5. GREENSCOPE sustainability indicators: Explanation of the selected GREENSCOPE indicators and their reference values. Furthermore, the selected indicators are compared with those of a simulation based on an alkaline electrolyser.
6. Process scale-up: Selection of the best method for carrying out the scale-up process and the results achieved by the method used.
7. Conclusions: Evaluation the assessment of the initial objectives and additional conclusions of the thesis. Possible future research is outlined.

2 Hydrogen fundamentals and production methods

This chapter reviews the basics of hydrogen as an energy source for the present and future, including its major consumption sectors and potential applications. It also covers the primary production methods, with an emphasis on blue and green hydrogen. The production of green hydrogen using water electrolysis is also discussed.

2.1 Hydrogen as an energy source

The hydrogen economy is based on the idea of placing hydrogen element as the new energy carrier of the world economy (Pandev *et al.*, 2017). This new paradigm is driven by society's need to replace the current model based on fossil fuels, which is depleting after research confirms that the peak of fossil fuels has been reached in recent years (Bardi, 2019). In addition, many international organisations have recently focused on reducing Greenhouse Gas (GHG) emissions, which cannot be mitigated without a decrease in the use of all fuels that emit pollutants during their combustion (Brandon and Kurban, 2017).

Many researchers have been introduced into this field, giving rise to multiple contributions (Yap and McLellan, 2023). One of the most revolutionary is the one developed by the sociologist Jeremy Rifkin. This research places hydrogen as the enabler of the third industrial revolution. The main reason for believing this is that hydrogen is the most abundant element on earth and therefore, in contrast to fossil fuels, its depletion can be considered infinite. Furthermore, Rifkin is based on the idea of positioning renewable energies as the main sources of energy for the production of electricity, given the instability of these, hydrogen emerges as the main energy source capable of being produced in a sustainable way and at the same time being stored continuously and for a prolonged period (Rifkin, 2003).

Recently, in 2019, the IEA developed a study for the G20 outlining the reasons for the principles needed to start believing in hydrogen as a truly green energy solution (IAE, 2019):

- No other natural resource offers the flexibility of hydrogen as a fuel, hence the report states that the next few years should bring a definite hydrogen boost.
- Its uses as a fuel for conversion into electricity have been extensively tested in various basic sectors of the economy, such as transport, industry and buildings.
- In the coming years, governments and institutions will only have to be willing to invest in technology to combine an energy model that is based on renewable energies and hydrogen production.

However, this transition will not be easy, as the following challenges need to be addressed (IAE, 2019):

- Hydrogen production using non-carbon energy resources is currently expensive.
- Infrastructure development remains sluggish.
- Currently, Hydrogen is produced mostly by natural gas and coal, and legislation restricts the growth of a green hydrogen sector.

At present, the main use of hydrogen is linked to the industrial sector. Petroleum through reforming, ammonia, and methanol production as well as the steel and iron sector are the main consumers of hydrogen produced worldwide. The main problem is that most of this hydrogen is produced by using fossil fuels (Dash, Chakraborty and Elangovan, 2023). Figure 2.1 shows the global consumption of hydrogen, in Millions of tonnes (Mt), by sector during 2021, as well as its percentage (IEA, 2022b).

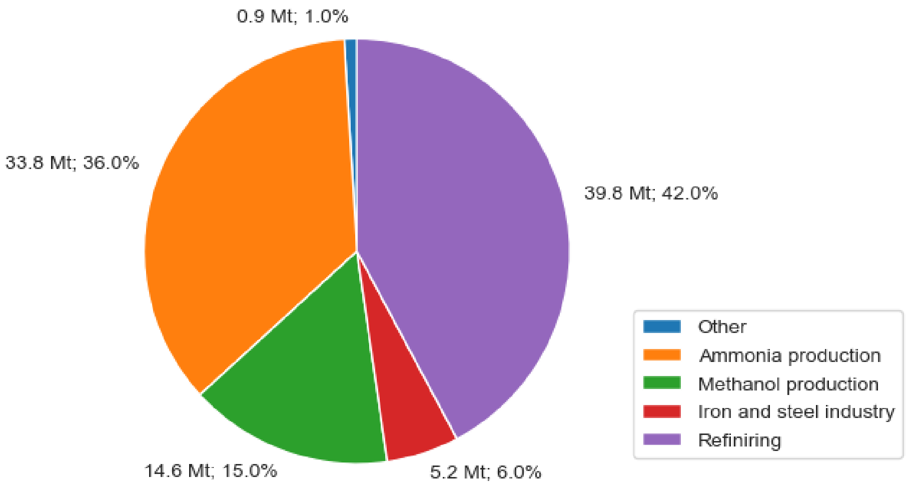


Figure 2.1: Hydrogen consumption by sector in 2021. Source: (IEA, 2022b).

The following sectors are some of the main energy consumers, so it is convenient to know their needs in order to achieve an energy transition based on hydrogen (Brandon and Kurban, 2017):

- Transport: The availability of refuelling stations and the cost of fuel cells will determine the potential use of hydrogen as a fuel in several means of transportation. Furthermore, it is challenging to develop low-carbon fuels for industries like aircraft and shipping, which raises the chance that hydrogen may replace fossil fuels in these fields.
- Buildings: In this sector, hydrogen could be incorporated into existing natural gas networks for residential and commercial buildings. Longer-term prospects could also include the direct use of hydrogen in fuel cells or hydrogen boilers.
- Electricity generation: The future of this sector is expected to lie in the use of renewable sources for electricity generation. Because hydrogen can be easily stored for long periods, it can be used as an alternative for electricity production when renewable sources are not available.
- Industrial: Hydrogen will continue to be used as an industrial feedstock, but the aim is to produce it more sustainably in order to reduce the life-cycle environmental impact of final products.

2.2 Hydrogen production methods

As the focus of this thesis is on hydrogen production, a more in-depth exploration of the different existing methods is deemed appropriate. The classification of hydrogen production, Figure 2.2, methods are accepted by the entire research community and are divided into these different types according to the energy sources used for its production (Bartlett and Krupnick, 2020):

- Grey hydrogen: Due to the low costs, it is the most widespread production type. It is estimated that 47 % of global hydrogen production in 2021 was by this method (IRENA, 2021). It is based on the use of fossil fuel reforming processes, mainly natural gas and coal. The trend is that in the coming years the production of hydrogen by this process, which emits high quantities of CO_2 as well as other GHG pollutants, will be reduced.
- Brown hydrogen: Gasification is the method of producing hydrogen from coal. The main drawback of this brown hydrogen compared to grey hydrogen is that it produces more CO_2 and various pollutants.
- Blue hydrogen: It is based on the use of Carbon Capture and Storage (CCS) as a process to reduce the large amount of CO_2 emitted during the production process of the two previous methods.
- Green hydrogen: The main objective of this method is the zero emission of CO_2 during the production of hydrogen. Water electrolysis powered by renewal is the most prominent process, see Chapter 2.3.

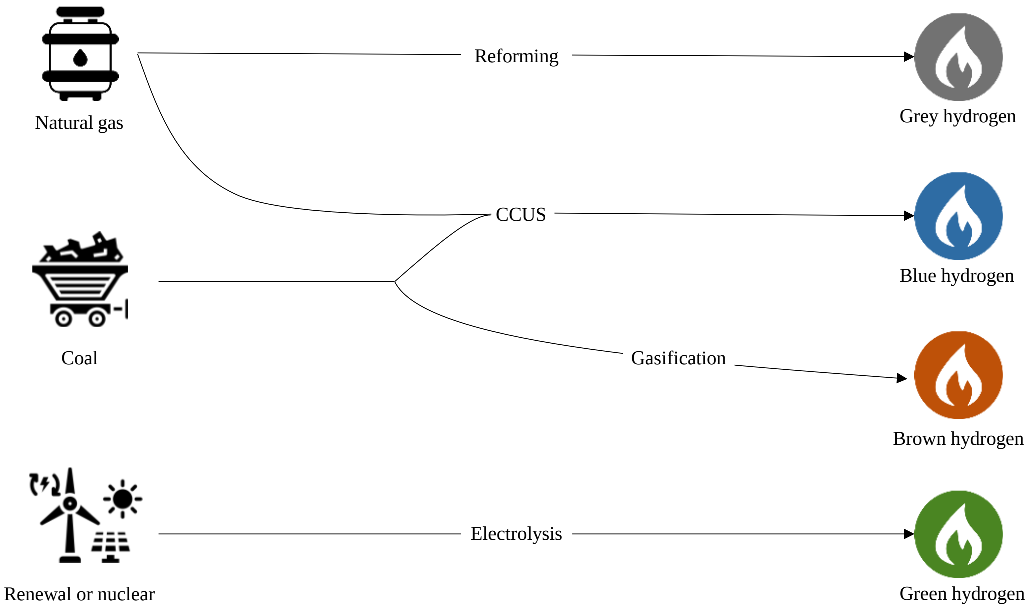


Figure 2.2: Hydrogen production methods. Source: (Bartlett and Krupnick, 2020).

Both blue and green hydrogen are considered essential for achieving sustainable hydrogen production in the future (Bartlett and Krupnick, 2020). Thus, it is appropriate to provide a more comprehensive explanation of the technologies behind them and their projected implementation.

The concept behind **blue hydrogen** is that the present methods for making hydrogen from fossil fuels, Steam Methane Reforming (SMR) or Auto-Thermal Reforming (ATR), might be combined with CCS technology to significantly reduce their carbon footprint. However, despite its promising future as a hydrogen production method, blue hydrogen currently has some drawbacks related to cost and CO_2 emissions throughout its lifecycle. This is linked to the fact that a specific infrastructure is needed for CO_2 capture and storage, which will significantly increase both the investment and operating costs of the plant. Furthermore, CO_2 emissions and dependence on fossil fuels are not mitigated by this type of hydrogen, since the extraction of fossil fuels continues to use polluting processes and the balance is therefore still negative (Howarth and Jacobson, 2021; Noussan *et al.*, 2021).

In contrast to the drawbacks of blue hydrogen, the production of **green hydrogen** offers a cleaner and more sustainable solution to reduce society's dependence on fossil fuels. Utilizing renewable sources of electricity to power the electrolysis of purified water, this process creates hydrogen without producing harmful emissions (Noussan *et al.*, 2021). In this sense, organizations such as the European Union has included green hydrogen in their plans to achieve zero emissions in the Eurozone, through an ambitious plan to boost hydrogen with a target of producing 10 Mt of hydrogen from renewable sources by 2030 (Noussan *et al.*, 2021). Figure 2.3 shows the possible global potential for blue and green hydrogen production in the coming years, which is expected to reach about 400 Mt in 2050 (KPMG Belgium, 2022).

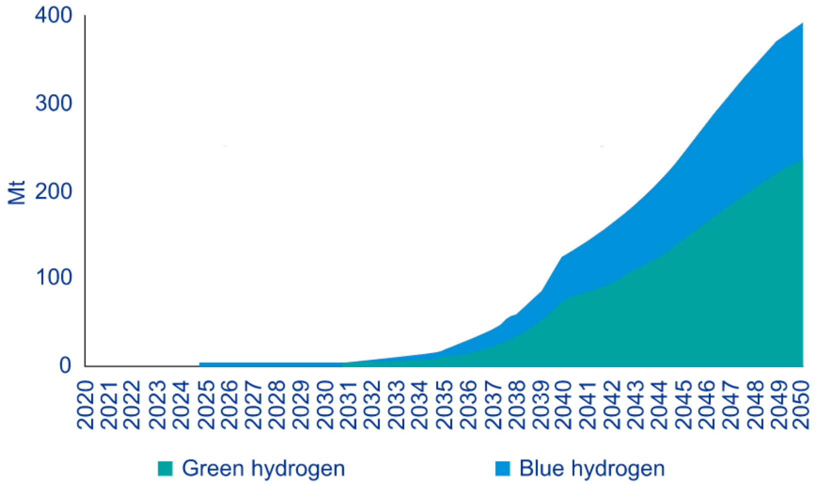


Figure 2.3: Future production of hydrogen. Source: (KPMG Belgium, 2022).

However, as with any breakthrough, not everything is positive from the outset. Unless the following challenges are resolved, the deployment of green and blue hydrogen will not bring about the energy revolution that has been predicted (Clark and Rifkin, 2006; Noussan *et al.*, 2021):

- Large cost: Energy from renewable sources, which is the core of green hydrogen generation through electrolysis, is more expensive to generate than fossil fuels. For this reason, grey and brown hydrogen are still the predominant sources of hydrogen production.
- High power consumption: Related to the previous point, the generation of hydrogen by electrolysis currently requires a high energy demand. As a result, operating costs are higher than those of hydrogen production facilities using natural gas, estimated to be around three times higher.
- Security inversion: The physical characteristics of hydrogen make it a highly flammable substance. For this reason, the safety measures required in hydrogen installations are highly restrictive regarding the manipulation and utilization of this substance.

2.3 Water electrolysis

As mentioned throughout this chapter, water electrolysis is emerging as the key driver of green hydrogen success. This technology is based on the use of renewable electricity to extract hydrogen from water without generating CO_2 emissions and any other pollutant substance, only water as a by-product. For this, an electrolyser is needed, which is the device that through the application of a direct electric current, typically between 1,4 and 2,0 V, can generate two chemical reactions: one at the cathode that produces hydrogen and the other at the anode that generates oxygen. Equations (2.1), (2.2) and (2.3) show the reactions described above (Godula-Jopek and Stolten, 2015).

Global electrolysis reaction:

$$H_2O(l) \rightarrow H_2(g) + \frac{1}{2}O_2(g) \tag{2.1}$$

Cathode reaction:

$$2H^+ + 2e^- \rightarrow H_2(g) \tag{2.2}$$

Anode reaction:

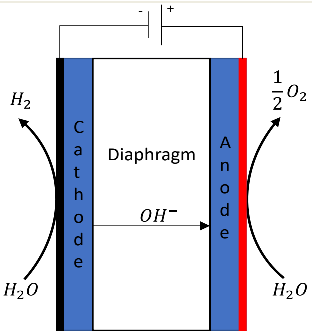
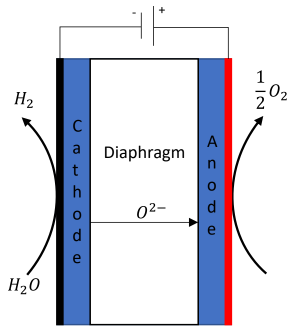
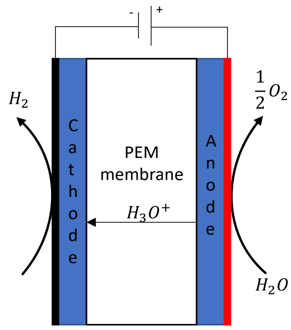
$$H_2O(l) \rightarrow \frac{1}{2}O_2(g) + 2H^+ + 2e^- \tag{2.3}$$

Depending on the operating conditions and the type of electrolyte, the conductive substance that enables the reactions, three different technologies can be used for the production of hydrogen from water electrolysis (Wang, Cao and Jiao, 2022):

- Alkaline Water Electrolysis (AWE): Known as the most developed technology, this method uses as an electrolyte a liquid alkaline solution, typically with KOH and NaOH, with a concentration of around 25 %. AWE has a lower adaptability to fluctuations and it is therefore not a good technology for the use of renewable energies. In addition, its lower current density, around 0,3 A/Cm2 does not make it as competitive as other methods.
- Solid Oxide Electrolysis (SOE): Yttria-stabilized zirconium oxide is the electrolyte utilised in this procedure. The high temperatures, 800 ºC, at which this process works is a major problem in terms of the durability of the catalytic converters, so the operating costs soar. In addition, the hydrogen produced must be treated for purification, adding a procedure to the whole system.
- Polymer Electrolyte Membrane (PEM): This technology utilizes a proton exchange membrane as its electrolyte, which is widely recognized as one of the most prominent technologies in this field. The reasons for choosing this technology for the development of the practical study of this thesis are explained in the following paragraph.

Table 2.1 provides a detailed analysis of the operational specifications of the water electrolysis technologies discussed above, allowing for a comprehensive evaluation of their strengths and limitations. The data indicate that PEM electrolysis is the most advantageous option, as it exhibits superior current density, energy efficiency, and operating pressure. Additionally, its seamless adaptability to electrical power changes makes it a highly desirable technology, allowing for efficient integration with renewable energy sources (Hancke, Holm and Ulleberg, 2022). As a result, PEM electrolysis has been selected as the technology for the process simulation, sustainability analysis, and process scale-up in this thesis, chapters 4, 5 and 6 of this thesis, respectively.

Table 2.1: Electrolysis technologies. Source: (Wang, Cao and Jiao, 2022).

	AWE	SOE	PEM
Cathode/ Anode	$4H_2O+4e^- \rightarrow 4OH^-+2H_2$/ $4OH^- \rightarrow 2H_2O+O_2+4e^-$	$2H_2O+4e^- \rightarrow 2O^{2-}+2H_2$/ $2O^{2-} \rightarrow 4e^-+O_2$	$4H^++4e^- \rightarrow 2H_2$/ $2H_2O \rightarrow 4H^++4e^-+O_2$
Current density	0,2 – 0,4 A/cm^2	0,3 – 0,5 A/cm^2	1 – 3 A/cm^2
Cost	1000-1500 €/kW	Under development	1500-2000 €/kW
Efficiency	62 – 82 %	81 – 92 %	67 – 82 %
Operating Temperature	≤ 90 ºC	800 ºC	≤ 80 ºC
Operating Pressure	<30 bar	1-5 bar	<60 bar
Energy demand	4,5 – 5,5 kWh/Nm^3	< 3,5 kWh/Nm^3	4,0 – 5,0 kWh/Nm^3
Response time	Minutes	-	Seconds
Durability	60.000 h	<20.000 h	80.000 h
Maturity	High	Lab scale	First applications
Schematic			

3 Sustainability

Firstly, this chapter reviews the difference between the concepts of green chemistry and chemical sustainability, on which the GREENSCOPE methodology described above is based. At the end of the chapter, based on a literature review, an analysis of the sustainability of green hydrogen is carried out. This analysis is divided according to the GREENSCOPE methodology.

3.1 Chemical sustainability

Over the years, the chemical industry has grown to become one of the basic industries for human development (Anastas, 2007). In 2018, global sales reached 3,347 billion dollars, a growth of 68% compared to 2008 (CEFIC, 2021). However, while the quality of life and production benefits of these products are widely demonstrated, it should be noted that they are also subject to environmental and human health costs that must be taken into account. For example, in the United States (US) 549,65 million pounds of waste were released into the atmosphere by the chemical sector, which are related to carcinogenic and toxic substances, emissions of gases and solid waste, among other pollutants produced (US EPA, 2018).

Although often used interchangeably, green chemistry and chemical sustainability are not exactly the same concepts. To reduce the impact of the chemical industry on the environment, the concept of green chemistry was developed by Anasts and Warner in 1998. This concept is founded on the principle of designing and utilizing chemicals with minimal pollution potential (Anastas and Warner, 1998). On the other hand, sustainable chemistry is based on the principles of green chemistry and goes further to consider not only the chemical reaction but also the environmental, social, and economic impact of the entire product life cycle, including the sourcing of raw materials, production, use, and disposal (García-Serna, Pérez-Barrigón and Cocero, 2007).

Sustainable chemistry tries to provide a comprehensive and long-term solution to the environmental challenges of traditional chemical processes. This involves evaluating the entire system and identifying ways to reduce the environmental impact of chemical processes while simultaneously promoting economic development and social well-being. Through the use of this approach, sustainable chemistry aims to create and utilize chemicals that are not only environmentally friendly but also socially responsible (Kirchhoff, 2005).

3.2 GREENSCOPE methodology

Using the approach of chemical sustainability and aiming to measure sustainability in any new or existing chemical process throughout its LCA, the EPA created the GREENSCOPE tool (US EPA, 2015). The methodology of this tool is based on a set of metrics, GREENSCOPE indicators, used to evaluate the environmental performance of chemical products and processes in four different principal areas: Environmental, Efficiency, Economic and Energy. These indicators provide a comprehensive framework for assessing the sustainability and environmental impact of chemicals, from raw material sourcing to disposal (Gonzalez and Smith, 2003).

To generate the GREENSCOPE indicators, Equation (3.1) is used, which compares the actual process scenario with the best-case scenario of 100% sustainability and the worst-case scenario of 0% sustainability. The GREENSCOPE tables provide a comprehensive set of indicators and their corresponding parameters for calculating both the best and worst-case scenarios (Ruiz-Mercado, Smith and Gonzalez, 2012; Ruiz-Mercado, Gonzalez and Smith, 2014). The great advantage of this approach is that it allows the comparison of the sustainability of similar chemical processes (Li *et al.*, 2016).

$$Sustainability\ score = \frac{Actual - Worst}{Best - Worst}\ x\ 100\ [\%] \qquad (3.1)$$

The taxonomy of GREENSCOPE indicators is based on the four main areas in which the methodology aims to assess sustainability goals. The following is a brief overview of the categories and their main characteristics (Ruiz-Mercado, Gonzalez and Smith, 2014):

- Environmental (66 indicators): Taking into account the philosophy of Green Chemistry, this group of metrics tries to quantify the impact of the chemical materials used in the process. This assessment is carried out for the raw material and also for the product and the materials released at the end of the process.
- Efficiency (26 indicators): Relation between the input and the output of the process product, energy, feedstocks, pollutants, cost and other metrics important during the LCA.
- Economic (33 indicators): A green transition is certainly a good goal, but without economic prosperity, a new technology cannot take off. Therefore, a variety of indicators are proposed to assess whether the investments, benefits and costs are sufficiently beneficial for a new process or technology to establish itself in the markets.
- Energy (14 indicators): Improved reaction chemistry can lead to a reduction in energy consumption, resulting in various benefits. Thus, with the goal to minimise energy consumption, a set of metrics are proposed to include in the whole LCA.

This GREENSCOPE methodology and some of its indicators are used in Chapter 5 of this thesis to quantify the sustainability of the process simulation, described in Chapter 4, of a hydrogen production plant based on PEM technology, as well as for comparison with another hydrogen production method.

3.3 Green hydrogen sustainability

By reviewing the existing literature, the following analysis is conducted to assess the current status of green hydrogen in terms of its environmental impact, efficiency, economic viability and energy efficiency, which are the same areas of assessment of the GREENSCOPE methodology.

Environmental

Green hydrogen is considered an environmentally carbon-neutral energy carrier. Its contribution to climate change is minimal, as the only by-product of its production is water and no GHG are emitted during the whole LCA. However, the environmental impact of hydrogen is not zero, the type of renewable energy source used, the origin of the water for the electrolysis process and the residues generated after the usage of the production equipment must be taken into account (Baykara, 2018).

Efficiency

The efficiency of a PEM electrolyser varies depending on the quality of the materials used, the design of the electrolyser, the operating temperature, the pressure, and the concentration of the electrolytes. In general, the typical efficiency of a PEM electrolyser can range from 67% to 82%. To improve efficiency, efforts should focus on optimising the geometry of the electrolytic cell, using more efficient catalysts and optimising the operating conditions of the electrolyser (Wang, Cao and Jiao, 2022).

Economic

The current lack of extensive green hydrogen production is mainly due to poor economic competitiveness, which is why most production is done using fossil fuels. The main costs of green hydrogen production are related to the cost of renewable electricity, the efficiency of the electrolysis process and the cost of the electrolysis equipment (Yue *et al.*, 2021).

Currently, as it is shown in Figure 3.1, the cost of producing green hydrogen is estimated to be between 2,50 - 6,50 $/kg, depending on the production method and location. In comparison, the cost of grey hydrogen is around 1 - 2,5 $/kg. Nevertheless, the cost of green hydrogen is expected to decrease as renewable energy becomes more affordable and the efficiency of the electrolysis process improves. By 2050, the cost of green hydrogen is expected to be approximately the same as that of grey hydrogen at present (KPMG Belgium, 2022).

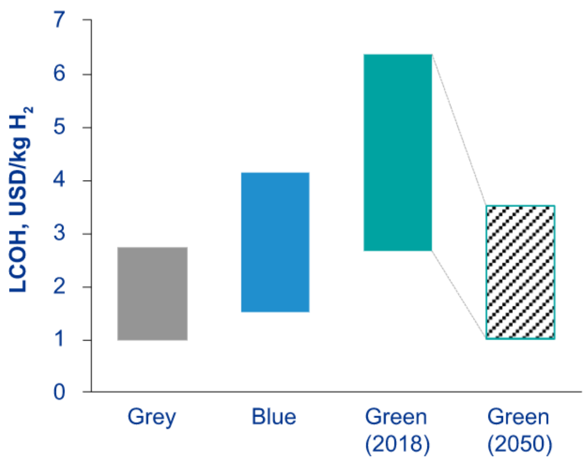


Figure 3.1: Hydrogen production costs. Extracted from: (KPMG Belgium, 2022).

Energy

In this area, GREENSCOPE methodology is focused on the concepts of the amount of energy required and the use of renewable energies. In the case of green hydrogen, it is estimated that the production of 1 kg requires 50-55 kWh of electricity, which is considered high energy consumption compared to some fossil fuels (Kurrer, 2020). This consumption depends on the efficiency of the electrolysis process and the energy source used to power it (Antweiler, 2020). As it is explained in Chapter 2.2, green hydrogen is expected to be a technology powered by renewable energies, so the energy renewal consumption value is potentially 100 %.

4 Process simulation

This chapter focuses on the process simulation for a PEM electrolyser. The methodology utilized is described, followed by an explanation and validation of the model utilizing existing data from the literature. The results are then discussed in a dedicated section.

4.1 Methodology

The methodology employed in this thesis for process simulation consists of the following steps:

1. Conduct a literature review to gain an understanding of the necessary equipment and software required for the simulation.
2. Comprehension of the inputs and outputs of the model used in Aspen HSYS and Aspen Customer Modeler (ACM) for the water electrolyser.
3. Validating the model through a comparison with data from similar simulations obtained from the literature.
4. Computing results to comprehensively understand the factors that affect the performance of the process.
5. Obtaining the data used in Chapter 5 to generate the GREENSCOPES indicators.

4.1.1 Simulated PEM water electrolysis plant

As discussed in Chapter 2.3, the most promising option for future hydrogen production plants is the PEM water electrolysis technology, which is why the chosen simulation plant is based on it.

The electrolyser is the central component of any water electrolysis production process, receiving electricity for working. It consists of two parts: the anode side, responsible for water supply and oxygen production, and the cathode side, which produces hydrogen. Once water is separated into its components, gas separators extract the products, while water is recycled for further use (Carmo *et al.*, 2013). Figure 4.1 illustrates the arrangement of these basic elements, including the inlet and outlet flows of each. Pumps, coolers, and mixers are auxiliary elements required to maintain appropriate temperatures and flow rates but are not considered essential and are not shown in the schematic. Nonetheless, they are used in subsequent simulations.

The simulated production plant is designed to handle electrical power between 0 and 6 kW, as the simulation tends to produce inaccurate results beyond this range. The operational temperature is evaluated at two different values, 40 and 55 ºC, while the operational pressure, in the anode side, is simulated at 7 and 30 bar. These values are selected based on a literature review of typical parameters used in similar simulations (Colbertaldo, Gómez Aláez and Campanari, 2017).

Figure 4.1 also shows the values of the flow rates in the electrolyser. Additionally, it is possible to observe that the operating values of temperature and pressure on the cathode side are set at 45 °C and 2,5 bar for the whole simulation process.

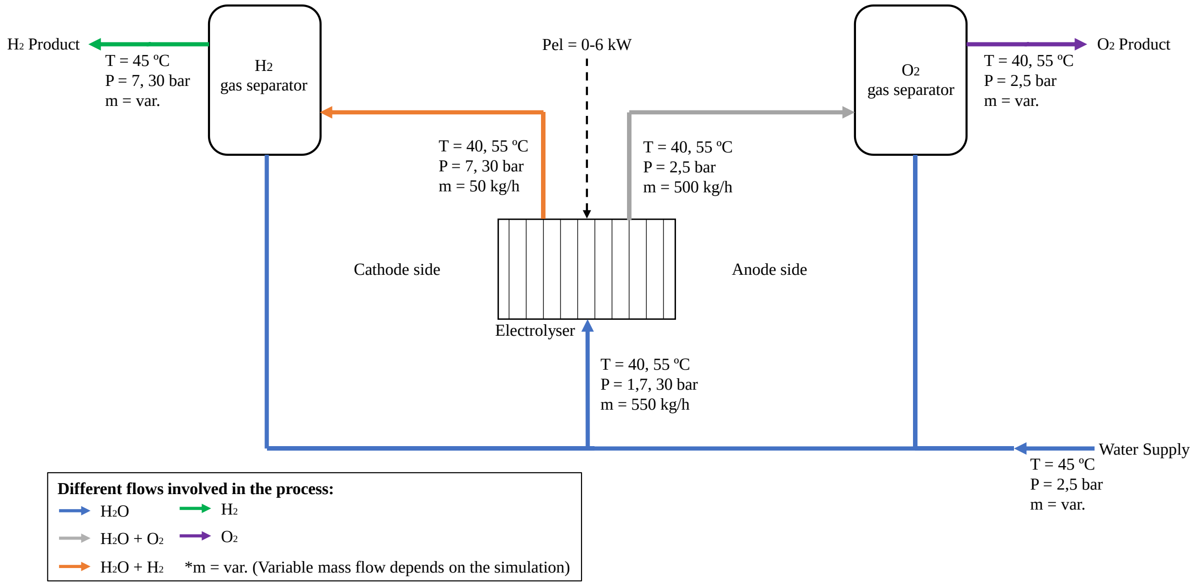


Figure 4.1: Basic schematic of the simulated PEM hydrogen plant.

4.1.2 PEM process simulation

Chemical Process Simulation (CPS) software is a crucial tool in the field of chemical engineering, enabling engineers to build virtual models of chemical processes and test different operating scenarios. Through CPS, engineers can gain a deep understanding of how the process behaves under different conditions, identifying areas for process improvement and optimization to maximize efficiency, minimize waste and reduce costs. With the help of CPS, chemical engineers can make informed decisions on process design and control, ensuring that the process operates optimally and meets the required product specifications (Casavant and Côté, 2004).

The present thesis identifies Aspen HYSYS, developed by AspenTech, as the most suitable software for simulating a PEM water electrolysis hydrogen production plant. This is because Aspen HYSYS incorporates an existing model of this technology, which was created using Aspen Custom Modeler (ACM) and added as a module to Aspen HYSYS. By utilizing this module, the simulation process becomes more efficient and effective, resulting in more accurate predictions of the plant's behaviour and performance (Aspen Technology, 2021).

The schematic diagram shown in Figure 4.2 represents the model used for simulating the different steady states selected for the hydrogen production system. To simplify the explanation the process is divided into two main parts: the anode (A) and the cathode (C). In the electrolyser (ACM), recycled water from the separation process enters through the C-IN and A-IN streams. After electrolysis separates the hydrogen and oxygen, resulting streams containing hydrogen mixed with water (C-OUT) and oxygen mixed with water (A-OUT) are obtained. These streams are then sent to the separators, where two separators, SEP-1 and SEP-2, are needed in the cathode part to extract the maximum amount of hydrogen from the system. However, on the oxygen side, only one separator (SEP-3) is required as it is not necessary to extract all of the oxygen. The water from the separators is recycled and reused in the process, while additional water is added to the anode side to ensure that the inserted mass flow into the electrolyser remains constant (A-MKUP).

Furthermore, to ensure an accurate simulation of the process, it is necessary to incorporate auxiliary elements that enable the solution to converge at all times and for any simulation. These auxiliary elements include pumps, coolers, mixers, stream splitters, recyclers, and an adjusting valve. The two latter are not real elements necessary in a real plant but ensure the proper convergence of all mass flows and maintain the desired operating temperature of the electrolyser.

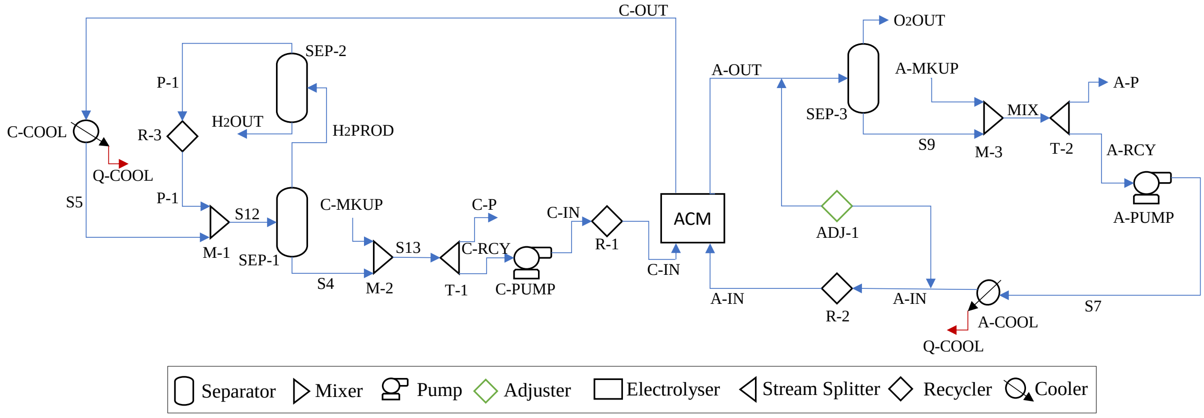


Figure 4.2: Aspen HYSYS schematic for the PEM simulation.

During the simulation, some difficulties were encountered in introducing the inputs properly and ensuring correct operation. For instance, to modify the electrical power supplied, parameters must be adjusted in the ACM module that represents the electrolyser. To adjust the temperature, the desired value must be modified in the ADJ-1 element, and to change the pressure, the flow pressure of C-IN must simply be adjusted.

In addition, it should be noted that the scheme includes an inlet for the water supply, C-MKUP, on the cathode side to balance the water flow in case of convergence problems. However, this flow was not modified during the simulation process, as no inconsistency was found, and the simulation worked correctly at all desired points.

Finally, it is important to point out that the obtained results were transferred to xlsx format files for further data manipulation using Python 3. Although Aspen HYSYS automates the process of transferring simulation software data to xlsx format, the task of importing this data into Python requires specialized knowledge and skills that have been acquired through the master's program. Possessing this knowledge is crucial for ensuring accurate data collection, processing, and analysis.

4.1.3 Limitations and considerations

The limited information available about the model and the inclusion of an ACM module has posed significant difficulties in properly understanding the simulation's operation. This, coupled with the limited experience with Aspen HYSYS simulation software, has required a great deal of effort to precisely modify the system's inputs and extract the necessary data for subsequent processes. Nonetheless, it can be affirmed that a deep understanding of both the ACM software and Aspen HYSYS has ultimately been attained, and successful simulations have been developed as a result.

On the other hand, regarding the model, it was developed for a low-scale hydrogen production plant. This can be seen in the fact that the maximum electrical power is limited to 6 kW before the system fails to converge. As a result, the values for hydrogen produced are very low and cannot be considered for a real process of hydrogen production. To address this issue, a scaling-up process is carried out in chapter 6 of this thesis.

It is worth noting that a hydrogen production plant typically requires additional components beyond those included in this simulation, such as equipment for hydrogen storage and process purification (IRENA, 2020). However, the focus of this process simulation is solely on gathering data related to hydrogen production. Thus, the hydrogen storage and purification equipment of the plant are not considered.

4.2 Simulation model

The simulation relies on three key inputs: the electrical power, the operational temperature and the operational pressure. Its outputs include the production of hydrogen and oxygen, water consumption, and heat losses. These outputs are obtained through the use of an ACM module in Aspen HYSYS. The PEM electrolyser, computed in the ACM model, is based on a voltage model and several equations that ensures the energy and mass balance models inside the electrolyser. This model and these equations are detailed in the present chapter and are extracted from the simulation develop by Colbertaldo.P et al., 2017 (Colbertaldo, Gómez Aláez and Campanari, 2017). For a better understanding, the simulation inputs and outputs are illustrated in Figure 4.3.

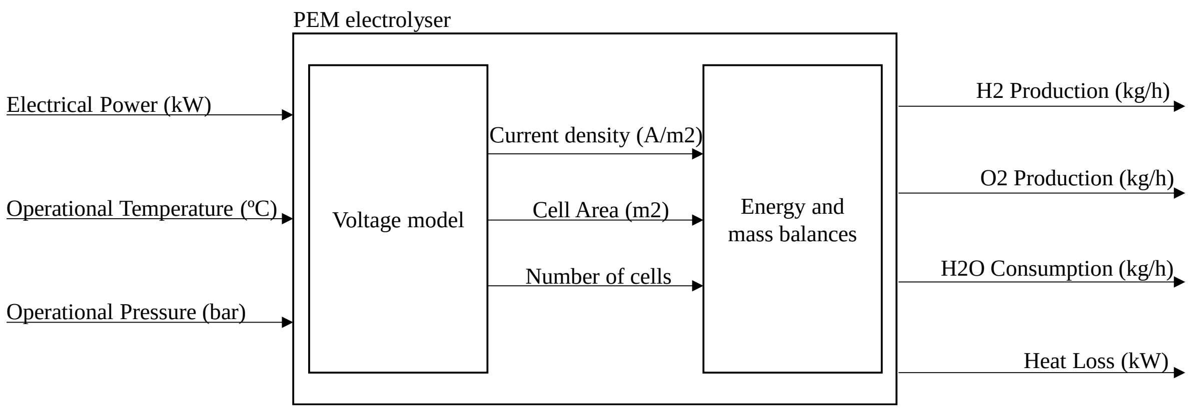


Figure 4.3: Basic schematic of the I/O and model used for the PEM electrolyser.

4.2.1 PEM electrolyser model description

Voltage model

The PEM electrolyser’s voltage model is determined by Equation (4.1), which calculates the total voltage required for a single cell to perform the electrolysis process. This model consists of several components, including the ideal voltage, the minimum voltage required, as well as various overvoltage, different losses, and factors resulting from the activation of the reaction incurred throughout the process (Colbertaldo, Gómez Aláez and Campanari, 2017; Aspen Technology, 2021).

$$V_{cell} = V_{id.} + \Delta V_{act.} + \Delta V_{ohm.} + \Delta V_{diff.} + \Delta V_{par.} \quad (4.1)$$

Where $V_{id.}$ is the ideal voltage, $\Delta V_{act.}$ is the activation voltage, $\Delta V_{ohm.}$ is the ohmic voltage, $\Delta V_{diff.}$ is the diffusion voltage and $\Delta V_{par.}$ is the parasitic voltage. The equations required to calculate the individual voltages are explained below.

The minimum voltage required to initiate an electrolysis process in a cell is known as the ideal voltage (Vid.), as described by Equation (4.2). The ideal voltage is closely linked to the Gibbs energy, which represents the minimum energy needed for the process to occur, as well as a pressure correction term.

$$V_{id.} = \frac{1}{nF}(\Delta G + RT_{op.}\ln(\frac{p_{H_2} + p_{O_2}^{0.5}}{a_{H_2O}})) \quad (4.2)$$

Where n is the number of electrons, F is the Faraday's constant, ΔG is the Gibbs free energy value, R is the gas constant, $T_{op.}$ is the operational temperature in the cell, p is the partial pressure for both elements H_2 and O_2 and a_{H_2O} is the water activity value.

For the reaction to take place, an activation voltage ($\Delta V_{act.}$), Equation (4.3), is required, which is based on the Tafel equation and incorporates Butler-Volmer's simplification (García-Valverde, Espinosa and Urbina, 2012).

$$\Delta V_{act.} = \Delta V_{act,cat.} + \Delta V_{act,an.} \quad (4.3)$$

Where $\Delta V_{act,cat.}$ is the activation voltage in the cathode side and $\Delta V_{act,an.}$ is the anode side voltage activation. These activation voltages have the same equation on both sides, which is described in Equation (4.4).

$$\Delta V_{act.,x} = \frac{RT_{op.}}{\alpha_x nF}\ln(\frac{i_u}{i_{0,x}}) \quad (4.4)$$

Where x represents the anode or the cathode, R is the gas constant, $T_{op.}$ is the operational temperature in the cell, α_x is the charger transfer coefficient, n is the number of electrons, F is the Faraday's constant, i_u is the useful current density and $i_{0,x}$ is the exchange current density.

According to Ohm's law, the electrical losses ($\Delta V_{ohm.}$) occurring in the anode, the cathode and the membrane during the electrolysis process are represented by Equation (4.5).

$$\Delta V_{Ohm.} = (R_{cat.} + R_{an.} + R_{mem.})\, i_u A_{cell} \quad (4.5)$$

Where $R_{cat.}$ is the cathode side resistance and is calculated using Equation (4.6), $R_{an.}$ is the anode side resistance and is calculated using Equation (4.6), $R_{mem.}$ is the membrane resistance and is calculated using Equation (4.7), i_u is the useful current density and A_{cell} is the active cell area.

$$R_x = \frac{t_x \rho_x}{A_x} \quad (4.6)$$

Where x represents the anode or the cathode, t_x is the electrode thickness, ρ_x is the resistivity and A_x is the active electrode area.

$$R_{mem.} = \frac{t_{mem}}{\sigma_{mem.} A_{mem.}} \quad (4.7)$$

Where $t_{mem.}$ is the membrane thickness, $\sigma_{mem.}$ is the conductivity based on the Springer model (Springer, Zawodzinski and Gottesfeld, 1991) and $A_{mem.}$ is the active membrane area.

Diffusion voltage ($\Delta V_{diff.}$), Equation (4.8), represents the diffusion losses that occur when mass transport is hindered by the concentration gradient between the membrane surface and the main stream in which the reaction takes place. These losses are the result of mass transport limitations due to the concentration gradient.

$$\Delta V_{diff.,x} = \frac{RT_{op.}}{\alpha_x nF} \ln \left(\frac{i_L}{i_L - i_u}\right) \tag{4.8}$$

Where x represents the anode or the cathode, R is the gas constant, $T_{op.}$ is the operational temperature in the cell, α_x is the charger transfer coefficient, n is the number of electrons, F is the Faraday's constant, i_u is the useful current density and i_L is the limiting current density.

Parasitic losses (ΔVpar.), are typically expressed as a change in current rather than an increase in voltage. Essentially, the current efficiency is determined by the ratio of the input current to the useful current, Equation (4.9). This ratio is evaluated using the Faraday efficiency, which in the case of a PEM system, it is common to be close to 100%. Consequently, the Faraday efficiency used in the simulations is 99%.

$$\eta_{far.} = \frac{Iu}{Istack} \tag{4.9}$$

Where I_u is the useful current which is calculated by multiplying the current density (i_u) by the active area of the cell (A_{cell}) and I_{stack} is the current in the cell.

Mass balance

The material balance evaluation in the electrolysis process is divided between the anode and cathode sides, and it is based on the assessment of the various flows involved. These flows include the water flow input, hydrogen production as described by Equation (4.10), oxygen production, electro-osmotic, diffusivity losses as described by Equations (4.11) and (4.12), respectively, and the pressure flow compensation as described by Equation (4.13).

$$\dot{N}_{H_2} = \frac{i_u A_{cell} N_{cells}}{nF} \tag{4.10}$$

Where i_u is the useful current density, A_{cell} is the active cell area, N_{cells} is the number of cells in the stack, n is the number of electrons and F is Faraday's constant.

$$\dot{N}_{H_2O}^{e-o} = \frac{n_d i_u A_{cell} N_{cells}}{F} \tag{4.11}$$

Where n_d is the coefficient related to the humidification of the membrane extracted from (Colbertaldo, Gómez Aláez and Campanari, 2017), i_u is the useful current density, A_{cell} is the active cell area, N_{cells} is the number of cells in the stack and F is the Faraday's constant.

$$\dot{N}_{H_2O}^{Diff.} = \frac{D_{H_2O}^{eff.} \Delta C A_{cell} N_{cells}}{t_{mem}} \tag{4.12}$$

Where $D_{H_2O}^{eff.}$ is the diffusivity function based in (Aspen Technology, 2021), ΔC is the comparison water composition in the anode and cathode side, A_{cell} is the active cell area, N_{cells} is the number of cells in the stack and $t_{mem.}$ is the membrane thickness.

$$\dot{N}_{H_2O} = -\frac{K_{Darcy} A_{cell} \rho_{H_2O} (P_{cat.} - P_{an.})}{\mu_{H_2O}} \tag{4.13}$$

Where K_{Darcy} is the membrane permeability, A_{cell} is the active cell area, ρ_{H2O} is the water density, $P_{cat.}$ and $P_{an.}$ are the pressure value in the cathode and anode sides respectively and μ_{H2O} is the water viscosity.

Energy Balance

The energy balance is determined by comparing the energy inputs and outputs of the system equal to the total energy capacity. The inputs include the electrical power and the energy content of the inlet water flow, while the outputs encompass the heat losses (as described by Equation (4.14) as well as the outflow energy from both the anode and cathode sides.

$$Q_{loss} = h_{free} A_{ext} (T_{op.} - 10 - T_{std.}) \tag{4.14}$$

Where h_{free} is the heat transfer coefficient based in (Aspen Technology, 2021), $A_{ext.}$ is the exterior area (Aspen Technology, 2021), $T_{op.}$ is the operational temperature in the cell and $T_{std.}$ is the standard temperature.

Table 4.1 contains constant values used in the model, which must be set as fixed parameters in the Aspen Customer Modeler code. These values have been extracted from the model provided by AspenTech (Aspen Technology, 2021).

Table 4.1: Simulation fixed parameters. Source: (Aspen Technology, 2021).

Parameter	Value
Anode current density ($i_{a,0}$)	$1 \cdot 10^{-10}$ A/cm^2
Cathode current density ($i_{c,0}$)	$1 \cdot 10^{-3}$ A/cm^2
Charger transfer coefficient (α_x)	0,5
Active cell area (A_{cell})	160 cm^2
Number of cells (N_{cells})	12
Resistivity (ρ_x)	7,5 mΩ cm
Cathode and anode thickness (t_x)	1,3 mm
Membrane thickness (t_{mem})	127 µm
Number of electrons (n)	4

4.2.2 Process validation

The process validation consists of comparing the simulated data with the model presented in (Colbertaldo, Gómez Aláez and Campanari, 2017). A comparison of the data is carried out by simulating the system at an operating temperature of 55 ºC and operating pressure of 30 bar. These operating conditions are chosen because they represented the maximum pressure that could be used without causing the simulation to fail and because they were consistent with the temperature used in one of the simulations presented in the reference article.

In general, it can be verified that the figures obtained through simulation are similar in shape and magnitude to those obtained in the reference article. However, any observed differences can be attributed to the use of a different model in the simulation presented in this thesis. This model incorporates certain equations suggested by AspenTech, which explain the discrepancies observed (Aspen Technology, 2021). Below are explained the reference figures used for the process validation.

Figure 4.4 illustrates the polarization curve, which demonstrates the relationship between the voltage cell and the current density. Furthermore, it provides insight into how the various voltages incorporated in the model change as the current density increases. As can be observed increasing the current density results in increased over voltages. This is reflected in the fact that resistive, diffusive, and activation voltages are all increased, leading to a higher overall voltage requirement for the plant at higher current densities, which also implies higher power.

Unfortunately, the simulated plant does not achieve current density values as high as those presented in the comparative data article. Therefore, the specific comparison is made at a current density of 1.3 A/cm2, where the voltage value for the simulated plant in this thesis is known to be 2.27 V. For the same data point in the reference article, the voltage is observed to fall between the values of 2.2 and 2.3 V. Thus, it can be concluded that there is a correspondence between both figures, validating the voltage model.

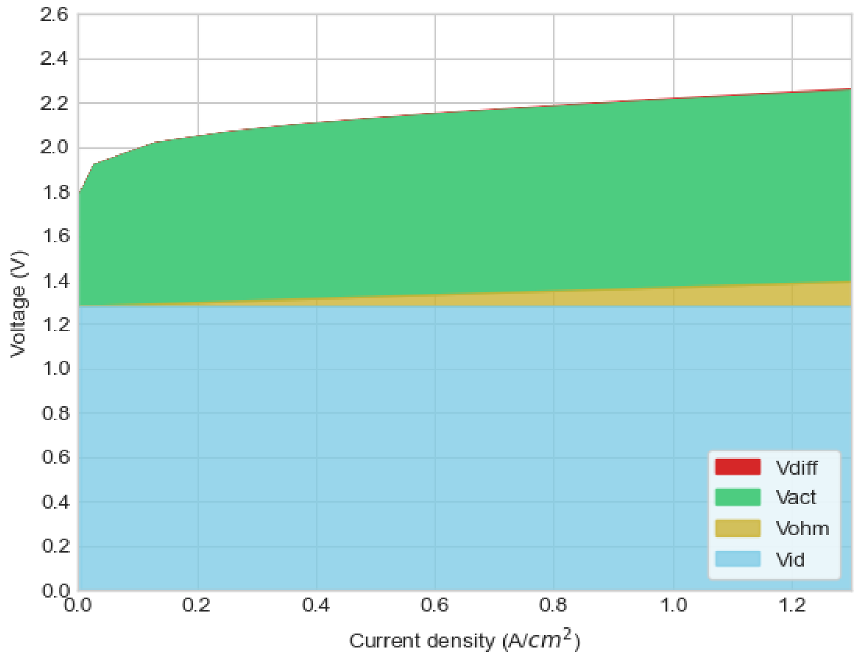


Figure 4.4: Polarization curve and influence of the different voltages.

The linearity of the relationship between the electric power supplied to the electrolyser and the amount of hydrogen produced is demonstrated in Figure 4.5. This indicates that the more electric power that is supplied to the system, the more hydrogen is produced (Colbertaldo, Gómez Aláez and Campanari, 2017). The figure depicts a power range of 0 to 6 kW, which is considered low for a real electrolyser, and hence, the simulation yields low hydrogen production values. However, it is important to note that the reference article does not provide data on higher electric power or hydrogen production. Therefore, the reference article can be utilized for data comparison.

The simulated data can confirm that the hydrogen production electrolyser for all simulated powers corresponds to the values reported in the reference article. The linearity and data coincidence can be demonstrated and allow a proper validation of the simulation. This fact is particularly evident for an electric power of 2 kW, which yields a production of 0.4 Nm3/h, matching the values in the reference data.

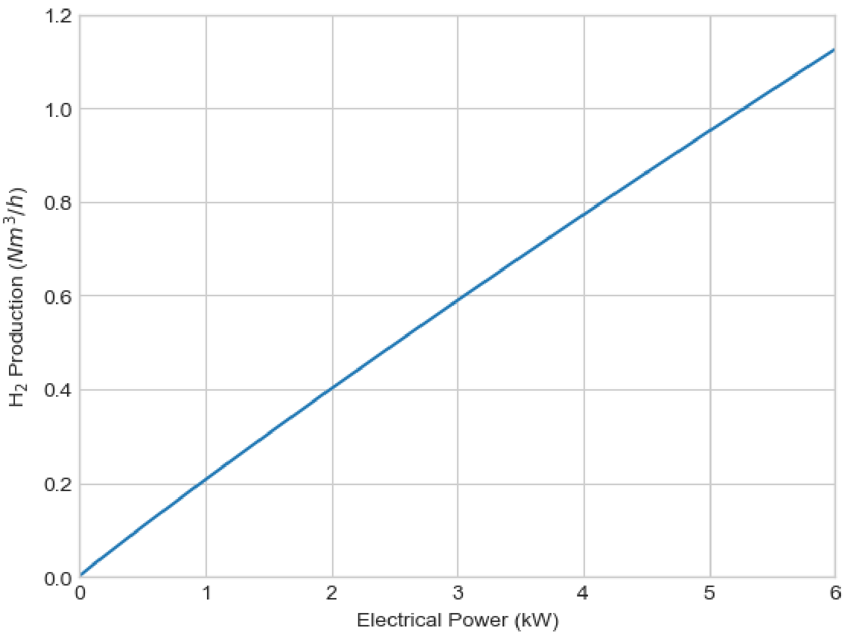


Figure 4.5: Hydrogen production rate regarding the electrical power input.

By presenting the above figures, the validity of the model can be demonstrated, and therefore it can be determined that the data obtained can be used for subsequent processes of model analysis for different operating points, computation of GREENSCOPE sustainability indicators, as well as for the scaling-up process.

4.3 Simulation results

The successful functionality of the simulated plant utilizing Aspen HYSYS is confirmed. As a result, the plant can be simulated at various operational points to examine the impacts of input variables on hydrogen production and overall plant performance. Table 4.2 displays the simulated operating points that allow further conclusions to be drawn on the performance of the plant.

Table 4.2: Operational points for simulations.

Colour coding	Temperature (ºC)	Pressure (bar)
Green	55	7
Purple	40	7
Blue	55	30

The procedure for the simulations is as follows:

1. Set the operational temperature and pressure.

    1.1. To set the pressure, it is necessary to change the value on the cathode side by modifying the parameter in the different modules.
    1.2. To set the temperature, the value of the target temperature needs to be changed in the ADJ-1 element module. This causes the operating temperature in the PEM ACM module to change to the desired temperature.
2. After setting the operational points, it is necessary to vary the electrical power input between 6 kW, maximum power input, and 1 kW, where the simulation starts to behave incorrectly.
3. Data needed for each operational point and power input is collected in Excel files.
4. Finally, Python 3 is used to analyse the data and extract conclusions from the simulation.

Figure 4.6 displays the production of hydrogen and oxygen at various operating points and different electrical powers. The solid lines represent hydrogen production, while the dashed lines denote oxygen production. The operating points are identified by colours, following the previously established code in Table 4.2.

Analysis of Figure 4.6 reveals that higher electricity input leads to increased production of hydrogen and oxygen at all operating points. However, the level of hydrogen production varies among the operating points due to the influence of temperature and pressure on the simulations. Specifically, temperature has a direct proportional effect, with higher temperatures resulting in higher hydrogen production. Conversely, pressure has an inversely proportional effect, where higher operating pressures lead to lower levels of hydrogen production.

This fact underscores the importance of finding a balance to achieve the optimal operating point. It is not only about finding the lowest operating pressure at the highest operating temperature but also about taking into account other crucial parameters. For instance, it is essential to consider that hydrogen produced must be stored at the highest possible pressure after the separation stage. Thus, the minimum pressure may not necessarily be the best operating point, as the energy requirements and costs for hydrogen compression in subsequent stages need to be considered. This highlights that operating at high pressures is one of the key advantages of PEM technology (Gutierrez, 2021). Therefore, reaching the optimal operating point requires careful balancing throughout the entire process, beyond merely maximising the amount of hydrogen extracted from the electrolysis process.

In Figure 4.6, is possible to observe oxygen production. For all operating points, the extracted oxygen remains constant. This is attributed to the separation capacity of the module used in the simulation. As a result, the level of oxygen produced could potentially be higher if not for the limitations of the oxygen production element. However, since the plant's primary objective is hydrogen production, a high level of oxygen production is not desirable as it would imply additional energy and economic costs.

The conclusions derived from this discussion of results are consistent with the findings discussed in other articles related to hydrogen simulation and production. Thus, it can be reaffirmed that the simulation of various operating points conducted in this study is valid and coherent for extracting results and drawing conclusions (Colbertaldo, Gómez Aláez and Campanari, 2017; Buttler and Spliethoff, 2018).

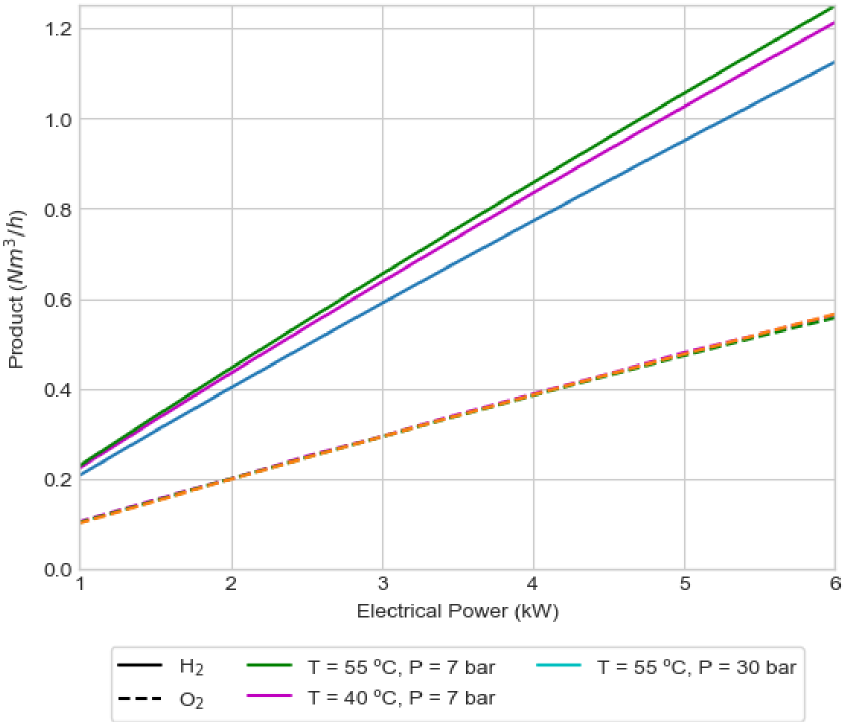


Figure 4.6: Hydrogen and oxygen production relative to electrical power input.

Presented below is Figure 4.7, which provides the correlation between voltage cell and current density, as well as the relationship between current density and efficiency. Where efficiency is defined as the ratio of energy extracted from the process in the form of hydrogen, using its LHV, to the amount of electrical energy input to the process. Similar to the previous figure, colour codes defined in Table 4.2 are employed to identify the operational point of the simulation. Solid lines are used to represent voltage cell, while dashed lines indicate the evolution of efficiency.

For the various simulated points, the voltage of the cell is different for the same value of current densities. This is directly correlated with the amount of hydrogen produced. In other words, when less product is extracted, higher losses occur, resulting in a higher voltage for the cell. This relationship is evident when comparing Figure 4.6 and Figure 4.7. For example, the green operating point, which corresponds to the highest amount of product extracted, exhibits lower values of cell voltage. On the other hand, the blue operating point shows the opposite behaviour.

In terms of efficiency, the observed relation is in line with the explanation of the voltage of the cell and the amount of hydrogen extracted. It is reasonable to expect that higher losses result in lower hydrogen production and lead to lower overall efficiency. As a result, the simulation in green, corresponding to the highest pressure, exhibits the poorest performance. This can be attributed to the fact that the model incorporates a material balance that accounts for losses in the flow, including the pressure differential between the cathode and the anode. Consequently, the larger difference in the operating point at 30 bar results in decreased performance compared to the simulations at 7 bar. However, it is important to note that determining the optimal operating point requires consideration of numerous other factors, including cost considerations.

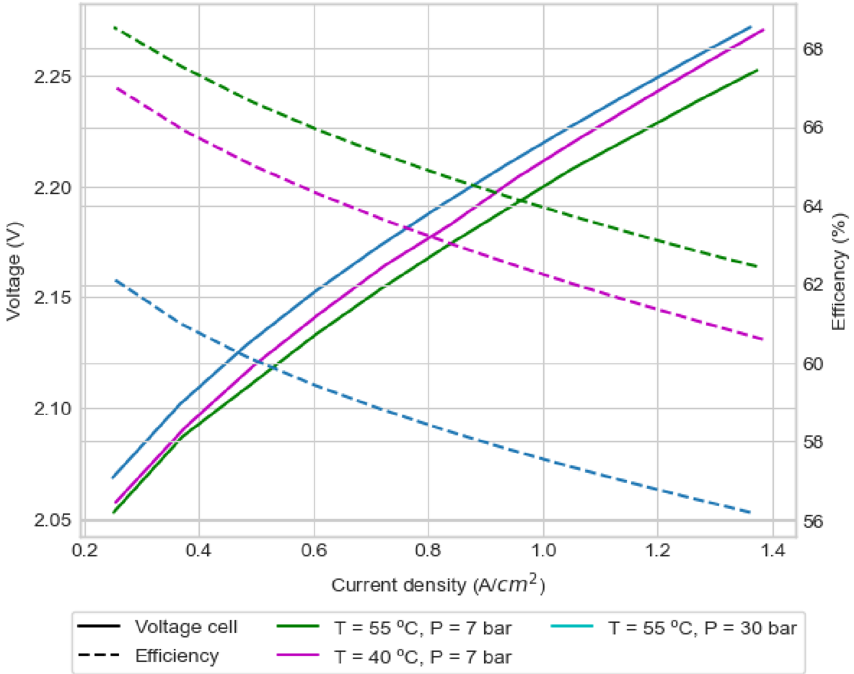


Figure 4.7: Voltage cell and efficiency compared with current density.

Figure 4.8 displays how operating points impact voltage, with each three point's maximum voltage represented by the colour code displayed in Table 4.2. The blue, green and magenta planes represent the electrical ratings of 6, 5.5 and 4 kW, respectively. It confirms that the highest voltage occurs at the highest pressure and the lowest voltage at the operating point with the lowest pressure and highest temperature. It serves to validate previous conclusions via a 3D representation.

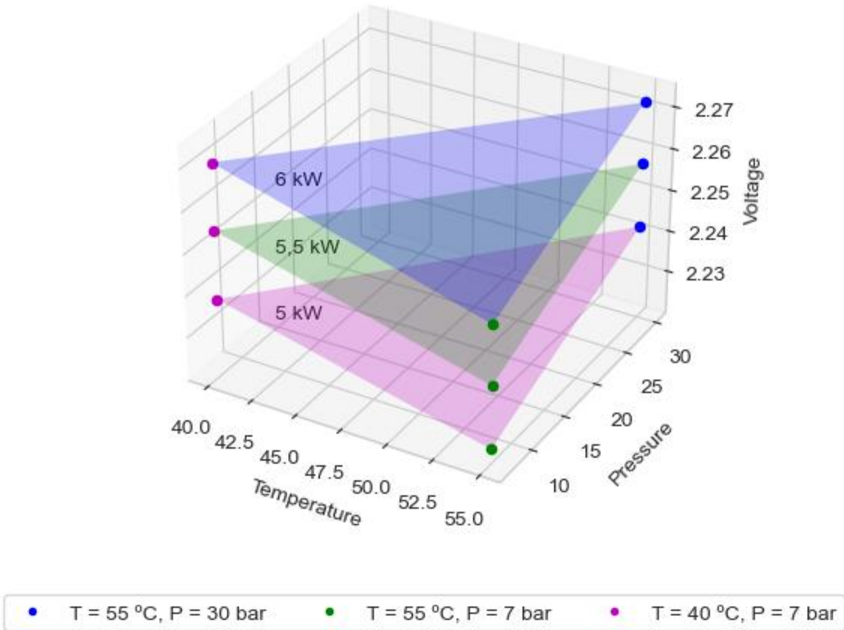


Figure 4.8: Effect of temperature and pressure on voltage cell.

The development of these simulations allows a better understanding of the impact of inputs on hydrogen production plants. Despite confirming that the simulations at 55°C and 7 bar give higher hydrogen production operating parameters, it is decided to use those corresponding to the blue operating point, 55°C and 30 bar, for the GREENSCOPE indicators. This is because the simulated data align more closely with those employed in large-scale production plants, with higher pressure levels that are more representative of the subsequent storage stage (Buttler and Spliethoff, 2018).

5 GREENSCOPE sustainability indicators

This chapter focuses on describing the GREENSCOPE indicators selected to evaluate the sustainability of PEM electrolysis technology. In addition, the alkaline production method is chosen as a point of comparison to determine if PEM electrolysis is a more sustainable option compared to other hydrogen production methods.

5.1 GREENSCOPE indicators for PEM water electrolysis

No similar research has been conducted on the application of these GREENSCOPE indicators to evaluate a PEM electrolysis process. As a result, the following proposal outlines indicators that can be utilized to assess the sustainability of hydrogen production plants that utilize PEM technology. It is important to note that in order to make these indicators comparable to other processes, reference values specific to each indicator must be established, as detailed in the explanation of each indicator. The normalization process is achieved using Equation (3.1), which is theoretically presented in Chapter 3.2 of this thesis.

i. **Global warming potential (GWP) – Environmental indicator:**

Chapter 2.2 notes that green hydrogen production methods aim at zero emission of pollutants. To quantify this environmental impact, the GWP indicator is chosen. This indicator compares the amount of CO_2 emitted to the amount of product generated, as shown in Equation (5.1). By doing so, it allows a threshold to be set that determines when the amount of CO_2 emitted becomes unacceptable for the amount of product extracted from the process (Ruiz-Mercado, Smith and Gonzalez, 2012).

$$GWP = \frac{m_{CO_2}}{m_{prod.}} \tag{5.1}$$

Where m_{CO_2} is the total CO_2 and equivalents emitted and $m_{prod.}$ is the total product mass. The best and worst scenario need it for the performance of the GWP indicator are directly extracted from the description of the indicator. The worst-case scenario assumes that all 100 of the mass released is CO_2 emissions, while the optimal scenario assumes that there is zero carbon dioxide emitted from the product (Ruiz-Mercado, Smith and Gonzalez, 2012).

ii. **Mass Loss Index (MLI) – Efficiency indicator:**

To gauge the maximum efficiency of a process, the amount of useful product extracted can be measured in relation to the mass of extracted material remaining unused. MLI indicator, Equation (5.2), assesses whether a process can generate sufficient levels of product, as a low mass efficiency results in inefficient utilization of resources (Ruiz-Mercado, Smith and Gonzalez, 2012).

$$MLI = \frac{m_{non-prod.}}{m_{prod.}} \tag{5.2}$$

Where $m_{non\text{-}prod.}$ is the total non-products release and $m_{prod.}$ is the total product mass. The MLI indicator's best and worst-case scenarios are based on its description. The worst-case scenario assumes that 100% of the extracted mass comprises O_2 emissions, which is the only non-product generated during water electrolysis. Conversely, the best-case scenario assumes that the entire extracted product consists of hydrogen (Ruiz-Mercado, Smith and Gonzalez, 2012).

iii. Fractional Water Consumption (FWC) – Efficiency indicator:

In the process of water electrolysis, water is the only raw material used. Therefore, it is important to determine whether the amount of resources used is appropriate compared to the amount of product extracted, or if there is an excessive consumption. For this purpose, the FWC indicator, Equation (5.3), is used, which compares the amount of product extracted with the amount of water consumed (Ruiz-Mercado, Smith and Gonzalez, 2012).

$$FWC = \frac{V_{fw}}{m_{prod.}} \tag{5.3}$$

Where V_{fw} is the volume of freshwater consumed and $m_{prod.}$ is the total product mass. For this indicator, the target for the best-case scenario is zero water consumption. However, for the worst-case scenario, the target should be customized to the specific process being evaluated, as the value used serves only as a reference. It is therefore set at 0,1 m^3/kg, as this value allows comparison between the two methods (Ruiz-Mercado, Smith and Gonzalez, 2012).

iv. Specific Energy Costs ($C_{E, spec.}$) – Economic indicator:

Electricity is a key component in the process of separating water into hydrogen and oxygen. Hence, it is relevant to assess whether the cost of this resource is significant. To this end, $C_{E,spec.}$ indicator, Equation (5.4), has been chosen to compare the electricity costs with the total production costs (Ruiz-Mercado, Smith and Gonzalez, 2012).

$$C_{E,spec.} = \frac{E_c}{Prod._C} \tag{5.4}$$

Where E_c is the energy cost and $Prod_c$ is the production cost. The optimal scenario for this indicator arises when there are no energy costs. The table doesn't specify a worst-scenario percentage value, just suggests that needs to be higher than 20% (Ruiz-Mercado, Smith and Gonzalez, 2012). As the cost of renewable energy is highly variable, determining how much is a high value is disparate, so it has been decided to take the average value, which is around 60% (IRENA, 2020; Nami *et al.*, 2022), and apply a 10% increase to bring it to 70%. The energy and the product cost are calculated by using the model developed by (Jovan and Dolanc, 2020). This model needs the estimation of the CAPEX of the simulated plants, which is calculated using the estimation model developed by (Reksten *et al.*, 2022).

v. Resource Energy Efficiency (η_E) – Energy indicator:

As an energy indicator, the evaluation of the amount of energy obtained in the process in the form of a product compared to the amount of energy input to the process is selected, Equation (5.5). In the case of water electrolysis, the flow of hydrogen leaving the process is the product obtained and the flow of water introduced is the energy introduced into the process (Ruiz-Mercado, Smith and Gonzalez, 2012).

$$\eta_E = \frac{E_{prod.}}{E_{in}} \tag{5.5}$$

Where $E_{Prod.}$ is the energy content in the product and the E_{in} is the total inlet energy mass. When the η_E indicator is equal to 1, it is considered to be the worst-case scenario, whereas when it is equal to 0, it is considered to be the optimal scenario. These values are indicated in the description of the GREENSCOPE indicators (Ruiz-Mercado, Smith and Gonzalez, 2012).

For the calculation of the energy content of both flows the specific heat capacity parameter is applied. This is 4184,00 J/kgK for water and 14421,00 J/kgK for hydrogen.

5.2 GREENSCOPE indicators comparison with Alkaline Water Electrolysis (AWE)

To calculate the selected indicators for the PEM electrolyser, the operating point of the simulation is chosen corresponding to an electrical power of 6 kW, a temperature of 55 °C and a pressure of 30 bar. This particular operating point is chosen because it is of a similar magnitude to the operating point used in the AWE simulation, which is selected to compare the sustainability analyses (Sánchez *et al.*, 2020). One of the reasons why the PEM simulation is compared to AWE technology is because they are two similar processes, but with different technology. Additionally, while PEM technology is prominent, it has not gained much traction in the market. Therefore, it is interesting to assess its sustainability compared to a more mature technology like AWE. The literature review in Chapter 2.3 provides a detailed explanation of these statements.

Table 5.1 summarizes the necessary data values for the computation of each sustainability indicator. It shows the data used for each indicator, which are applied in the formulas presented in Chapter 5.1.

Table 5.1: Data used for the GREENSCOPE indicators for PEM and AWE.

	PEM	AWE	GREENSCOPE indicator
H_2 product (kg/h)	0,101	0,220	GWP, MLI, FWC, η_E
O_2 extracted (kg/h)	0,026	1,355	MLI
CO_2 emissions (kg/h)	0,000	0,000	GWP
H_2O consumption (m^3/h)	0,002	0,002	FWC, η_E
Production cost (\$/kg H_2)	7,170	6,249	$C_{E, spec.}$
Energy cost (\$/kg H_2)	1,779	1,363	$C_{E, spec.}$

The data extracted from the simulations, along with the indicators described in Chapter 5.1, are used to create Table 5.2. This table presents the percentages of the indicators after normalization, using the reference values specified for each indicator.

Table 5.2: Normalization of GREENSCOPE indicators.

	GWP (%)	MLI (%)	FWC (%)	$C_{E, spec.}$ (%)	η_E (%)
PEM	100	99,74	84,68	40,91	77,50
AWE	100	93,84	92,14	48,04	79,84

After normalizing the indicators, both simulations are suitable for comparison. Therefore, a detailed analysis of each indicator and the reasons for the differences between the two production methods is presented below.

After thoroughly evaluating all GREENSCOPE environmental indicators, it is confirmed that all methods of green hydrogen production consistently achieve a perfect score of 100%. These methods avoid the use of hazardous materials and do not generate any polluting gases during their reactions. As a result, the selected **GWP** indicator is utilized as a precise and visually effective representation of the complete absence of gas or pollutant emissions in the production of green hydrogen. The score of this indicator is 100 % for both production methods, that evidence of the zero-emission of CO_2 pollutants during the process.

The results of the **MLI** indicator show that the PEM process achieves a normalization value of 99,74 %, whereas the AWE process yields a value of 93,84 %. This indicates that AWE processes are less efficient in terms of hydrogen production, as they proportionally generate more oxygen compared to PEM processes. However, the positive aspect is that oxygen, as a byproduct, is environmentally safe and can be utilized in other processes for example to reduce CO_2 emissions or in hospitals where it is needed for patient care (Kato *et al.*, 2005).

In terms of the **FWC** indicator, the AWE process demonstrates superior efficiency in utilizing the required water resource for its operation compared to the PEM process. While the PEM normalization of the indicator reflects a level of 84,68 %, the AWE process achieves significantly higher levels, reaching close to 92,00 %. This indicates that, for the same amount of water input in both processes, the AWE process generates a larger quantity of hydrogen. Thus, the AWE process is deemed more efficient, which can be attributed to the maturity of the technology and its widespread establishment in the market (Wang, Cao and Jiao, 2022).

The normalized $\mathbf{C_{E,\ spec.}}$ indicator value for the PEM process is 40,91 %, while for the AWE process, it is 48,04 %, indicating that energy cost has a greater impact on both processes. It is possible to see how the more mature AWE technology has a better cost distribution, although the difference is not very large. In the future, it is expected that this indicator may have a higher value, as the reduction of renewable energy costs is expected to decrease with the promotion of and investment in green technologies. For example, over the last 10 years, the cost of solar energy has decreased by around 88%, a value that is expected to decrease further (Osman *et al.*, 2023).

The $\boldsymbol{\eta_E}$ indicators for hydrogen production using PEM technology and AWE are 77,50 % and 79,84 % respectively, indicating that AWE has slightly higher efficiency compared to PEM. This can be attributed to the fact that PEM technology utilizes a solid polymer electrolyte with higher electrical resistance, resulting in increased energy losses as heat during electrolysis, leading to lower overall efficiency when compared to AWE (Kumar and Vurimindi, 2019).

To summarize the previous ideas and enhance the visual representation of the data from Table 5.2, Figure 5.1 is created. This figure displays the normalized values for all the indicators, along with the comparison between PEM and AWE technologies, depicted in blue and orange respectively. This graphical visualisation shows how the economic indicator, $\mathbf{C_{E,\ spec.}}$, is the worst value, reinforcing the idea that electrolysers to produce green hydrogen are not present in the market due to their low economic competitiveness.

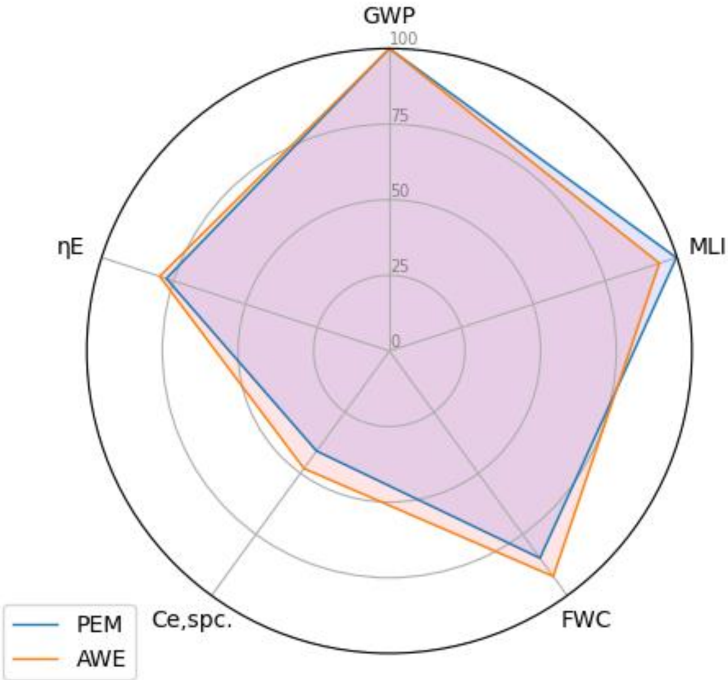


Figure 5.1: PEM and AWE comparison of GREENSCOPE indicators.

6 Process scale-up

This chapter explains the process scale-up that is carried out after simulating the PEM electrolyser. Firstly, the approach and methodology employed are discussed, followed by the linearisation of the data and the obtained results.

6.1 Approach and methodology

The purpose of this chapter is to identify a method to obtain simulated values that closely represent real-world data. This process is the preliminary step before optimizing the operating conditions of the process by maximizing the GREENSCOPE indicators. The reason for this scale-up process is that the simulation performed in Chapter 4 falls short of generating values suitable for large-scale hydrogen production.

The primary objective of the scaling-up process is to enable mathematical prediction of a less expensive prototype's performance before constructing the final product for use in the more costly and untested real-world process over the long term (Wieringa, 2014). In the field of PEM technology, scaling-up methodologies are not yet widely adopted. Only three different approaches have been identified so far. The first approach involves utilizing neural networks and implementing deep learning techniques (Tian, 2020), although this approach is not adopted due to lack of time. The second approach involves dimensional scaling through the Buckingham π theorem (Polverino *et al.*, 2019), which has been explored in fuel cell research but is not easily applicable to hydrogen production and, therefore, not suitable for this thesis. The third approach entails utilizing the mathematical PEM models to generate data within a desired range (Awasthi, Scott and Basu, 2011). However, this method lacks the flow simulation aspect that the Aspen HYSYS program provides.

For the above reasons, an alternative method has been sought to scale the values obtained from the simulation, allowing comparison with existing electrolysers on the market. Therefore, a methodology based on linearisation has been chosen to transform the data, making them suitable for the computation of regression models. The simulated data pertain to the operating conditions of 30 bar pressure and 55°C temperature. Furthermore, to verify the suitability of the simulated installation for large-scale hydrogen production, the benchmarking study conducted by Buttler and Spliethoff is used as a reference. This study includes graphical representations of the current market's PEM electrolysers (Buttler and Spliethoff, 2018).

6.2 Data linearisation

The objective of data linearisation is to apply a regression model to data that initially do not have a linear dependence. To achieve this, the data must first be plotted and analysed to determine their shape. Subsequently, a data conversion technique can be employed, which involves applying a mathematical function, such as logarithmic, inverse, square root, or others, to the variable that does not initially exhibit a linear relationship. This conversion enables the data to be transformed into a linear form, making it amenable to regression modelling and other statistical analyses (James *et al.*, 2021).

Table 6.1 presents a detailed explanation of the relationships of the variables for which regression models are sought, including the data transformations performed and the variables to which it applies. The table also includes the regression models ultimately used, along with their corresponding R-squared values, which determine their suitability for use. Notably, all R-squared values are close to 1, indicating the high degree of fit and confirming the suitability of the generated regression models for the study's purposes.

Table 6.1: Data linearization for scaling-up process.

Abscissa (x)	Ordinate (y)	Transformation	Regression model	R-squared
Current density	Voltage cell	$\sqrt{x}$	$y = 0{,}39\ \sqrt{x} + 1{,}85$	0,96
Current density	Specific work	log (x)	$y = 0{,}42\ \log(x) + 5{,}27$	0,94
Current density	Efficiency	log (x)	$y = -0{,}04\ \log(x) + 0{,}57$	0,99

The following figures provide an illustrative example of how the current and voltage density data have been linearised. Figure 6.1 shows the nonlinear relationship between current density and voltage cell. Figure 6.2 shows how the application of a square root transformation to the abscissa results in the linearisation of the data, which is then modelled using a regression model.

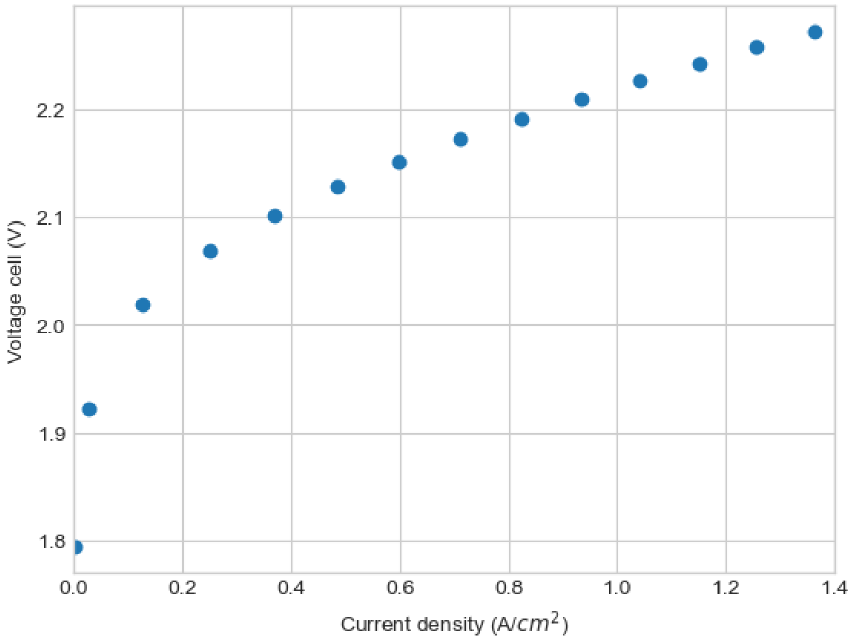


Figure 6.1: Non-linear correlation between current density and voltage cell.

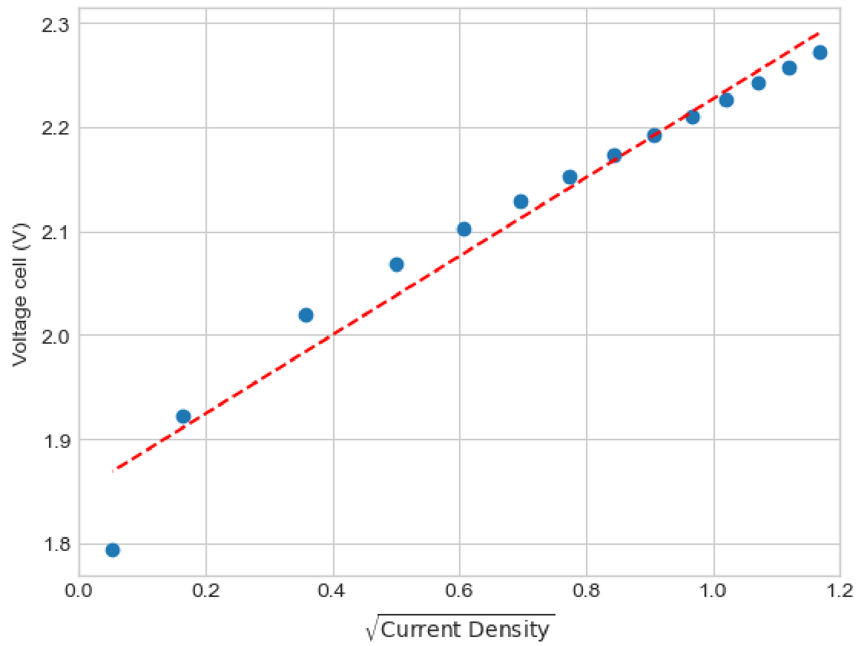


Figure 6.2: Linearisation of the current density.

6.3 Scaling-up validation process

After linearising the variables, Figure 6.3 is created to reproduce Figure 7 of the article presented by Buttler and Spliethoff, which serves as a benchmark for comparing the scaling process (Buttler and Spliethoff, 2018). The figure displays the current density against the voltage, specific work, and cell efficiency. The polarization line is shown in blue, while the efficiency using the lower heating value (LHV) as the reference value is displayed in grey dashed lines. Furthermore, a second y-axis is added, representing the specific work values.

Compared with the polarization curves of actual PEM electrolysers presented in the article by Buttler and Spliethoff, it is noticeable that the one obtained from the simulated case exhibits a logical maximum current value, falling within the range of those operating with similar points. However, the minimum voltage is approximately 1,8 V, while in commercial electrolysers, this value is consistently below 1,75 V and close to 1,5 V. Hence, simulating at low values is unsuitable. Although an attempt was made to address this problem via regression, the R-squared values obtained were too low.

The specific work values are found to be adequately adjusted to those shown in the reference article. Hence, it can be concluded that this variable can be compared to that of real electrolysers. Furthermore, the efficiency values obtained are realistic, when compared to those shown in the article, indicating that the scale-up of the simulation is satisfactory for the efficiency of the electrolyser.

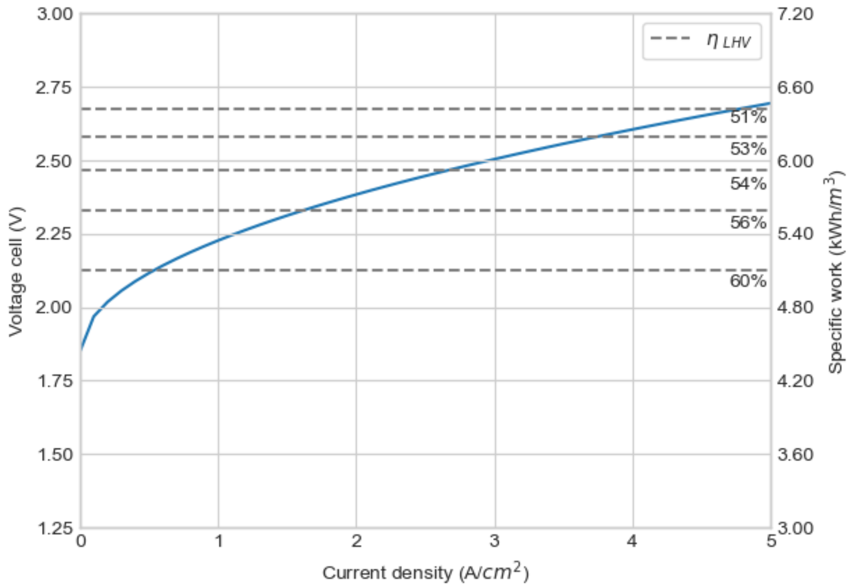


Figure 6.3: Scaling-up process for the simulated model.

7 Conclusions

This chapter begins with a review of the objectives established at the outset of the thesis. Subsequently, general conclusions are drawn, followed by suggestions for possible future research.

7.1 Assessment of initial objectives

Chapter 1.3 outlines the objectives of the present thesis. Below, a comprehensive review is undertaken to determine if these objectives were achieved, and if so, which were the main achievements:

- O1. A comprehensive literature review was conducted on green hydrogen production methods, as detailed in Chapter 2, and chemical sustainability, as detailed in Chapter 3. As a result of this review, an electrolyser based on PEM technology was discovered as the most effective method for producing green hydrogen. Additionally, the GREENSCOPE approach was identified as the most suitable method for evaluating the sustainability of the process.
- O2. The Aspen HYSYS simulation was successfully performed and validated using literature data, as detailed in Chapter 4. Additionally, the simulation of different operating points provided a comprehensive understanding of the effects of temperature, pressure, and electrical power on the PEM electrolyser.
- O3. In Chapter 5, the sustainability analysis was completed using the GREENSCOPE approach. The chosen indicators were used to evaluate the sustainability of the process and enabled a comparison with a simulated electrolyser based on AWE technology.
- O4. The scaling-up process was carried out using a technique based on data linearisation and regression, as described in Chapter 6. This method allowed for scaling-up the data of the simulation and a comparison of its performance, based on literature data, with electrolysers present in the market.

7.2 General conclusions

After the literature review was conducted, it was established that the predominant hydrogen production methods in use at the time generated significant CO_2 emissions. As a result, green hydrogen production through water electrolysis, powered by renewable electricity, emerged as the most promising solution. PEM electrolysers were found to be particularly well-suited for accommodating the variability of renewable energy sources and allowing for operation at higher pressures, ultimately reducing costs. Therefore, this method of hydrogen production was chosen for the development of the thesis.

The Aspen HYSYS simulation exhibited satisfactory performance when compared to the reference simulation. However, it was not possible to accurately simulate electrical power ranges below 1 kW, leading to the operating range being restricted to 1-6 kW for subsequent operational point analysis. Through analysing different points, it was determined that higher temperature and electrical power levels increased hydrogen production, while pressure had an inversely proportional effect. This finding is consistent with observations made in various real and simulated PEM electrolysers documented in the literature.

The sustainability analysis performed consisted of calculating indicator values according to the GREENSCOPE methodology, which was used for the first time in this thesis to evaluate hydrogen production methods. In addition, a sustainability comparison was made between PEM and AWE technologies. The environmental indicator, GWP, was found to be 100 for both technologies as green hydrogen production does not generate CO_2 emissions. With regards to the efficiency indicators, the proportion of hydrogen produced, MLI, was found to be higher for PEM technology than for AWE, while water consumption, FWC, compared to the amount of hydrogen generated was better for AWE technology. In terms of the economic indicator, $C_{E, spec}$., it was observed that the weight of energy costs was higher in the case of PEM technology. Finally, the efficiency indicator showed that the energy efficiency, ηE, was slightly worse for PEM technology.

The scaling-up process was performed using techniques that differ from those presented in the literature. Specifically, data linearisation and regression were used. This process confirmed that the simulation can be compared satisfactorily with commercially available PEM electrolysers. However, the only issue encountered was the lower voltage value, which was slightly above the normal operating range of commercially available electrolysers. This is consistent with the previously explained issue of inadequate simulation performance at low electrical power ranges.

7.3 Future research

Following are presented some future works that can be conducted to extend the research developed in the present thesis:

- Dynamic and increment range simulation: Understanding the evolution of heat transfer inside the electrolyser could help to determine more accurately the optimal operating point at which the electrolyser operates. Moreover, being able to increase both low and high-power ranges simulation would help the simulation achieve results closer to real electrolysers.
- Process optimization: Develop an algorithm to obtain the optimal operating point of the simulation in terms of maximisation of the GREENSCOPE indicators. This will allow the PEM simulation to perform better in certain sustainability areas where it currently lags behind AWE technology. This should be done after process scaling-up if the simulation cannot reach a high enough simulation range to be realistic.
- Process scale-up using Artificial Neural Networks (ANN) or Buckingham π theorem: In the existing literature, there are scaling-up processes for hydrogen fuel cells simulations using PEM technology, but there are no methods based on ANN or Buckingham π theorem for electrolysers simulations. The development of such methods could lead to significant cost reductions in the development of commercialised electrolysers.

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Appendices

Appendix A: Initial thesis description.

FMH606 Master's Thesis

Title: Modelling of hydrogen technologies for sustainability

USN supervisor: Gaurav Mirlekar, Associate Professor, USN

External partner: Not yet assigned

Task background:
The use of hydrogen as an energy carrier holds promise for the future. Hydrogen value chainfrom production technologies (for example, natural gas reforming and water electrolysis) to storage, transport, and utilization play an important role in addressing energy challenges.
These processes are modeled using mass and energy balances consisting of differential and algebraic equations. The main objective of the thesis is to perform steady-state or dynamic modelling studies of hydrogen production based on polymer electrolyte membrane (PEM) electrolysis and compare results with experimental data available from a hydrogen production plant. The evaluation of optimum solution for producing hydrogen, using hydrogen for energy storage, and subsequently utilizing hydrogen for efficient power generation should be conducted. Furthermore, these processes must be analyzed as sustainable energy systems. Using the GREENSCOPE indicators, the sustainability of the process will be quantified and optimized.

Task description:
1. Literature review on hydrogen technologies, process modelling and sustainability.
2. Evaluate possible process configuration in simulation software, such as Aspen HYSYS,Aspen Plus or programming languages such as MATLAB or python.
3. Perform process simulation, conduct optimization, and critically analyse theperformance in terms of sustainability.

Student category: Reserved for JORDI BÉJAR RABASCALL (IIA student)

Is the task suitable for online students (not present at the campus)? No

Practical arrangements: A computer and software are required to perform simulation andmodelling tasks.

Supervision:
As a general rule, the student is entitled to 15-20 hours of supervision. This includes necessary time for the supervisor to prepare for supervision meetings (reading material tobe discussed, etc).

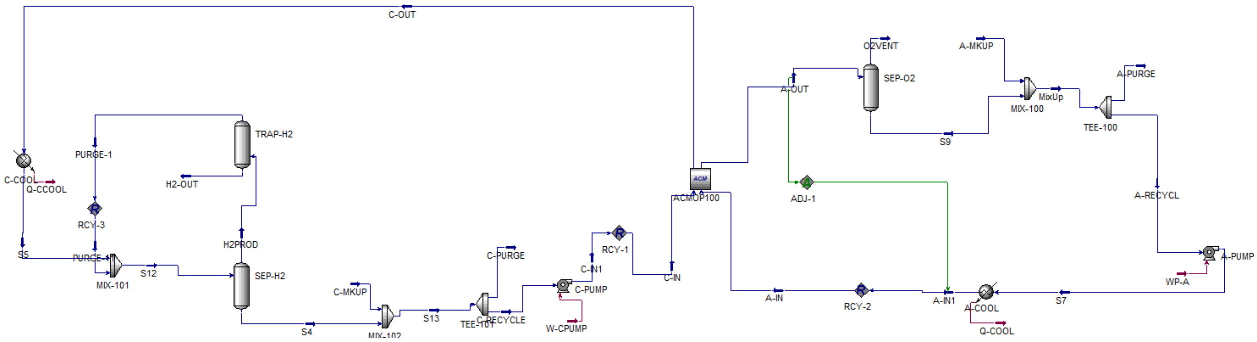
Signatures:

Supervisor (date and signature): Gaurav Mirlekar 01.02.2023

Student (write clearly in all capitalized letters): Jordi Béjar Rabascall

Student (date and signature): 01/02/2023

Appendix B: Aspen HYSYS Simulation schematic.



Appendix C: Python code for computing the GREENSCOPE indicators.

```
#Imports
import matplotlib.pyplot as plt
import pandas as pd
from math import pi
import plotly.express as px
import pandas as pd

#import data
df_gs = pd.read_excel("data_Greenscopeindicators.xlsx")

#Exploratory analysis
print(df_gs.head())
df_gs.axes

#Indicator 1 - Global Warming Potencial (GWP) - Enviromental (23)
co2_pem = df_gs["Kg CO2 Prod"][0]
co2_awe = df_gs["Kg CO2 Prod"][1]
h2_pem = df_gs["H2 prod  (kg/h)"][0]
h2_awe = df_gs["H2 prod  (kg/h)"][1]
fwc_w = 100
fwc_b = 0
gwp_pem = co2_pem/h2_pem
gwp_awe = co2_awe/h2_awe
pgwp_pem = ((gwp_pem-fwc_w)/(fwc_b-fwc_w ))*100
pgwp_awe = ((gwp_awe-fwc_w)/(fwc_b-fwc_w ))*100

#Indicator 2 - Fractional water consumption (FWC) - Efficency (24)
h2o_pem = df_gs["H2O cons (kg/h)"][0]*0.001 #Pas from kg/h to m3/h
h2o_awe = df_gs["H2O cons (kg/h)"][1]*0.001 #Pas from kg/h to m3/h
h2_pem = df_gs["H2 prod  (kg/h)"][0]
h2_awe = df_gs["H2 prod  (kg/h)"][1]
fwc_pem = h2o_pem/h2_pem
fwc_awe = h2o_awe/h2_awe
fwc_w = 0.1 #Change
fwc_b = 0
pfwc_pem = ((fwc_pem-fwc_w)/(fwc_b-fwc_w ))*100
pfwc_awe = ((fwc_awe-fwc_w)/(fwc_b-fwc_w ))*100

#Indicator 3 - Mass lose Index (MLI) - Efficency (11)
o2_pem = df_gs["O2 (kg/h)"][0]
o2_awe = df_gs["O2 (kg/h)"][1]
mli_pem = o2_pem/h2_pem
mli_awe = o2_awe/h2_awe
mli_w = 100
```

```
mli_b = 0
pmli_pem = ((mli_pem-mli_w)/(mli_b-mli_w ))*100
pmli_awe = ((mli_awe-mli_w)/(mli_b-mli_w ))*100

#Indicator 4 - Specific Energy Cost (Ce,spc.) - Economic (23)
dy = 365 #Days per year
oy = 15 #Years of operation
r = 0.8 #Operational rate
ppem = df_gs["Power (kW)"][0]
pawe = df_gs["Power (kW)"][1]
capex_pem = ((585.85+9458.2/ppem*ppem**0.622)*(2023/2020)**-158.9)*ppem #$ (initial
inversion)
capex_awe = ((301.04+11603/pawe*pawe**0.649)*(2023/2020)**-27.33)*pawe #$ (initial
inversion)
opex_pem = capex_pem*0.05*oy #$ in 15 years
opex_awe = capex_awe*0.05*oy #$ in 15 years
th2_pem = h2_pem*24*dy*oy*r #kg in 15 years
th2_awe = h2_awe*24*dy*oy*r #kg in 15 years
PC1_pem = (opex_pem + capex_pem)/th2_pem #$/kg
PC1_awe = (opex_awe + capex_awe)/th2_awe #$/kg
ec_pem = ppem/h2_pem #kwh/kg
ec_awe = pawe/h2_awe #kwh/kg
PC2_pem = df_gs["Electrecity cost ($/MWh)"][0]*0.001*ec_pem #$/kg
PC2_awe = df_gs["Electrecity cost ($/MWh)"][1]*0.001*ec_awe #$/kg
Cp_pem = PC1_pem + PC2_pem #$/kg
Cp_awe = PC1_awe + PC2_awe #$/kg
ce_pem = PC2_pem/Cp_pem
ce_awe = PC2_awe/Cp_awe
ce_w = 0.7
ce_b = 0
pce_pem = ((ce_pem-ce_w)/(ce_b-ce_w ))*100
pce_awe = ((ce_awe-ce_w)/(ce_b-ce_w ))*100

#Indicator 5 - Resource energy efficiency (ηE) - Energy (6)
cph2o = df_gs["Cp h20 (J/kgK)"][0]*0.001 #kJ/kgk
cph2 = df_gs["Cp h2 (J/kgK)"][0]*0.001 #kJ/kgk
h2o_pem = df_gs["H2O cons (kg/h)"][0]
h2o_awe = df_gs["H2O cons (kg/h)"][1]
inkjh_pem = h2o_pem*cph2o*df_gs["T (ºC)"][0]
inkjh_awe = h2o_awe*cph2o*df_gs["T (ºC)"][1]
okjh_pem = h2_pem*cph2*df_gs["Tout (ºC)"][0]
okjh_awe = h2_pem*cph2*df_gs["Tout (ºC)"][1]
r_pem = okjh_pem/inkjh_pem
r_awe = okjh_awe/inkjh_awe
r_w = 1
r_b = 0
pr_pem = ((r_pem-r_w)/(r_b-r_w ))*100
pr_awe = ((r_awe-r_w)/(r_b-r_w ))*100
```

```
ind = ["GWP","MLI","FWC","Ce,spc.","ηE"]
pem = [pgwp_pem, pmli_pem,pfwc_pem , pce_pem, pr_pem, pgwp_pem]
awe = [pgwp_awe,pmli_awe ,pfwc_awe , pce_awe, pr_awe, pgwp_awe]
N = len(ind)
angles = [n / float(N) * 2 * pi for n in range(N)]
angles += angles[:1]
ax = plt.subplot(111, polar=True)
ax.set_theta_offset(pi / 2)
ax.set_theta_direction(-1)
plt.xticks(angles[:-1], ind)
ax.set_rlabel_position(0)
plt.yticks([0,25,50,75,100], ["0","25","50","75","100"], color="grey", size=7)
plt.ylim(0,100)
values=pem
ax.plot(angles, values, linewidth=1, linestyle='solid', label="PEM")
ax.fill(angles, values, 'b', alpha=0.1)
values=awe
ax.plot(angles, values, linewidth=1, linestyle='solid', label="AWE")
ax.fill(angles, values, 'r', alpha=0.1)
plt.legend(loc='upper right', bbox_to_anchor=(0.1, 0.1))
plt.figlegend(handles, labels, loc='upper left', bbox_to_anchor=(0.75, 1))
```

Appendix D: Python code for the data linearisation, regressions models and scaling-up process

```
#Imports
import pandas as pd
import numpy as np
import matplotlib.pyplot as plt
from scipy.optimize import curve_fit
from scipy.stats import linregress
from scipy.special import jv
import math
from sklearn.linear_model import LinearRegression

#Import data
df_ACM = pd.read_excel("data_ACM.xlsx")
df_S = pd.read_excel("data_Steams.xlsx")
df_E = pd.read_excel("data_Energy.xlsx")
df_ID = pd.read_excel("data_IDinfo.xlsx")

#Plot Current density vs Vcell
plt.style.use('default')
plt.style.use('seaborn-whitegrid')
Vcell = df_ACM ["VCELL (V)"]
CurrDen = df_ACM ["I_USEFUL (A/m2)"]
Vcell = np.array(Vcell)
CurrDen = np.array(CurrDen)*0.0001
plt.scatter(CurrDen, Vcell)
plt.xlabel('Current density (A/$cm^2$)')
plt.ylabel('Voltage cell (V)')
plt.xlim(0,1.4)
plt.show()
#Linearization
sqrt_x = np.sqrt(CurrDen)
model = LinearRegression()
model.fit(sqrt_x.reshape(-1,1), Vcell)
```

```
r_squared = model.score(sqrt_x.reshape(-1,1), Vcell)
m = model.coef_[0]
n = model.intercept_
y_predicted = m * sqrt_x + n

plt.scatter(sqrt_x, Vcell)
plt.xlabel('Current density (A/$cm^2$)')
plt.ylabel('Voltage cell (V)')
plt.plot(sqrt_x,y_predicted, color = "red", linestyle = "--")
plt.xlabel(r'$\sqrt{\mathrm{Current\ Density}}$')
plt.ylabel('Voltage cell (V)')
plt.xlim(0,1.2)
plt.show()

print(m)
print(n)
print('R-squared:', r_squared)

#Scale up
n_x = np.arange(0, 5.1, 0.1)
n_y_v = m * np.sqrt(n_x) + n
plt.plot(n_x,n_y_v, color = "blue", linestyle = "-")
plt.xlabel('CurrDen')
plt.ylabel('Vcell')
plt.show()
print(n_y_v)

#Plot Current density vs Specific work (Kwh/Nm3)
plt.style.use('default')
plt.style.use('seaborn-whitegrid')
H2Prod = df_S.loc[df_S['Steam'] == 'H2PROD', 'Mass Flow [kg/h]']
ElecP = df_ACM ["WSTACK (kW)"]
ElecP = np.array(ElecP)
H2Prod = np.array(H2Prod)*11.126
```

```
sw = ElecP/H2Prod
plt.scatter(CurrDen, sw)
plt.xlabel('CurrDen')
plt.ylabel('Specific Work')
plt.show()

#Linearization
log_x = np.log(CurrDen)
plt.scatter(log_x, sw)
plt.xlabel('CurrDen')
plt.ylabel('Specific Work')

model = LinearRegression()
model.fit(log_x .reshape(-1,1), sw)
r_squared = model.score(log_x .reshape(-1,1), sw)

m = model.coef_[0]
n = model.intercept_
y_predicted = m * log_x + n
plt.plot(log_x,y_predicted, color = "red", linestyle = "--")
plt.xlabel('CurrDen')
plt.ylabel('Specific Work')
plt.show()
plt.show()

print(m)
print(n)
print('R-squared:', r_squared)

#Scale up
n_x = np.arange(0.1, 5.1, 0.1)
n_y = m * np.log(n_x) + n
n_x = np.insert(n_x, 0, 0)
n_y_sw = np.insert(n_y, 0,2.37899751)
```

```
plt.plot(n_x,n_y_sw, color = "blue", linestyle = "-")
plt.xlabel('CurrDen')
plt.ylabel('Specific Work')
plt.show()

#Current Density vs Eff
plt.style.use('default')
plt.style.use('seaborn-whitegrid')
H2Prod = df_S.loc[df_S['Steam'] == 'H2PROD', 'Mass Flow [kg/h]']
H2Prod = np.array(H2Prod)*1000/3600
CurrDen = df_ACM ["I_USEFUL (A/m2)"]
CurrDen = np.array(CurrDen)*0.0001
LHV = 120
Eff = (H2Prod*LHV)/ElecP
CurrDen = CurrDen[:-1]
Eff = Eff[:-1]
plt.scatter(CurrDen, Eff)
plt.xlabel('CurrDen')
plt.ylabel('Eff')
plt.show()

#Linearization
logx = np.log(CurrDen)
model = LinearRegression()
model.fit(logx.reshape(-1,1), Eff)
r_squared = model.score(logx.reshape(-1,1), Eff)

m = model.coef_[0]
n = model.intercept_
y_predicted = m * logx + n
plt.scatter(logx, Eff)
plt.xlabel('CurrDen')
plt.ylabel('Eff')
plt.plot(logx,y_predicted, color = "red", linestyle = "--")
```

```
plt.xlabel('CurrDen')
plt.ylabel('Eff')
plt.show()

print(m)
print(n)
print('R-squared:', r_squared)

#Scale up
n_x = np.arange(0.1, 5.1, 0.1)
n_y = m * np.log(n_x) + n
n_x = np.insert(n_x, 0, 0)
n_y_e = np.insert(n_y, 0,0.9178704303319385)
plt.plot(n_x,n_y_e, color = "blue", linestyle = "-")
plt.xlabel('CurrDen')
plt.ylabel('Eff')
plt.show()
print(m * np.log(0.5) + n)
print(m * np.log(1.5) + n)
print(m * np.log(2.5) + n)
print(m * np.log(3.5) + n)
print(m * np.log(4.5) + n)
```

Appendix E: Python code for the scaling-up process validation

```
import matplotlib.pyplot as plt
import matplotlib.ticker as ticker


fig, ax1 = plt.subplots()
ax1.plot(n_x, n_y_v)
ax1.set_xlabel('Current density (A/$cm^2$)')
ax1.set_ylabel('Voltage cell (V)')
ax1.set_xlim([0, 5])

# Set the y tick values and tick labels for ax1
ax1.set_ylim([1.25, 3])
ax1.yaxis.set_major_locator(ticker.MultipleLocator(0.25))
ax1.yaxis.set_major_formatter(ticker.FormatStrFormatter('%.2f'))

ax2 = ax1.twinx()
line2, = ax2.plot(n_x, n_y_sw)
line2.set_visible(False)
ax2.set_ylabel('Specific work (kWh/${m^3}$)')

# Set the y tick values and tick labels for ax2
ax2.set_ylim([3, 7.20])
ax2.yaxis.set_major_locator(ticker.MultipleLocator(0.6))
ax2.yaxis.set_major_formatter(ticker.FormatStrFormatter('%.2f'))
# Add vertical dashed lines for the efficiency values
# Add a vertical line at x=3
ax1.axhline(y=0.39*np.sqrt(0.5)+1.85, color='gray',linestyle = "--")
ax1.axhline(y=0.39*np.sqrt(1.5)+1.85, color='gray',linestyle = "--")
ax1.axhline(y=0.39*np.sqrt(2.5)+1.85, color='gray',linestyle = "--")
ax1.axhline(y=0.39*np.sqrt(3.5)+1.85, color='gray',linestyle = "--")
ax1.axhline(y=0.39*np.sqrt(4.5)+1.85, color='gray',linestyle = "--")

# Add a text to explain the efficiency represented by the vertical line
```

```
ax1.text(4.7, 2.07, "60%", rotation=0)
ax1.text(4.7, 2.27, "56%", rotation=0)
ax1.text(4.7, 2.4, "54%", rotation=0)
ax1.text(4.7, 2.52, "53%", rotation=0)
ax1.text(4.7, 2.63, "51%", rotation=0)

#Legend
custom_label = [plt.Line2D([0], [0], color='gray', linestyle='--')]
ax1.legend(custom_label, ['$\eta$ $_{LHV}$'],frameon=True,loc='upper right')

plt.show()
```