CURRENT STATE AND DECARBONISATION OPTIONS FOR THE DUTCH POLYOLEFINS INDUSTRY

An overview on polyolefins production in the Netherlands, and what are the possibilities to reduce its impact on climate change



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Abstract

This master thesis deals with the decarbonisation options for the Dutch polyolefins industry. The main products (LDPE, LLDPE, HDPE, PP) are mostly used for packaging (60%), construction (10%), and automotive (7%). Three producers have been investigated: Dow (in Terneuzen, Zeeland), SABIC (in Geleen, Limburg), and Ducor (in Rozenburg, South-Holland). Their aggregate production of polyolefins is 2,570 kt/y, which is 2.5 times higher than the Dutch demand and equal to 8.5% of the total European production. The production sites consume 4,774 TJ/y of electricity and 644 TJ/y of heat in total, which are linked with the yearly emission of around 800 kt CO₂-eq. The main polymerisation technologies are high-pressure, solution, slurry, and gas-phase, causing the emission of 0.20-0.32 kg CO₂-eq/kg of product. As most of the GHGs are emitted upstream and downstream the polymerisation phase, this study focuses on the decarbonisation options associated to the production of feedstock and the endof-life treatments. Olefin monomers can be produced from bio-ethanol, which is obtained from the fermentation of sugar-rich crops (e.g. sugar beets). The carbon emissions are greatly reduced, but land and water use have to be considered, and the cost of the product is 60% higher than when using fossilbased feedstock. Another possible route is the gasification of bio-waste (e.g. from agriculture, the paper industry and cooking oils) and plastic waste, although the process is technically complicated and not yet ready for large scale applications. Besides the already-adopted mechanical recycling, potential circular economy options are dissolution recycling and pyrolysis, a thermochemical process able to handle hard-to-recycle plastics and recover the olefin monomers. Both technologies are considered promising by the stakeholders, but they are still under development and is thus uncertain if they are the best choices to process the plastic waste. In conclusion, more economic incentives and more innovation are needed to successfully decarbonise the Dutch polyolefins industry.

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Structure of the thesis

N°	Chapter	Description
1	Introduction	This chapter defines the problem and the knowledge gap that led to the start of the research. Scope and objectives are then explicated in the form of research questions.
2	Background information	This chapter presents background knowledge and definitions that, although are not necessarily used in the research, are useful to increase the familiarity with polyolefin plastics.
3	Methodology	This chapter offers an overview of the research methods used to answer all the research questions, listing the main sources, assumptions, and data processing steps.
4	Results	This chapter goes through the research questions one by one according to the methodology, presenting the findings in a combination of text, tables, and figures.
5	Discussion	This chapter hosts the discussion about the limitations of the research, and the societal impacts and scientific relevance of the results.
6	Conclusion	This chapter serves as the culmination of the thesis, summarizing together all the main findings and their societal and scientific relevance.

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List of abbreviations

BAT	Best Available Technique
CAPEX	Capital Expenditures
CCS	Carbon Capture and Storage
CCU	Carbon Capture and Utilization
СНР	Combined Heat and Power
СО	Carbon Monoxide
CO2	Carbon Dioxide
EEI	Energy Efficiency Improvement
EFA	Energy Flow Analysis
EU ETS	European Union Emission Trading System
GHG	Greenhouse Gases
GWP	Global Warming Potential
HDPE	High-density Polyethylene
IHP	Industrial Heat Pump
LDPE	Low-density Polyethylene
LLDPE	Linear Low-density Polyethylene
MFA	Material Flow Analysis
MSF	Municipal Solid Waste
MTO	Methanol-to-Olefins
MIDDEN	Manufacturing Industry Decarbonisation Data Exchange Network
NEa	Dutch Emission Authority (Nederlandse Emissieautoriteit)
NOX	Nitrogen Oxides
OPEX	Operating Expenditures
PE	Polyethylene
РР	Polypropylene
PV	Photovoltaic
TRL	Technology Readiness Level
WGS	Water-Gas Shift

1. INTRODUCTION

1.1 Societal background and problem definition

In the last decades, planet Earth and human society have experimented tremendous changes, with the world population growing by 42% (World Bank, 2020), the final consumption of energy increasing by 51%, and CO2 emissions rising by 58% (IEA, 2020) in the period from 1990 to 2017. This incredible growth of human activity has unequivocal impacts on the climate system, and the observed environmental changes have reached unprecedented magnitudes. Anthropogenic greenhouse gases (GHG) emissions led to increasing temperatures of atmosphere and oceans, and the last 30 years have been the warmest of the last 14 centuries, with most projections forecasting an increase of 2-4°C in 2100 (IPCC, 2014). The negative effects of climate change are widespread over all continents and include the rapid increase of heatwaves, floods, droughts, hurricanes, ocean acidification, desertification, the melting of the polar ice caps, and the consequent rise of the sea level (NASA, 2020), which is forecasted to grow between 0.3 and 1.0 meters before the end of the century (IPCC, 2014). The worry for the future of mankind has led to the development of environmental sciences and the creation of worldwide treaties like the Kyoto Protocol in 1997 and the Paris Agreement in 2016, when 196 state parties ratified their common determination to reduce GHG emissions and keep the global temperature rise below 2°C (UNFCCC, 2020). According to the EU Climate & Energy framework, in the European Union this effort has been quantified with the target of cutting 40% of GHG emissions by 2030 (European Commission, 2020), and the new European Green Deal has the goal to make the EU climate neutral by 2050, making energy, buildings, industry, and transport sustainable and boosting circular economy (EU Green Deal, 2020).

Parallel to the global climate change debate, worldwide actions are needed to reduce the consumption of natural resources and manage the consequent waste streams, including litter and pollution. In the early 1970s, the Club of Rome published its famous report "The Limits to Growth", advocating the idea that the ever-increasing demands of the global economy would have exhausted the reserves of materials and fossil fuels within a few decades (Meadows et al., 1972). Although new reservoirs were discovered and the prediction resulted wrong, it is undeniable that the current consumption patterns are not sustainable in the long run. From 1990 to 2016, the global production of oil grew by 40% and reached 4.4 million ktoe, with about a quarter of it being supplied to the industry sector and nonenergy uses, i.e. the oil is used as raw material for the production of plastics, bitumen, lubricants, etc. (IEA, 2020). It is safe to assume that one of the drivers of oil demand is plastics manufacturing, which in the last 30 years has triplicated (World Economic Forum, 2020), reaching an annual production of 360 million tonnes in 2018 (PlasticsEurope, 2019). When considering the cumulative impact of the last century, it is estimated that 6,300 million tons of plastic waste have been generated, of which almost 80% was amassed in landfills or dispersed in the environment (Geyer et al., 2017). Among the deleterious effects of plastic pollution, a large number of marine species (including seabirds) is harmed and/or killed through entanglement and ingestion of plastic debris (Derraik, 2002), and the trophic transfer process can potentially spread microplastics throughout the entire food chain, with hazardous effects on the health of humans and animals (Suaria et al., 2016). The buoyant properties of most plastics and the circulation patterns of ocean currents have led to the formation of the Great Pacific Garbage Patch, an area of 1.6 million km² with an estimated amount of 1.8 trillion floating plastic pieces, causing incalculable effects on the local and global biodiversity, food supply security, and health (Lebreton et al., 2018).

In 2015 the Member States of the United Nations adopted the "2030 Agenda for Sustainable Development", which includes 17 Sustainable Development Goals (SDG) to bring peace, progress, and prosperity to humanity and to our planet. Plastics pollution, especially micro-plastics, is part of SDG 3

(good health and well-being), SDG 6 (clean water and sanitation), SDG 14 (life below water), and SDG 15 (life on land). The energy and raw materials required for plastics manufacturing, on the other hand, have huge impacts on SDG 12 (responsible consumption and production) and SDG 13 (climate action) (UN, 2020), as well as the carbon emissions caused by the incineration of plastic waste. One of the signatories is the Netherlands, the fourth largest chemical producer in Europe and tenth worldwide, with a sector turnover of 50 billion euros that constitutes 6% of the national GDP (World Bank, 2020) and 13% of the industrial added value (VNCI, 2020). Like many other European countries, the Netherlands has developed a national plan to achieve the targets agreed on in the Paris Agreement, and its "klimaat-akkoord" has the ambition of reducing GHG by 49% in 2030 compared to 1990 levels, with the industrial sector alone reducing its emissions by 59% (Klimaat-akkoord, 2019). Among the other subsectors, manufacturers of plastic monomers and polymers are thus in the need of improving the energy and material efficiency of their production processes and reduce the associated GHG emissions. Additionally, last year the Ministry of Infrastructure and Water Management launched the Dutch Plastic Pact, signed by more than 90 manufacturers and retailers to make single-use plastic products and packaging more sustainable and more suitable for reuse (RIVM, 2020), and a European Plastic Pact involving a vast network of companies, states, and organisations (EPP, 2020).

1.2 Scientific background and knowledge gap

Given the relevance of the topic, the scientific literature regarding energy savings and reduction of CO₂ emissions is rich. Dunkelberg and colleagues, for instance, researched innovative options to optimize the efficiency of the plastics industry in three different locations in the USA, Canada and Germany, focusing on innovative smart-grids to shift the energy supply between electricity and gas (Dunkelberg et al., 2018) Another study analysed energy-saving measures and waste heat recovery options for plastic processing plants (Schlüter & Rosano, 2016), and the energy efficiency of plastic polymer plants operating under different climatic conditions has been investigated too (Khripko et al., 2016). To the best of the author's knowledge, however, no studies are covering the specific situation and/or providing decarbonisation options and strategies for the plastics industry in the Netherlands. To facilitate the achievement of its national targets, the Dutch government has entrusted PBL (Netherlands Environmental Assessment Agency) and TNO (Netherlands Organisation for Applied Scientific Research) to fill this knowledge gap. The collaboration of the two entities resulted in the creation of the MIDDEN project (Manufacturing Industry Decarbonisation Data Exchange Network), which has the goal of gathering up-to-date information on Dutch industrial locations, processes, and products, along with a wide variety of decarbonisation options for those processes (PBL, 2020).

1.3 Research scope, objective, and questions

As the MIDDEN covers the whole Dutch industrial sector, many researchers from TNO and PBL are involved in the projects, each focusing on a different sub-sector (e.g. paper, ceramics, natural gas, etc). This paper is dedicated to the production of polyolefins, namely polyethylene (PE) and polypropylene (PP), which together constitute half of the European demand for plastics (PlasticsEurope, 2019).

The goal of this master's thesis and connected MIDDEN report is thus to provide a complete and transparent overview of the PE and PP industry in the Netherlands, the connected energy and material flows, the associated emissions of GHG and the decarbonisation options able to reduce the impact on the environment. This objective will be achieved by answering the following main research question:

What is the current state of polyolefins production in the Netherlands, and what are the potential decarbonisation options to mitigate its impact on climate change?

This research question can be divided into four sub-questions:

- 1. What are the main applications of polyolefins, and what is the volume demand of each consumer sector in the Netherlands?
- 2. Who are the main producers of polyolefins in the Netherlands, and what are their production volumes, energy use, and carbon emissions?
- 3. What are the main polymerisation processes, and what are their associated costs, energy and material uses, and CO₂ emissions?
- 4. Which available decarbonisation options are considered as most promising by the stakeholders, and to what extent are they technically, economically, and environmentally comparable to the current technologies?

1.4 Societal and scientific relevance

The master's thesis will be written while working at TNO Utrecht as part of the MIDDEN project, with the goal of supporting industry and policymakers in their efforts to achieve deep decarbonisation in the Netherlands. By answering the aforementioned research questions with both qualitative and quantitative information, this master's thesis and the associated MIDDEN report will provide the Dutch government and plastics industry with the necessary data and tools to improve the decision-making process, thus further helping to achieve the national and European goals of the Netherlands to tackle climate change and reduce the environmental impact of plastics production. Additionally, the methodological approach of this study may serve as a model for future research in other countries and/or sectors, possibly facilitating scientific findings and decarbonisation roadmaps.

2. BACKGROUND INFORMATION

2.1 Plastics types

Plastics (from the Greek word "plastikos", meaning fit for moulding) is the term used to describe the wide range of synthetic materials used in a wide and growing range of industrial and consumer applications, from packaging and clothing to construction and electronics. Plastics are extremely versatile materials with lightweight, thermal and electrical insulation, and resistance to corrosion. Their characteristic of being easily moulded into complex shapes allow them to be integrated into composite products, and the aforementioned properties can be enhanced or modified with additives (e.g. flame retardants, colours, reinforcing fillers, and foaming agents) to meet the demands of specific applications (PlasticsEurope, 2020a).

Plastics can be divided into two major categories: thermoplastics and thermosets. The first group has a simple structure made by chemically independent macromolecules then melt upon heating and can then shaped, welded, and solidified after being cooled; this process can be repeated multiple times, allowing thermoplastics to be reprocessed and recycled. The second group is made by crosslinking polymer chains and is thus generally stronger than thermoplastic materials, but once hardened it cannot be reshaped. As seen in Figure 1, examples of thermosets include PUR (used for insulating foams, flooring, mattresses, etc) and epoxy resins (used for protective coatings and sealings, casting and electronics encapsulation, adhesives, etc).



Figure 1: Main categories of plastics. Compiled from a collection of sources.

2.2 European plastics market

In 2018, plastic production reached 360 million tonnes globally and 60 million tonnes in Europe, of which almost 80% was consumed by the six largest European countries (Germany, Italy, France, Spain, United Kingdom, and Poland) and the Benelux countries (Belgium, Netherlands, and Luxembourg) (PlasticsEurope, 2019). Despite the existence of dozens of plastic types, Figure 2 shows how almost half of the European consumptions consists just of polyethylene (PE) and polypropylene (PP).



Figure 2: Distribution of European plastic demand in 2018, based on data from (PlasticsEurope, 2019).

Overall, the European manufacturing and processing plastics industry had a turnover of more than 360 billion euros with a positive trade balance of more than 15 billion euros with extra-European countries, thus being the 7th most important sector for industrial value added (PlasticsEurope, 2019).

2.3 Development history of polyethylene (PE)

Polyethylene was first synthesized in 1930 by Professor Carl Shipp Marvel after an experiment involving ethylene gas and an organometallic catalyst at high temperatures, but DuPont, the company Marvel was working for, failed to realise the commercial possibilities of the product (Hutley & Ouederni, 2016). In 1933, another experiment by Eric William Fawcett and Reginald Oswald Gibson of the Imperial Chemical Industries (ICI) Research Laboratory led to the high-pressure polymerization of what is now called low-density polyethylene (LDPE) (Flavell-While, 2020). The process was patented by ICI in 1937 and in the following years many PE manufacturing plants were built to satisfy the increasing demand of telegraph cable insulation and, later, the high-frequency radar cables used during World War II by the Allied forces. LDPE is a very flexible and durable plastic, and nowadays it is mostly used to manufacture reusable food containers and trays, disposable shopping bags and packaging, squeezable bottles and cling-film (A&C Plastics, 2020). The process to produce high-density polyethylene (HDPE) was then discovered in 1953 by researchers Hogan and Banks of Phillips Petroleum Company using an innovative chromium oxide catalyst. Although the process was patented and manufacturing plants were built, the product did not match with the market needs and the company faced some financial problems, until it partnered with the Wham-O Company to make over 100 million HulaHoops with HDPE tubes (Hutley & Ouederni, 2016). The catalyst and the process were then licenced to other manufacturers, the market evolved, and HDPE became one of the most used polymers in the world for reusable bags and bottles, toys, pipes for water and sewage, folding chairs and houseware (A&C Plastics, 2020). Finally, the patent for linear low-density polyethylene (LLDPE) was filed by the DuPont Company in 1957, but the production remained low until in 1978 the Union Carbide Company announced their Unipol process and coined the name of the product, which is now the main choice for packaging film, especially in the food sector (Hutley & Ouederni, 2016). As shown in Figure 3, the three major types of polyethylene (LDPE, HDPE, and LLDPE) currently form a market of over 100 million tons and a value of over 180 billion dollars.



Figure 3:Global polyethylene production in kt (Hutley & Ouederni, 2016).

The milestones of polyethylene development history can be summarized as follows:

- 1937: First commercialization of LDPE
- 1953: First commercialization of HDPE
- 1975: PE plastics reach a global production of 10,000 tons
- 1978: First commercialization of LLDPE
- 2000: PE plastics reach a global production of 50,000 tons
- 2018: PE plastics reach a global production of 100,000 tons

2.4 Development history of polypropylene (PP)

In 1953, Professor Karl Ziegler and researcher Heinz Breil of the Max-Planck Institute discovered an innovative method to prepare high-density PE using a new organo-metallic catalyst, and adopted an aggressive licensing approach, earning enough money to sustain the Max-Planck Institute budget for the next 40 years (Martin, 2007). After hearing one of Ziegler lectures, Professor Giulio Natta and his team carried out experiments with the new catalyst and using propylene as a feedstock, and in 1954 managed to produce polypropylene (PP), a result that Ziegler's team had not yet achieved at the time (Hutley & Ouederni, 2016). For the discovery of the catalyst, and for its application to produce PP, both Ziegler and Natta won a Nobel Prize in Chemistry in 1963. Polypropylene was first made in 1951 by Hogan and Banks of Phillips Petroleum while experimenting on the chromium catalyst for HDPE, but due to legal procedures with the US Patent Office, their patent was issued only in 1983 (Hammond &

Saunders, 1991). PP is flexible, durable, and resistant to heat and acids, and it thus the plastic of choice for food containers (e.g. Tupperware), laboratory equipment, and automotive components (A&C Plastics, 2020). As seen in Figure 4, PP currently forms a market of almost 90 million ton and over 130 billion dollars.



Figure 4: Global polypropylene production in kt (Hutley & Ouederni, 2016).

The milestones of polypropylene development history can be summarized as follows:

- 1954: First commercialization of PP
- 1987: PP plastics reach a global production of 10,000 tons
- 2010: PP plastics reach a global production of 50,000 tons
- 2018: PP plastics reach a global production of 90,000 tons

3. METHODOLOGY

A general and summarized overview of the research steps can be seen in Figure 5, linking together questions, methods, and expected results.



Figure 5: Research framework showing the connection between questions, methodologies, and results.

The following sections will present in detail the specific methodologies used to gather and process the data relative to each research question.

3.1 Market analysis of Polyolefin products and applications

The first part of the research was dedicated to answering the question: "What are the main applications of polyolefins, and what is the volume demand of each consumer sector in the Netherlands?".

The first part of the question was answered by reviewing a huge amount of scientific literature, reports, websites, and other online sources regarding polyolefin products and applications. The description of each polyolefin was coupled with an overview of its physical properties, namely:

- **Density**: the ratio between the mass and the volume of the material, it is measured in g/cm³ or kg/m³.
- **Tensile yield strength**: the stress a material can withstand without permanent deformation, it is measured in MPa or N/mm².
- Shore hardness (D scale): it measures the resistance of a material to be penetrated by an indenter, it goes from 0 (softest) to 100 (hardest).
- **Continuous service temperature**: the maximum temperature (measured in Celsius) above which the mechanical properties of the material degrade significantly.

Finding the volume demand of each Dutch consumer sector, however, was more complicated. The main sources for information were the reports prepared by PlasticsEurope, the leading association of plastic manufacturers in Europe. Having more than 100 member companies, which are responsible for more than 90% of the polymers produced in the European Union (PlasticsEurope, 2020b), PlasticsEurope was considered a reliable source to get data regarding the use of polyolefins. Nevertheless, although the organisation gathers and publishes many facts and graphs about plastic production and consumption, it does not share the exact numbers to the public. To get the values regarding the use of polyolefin products in the main economic sectors of the Netherlands, it was thus necessary to use some creativity and connect the clues.

As seen in Figure 6, the graph presented by PlasticsEurope offers a visual overview of the consumption of many polymer types (including polyolefins) by different economic sectors. The only explicit numbers, however, are the total shares of each sector, without a distinction per polymer type. Using only the information of this graph it was not possible, for example, to know the amount of polypropylene (PP) used by the automotive sector or the amount of High-density Polyethylene (HDPE) used for building and constructions.



Figure 6: Plastics demand by segments and polymer types in 2018 (PlasticsEurope, 2019).

It was thus necessary to extract these values from the graph. To convert the visual data in actual numbers, the free software Web Plot Digitizer was used to measure the area of each sphere in pixels, as seen in Figure 7.



Figure 7: Screenshot of the measuring process using Web Plot Digitizer. The areas of the circles were measured in pixels.

The process was applied for all the polyolefin resin types, which in the case of polyethylene were then renamed to be coherent with the framework of this thesis. Namely "PE-LD/PE-LLD" was renamed as "LDPE/LLDPE" and PE-HD/PE-MD was renamed as "HDPE". The following step was to convert the measured amounts from pixels to percentage values. Knowing that the total polymer demand share in 2018 was 17.5% for LDPE, 12.2% for HDPE, and 19.3% for PP (PlasticsEurope, 2019), it was possible to calculate the ratio between the total amount of pixels and the polymer share and use the following formula to calculate the percentage values.

$$Share_{i,j} = \frac{Pixels_{i,j}}{Pixels_{i,tot}/Share_{i,tot}}$$
[%]

Where "i" is the product (e.g. HDPE) and "j" is the consumer sector (e.g. automotive).

For lack of better information, it was assumed that the Dutch society and economy are very similar to the European average, and it was known that the yearly demand for plastics in the Netherlands is equal to 2,200 kt (PlasticsEurope, 2019). As a final step, the following formula was applied, making it possible to create a treemap graph for each of the consumer sector.

$$Demand_{i,i} = Share_{i,i} \cdot Yearly Dutch Demand$$
 [kt]

Where "i" is the product (e.g. LLDPE) and "j" is the consumer sector (e.g. construction).

3.2 Investigation of Polyolefin producers in the Netherlands

The second part of the research was dedicated to answering the question: "Who are the main producers of polyolefins in the Netherlands, and what are their production volumes, energy use, and carbon emissions?". The list of producers falling inside the research boundaries of the MIDDEN project was provided by TNO and PBL and consisted in Dow Chemical Company, SABIC Limburg, and Ducor Petrochemicals. Information regarding these three companies was then found on their websites, leading to the localisation of the active polyolefin production plants in The Netherlands.

As Dow and SABIC are part of the European Emissions Trading Scheme (EU ETS), their carbon emissions are reported to the Dutch Emission Authority (Nederlandse Emissieautoriteit, or NEa). For these two producers, it was thus possible to retrieve the emissions of CO_2 for 2019. As a replacement for the NEa registry, the carbon emissions for Ducor were obtained from the emission registry managed by the National Institute for Public Health and the Environment (RIVM). It is important to remember, however, that the emissions declared to NEa and RIVM are the total sum of greenhouse gases directly emitted by all the processes of the production site, making it difficult to isolate the share relative to the polymerisation only.

Finally, to obtain the production volumes and the energy consumption it was necessary to get in direct contact with the manufacturers. The communication consisted of multiple phone calls, emails, online meetings, and was concluded with a survey filled in by the companies' representatives and covered by confidentiality. Besides providing valuable data, the manufacturers also verified or corrected the information previously collected on websites and public reports.

All the collected knowledge about the polyolefin producers has become part of the official MIDDEN dataset managed by PBL, which explicitly lists the following indicators:

- Plant.
- Year.
- Corporate group.
- Number of employees.
- Street address.
- Postal code.
- Town/location.
- Website.
- Permit number (NEa).
- Name of production site (NEa).
- Infrastructure electricity [GWh/year].
- Infrastructure heat [TJ/year].
- Infrastructure feedstock [kt/year].

An additional sheet of the MIDDEN dataset is reserved for the plant configuration data. One column was created for each different production line (e.g. LLDPE by Dow, HDPE by SABIC, PP by SABIC, etc), explicitly listing the following indicators:

- Plant.
- Configuration.
- Year.
- Section.
- Technology.
- Capacity [kt/year].
- Capacity utilisation [%].
- Year of next investment decision.
- Infrastructure electricity [GWh/year].
- Infrastructure heat [TJ/year].

Although the MIDDEN dataset is transparent about the sources of the data, the final report published by PBL, as well as this thesis, had to hide some information due to confidentiality reasons. Therefore, the result section 4.2 will show the aggregate annual values of production volume of polyolefins, electricity consumption, heat consumption, and carbon emissions in the Netherlands, without specifying the relative contribution of each polyolefins producer.

3.3 Techno-environmental analysis of Polymerisation processes

The third part of the research was dedicated to answering the question: "What are the main polymerisation processes, and what are their associated costs, energy and material use, and CO_2 emissions?".

The main sources were the Best Available Techniques (BAT) Reference Documents (BREF) prepared by the European IPPC Bureau at the Institute for Prospective Technological Studies (European Commission, 2007), the Eco-profiles reports on ethylene and propylene (PlasticsEurope, 2012, 2014b), and the interviews with the polyolefin producers involved in the research (Dow, SABIC, Ducor), especially regarding the costs.

All the collected information about the polymerisation processes has become part of the MIDDEN dataset, which explicitly lists the following indicators:

- Technology.
- Year.
- Technology description.
- Capacity [kt/year].
- Investment per unit of capacity [€].
- Annual O&M costs per unit of capacity [€].
- Technical lifetime.
- Electricity per unit of capacity [TJ].
- Heat per unit of capacity [TJ].
- Feedstock per unit of capacity [kt].
- Output per unit of capacity [kt].
- CO₂-eq emissions per unit of capacity (polymerisation) [kt].
- CO₂-eq emissions per unit of capacity (cradle-to-gate) [kt].

To better understand the relative importance of the polymerisation process, data regarding the emissions of CO₂-eq has been separated into "polymerisation only" and "cradle-to-gate", which includes the extraction of raw materials (e.g. oil) and the conversion into feedstock (i.e. ethylene).

As in the case of production volumes and energy consumption, all the collected data is stored in the MIDDEN dataset delivered to PBL, while only the average values will be displayed in this thesis due to confidentiality reasons.

3.4 Techno-economic analysis of Decarbonisation options

The fourth part of the research was dedicated to answering the question: "Which available decarbonisation options are considered as most promising by the stakeholders, and to what extent are they technically, economically, and environmentally comparable to the current technologies?".

After the first discussion with experts from TNO and PBL, it was decided that this MIDDEN report would have been dedicated to the analysis of sustainable bio-based processes and circular economy options, specifically innovative recycling technologies. As explained in previous sections, the majority of the GHG emitted by the polyolefins industry have an indirect nature (downstream and upstream) and are not directly caused by the polymerisation processes. Therefore, the most promising decarbonisation

options are the ones targeting the whole life-cycle and providing alternatives to the production of fossil-based feedstock.

This choice was also motivated by communication with relevant stakeholders (such as the Chemelot Industrial Park in Limburg) and by the two following reports:

- VNCI, the Dutch association of chemical producers, analysed possible decarbonisation pathways to 2050, with a focus on bio-based materials and circular economy, showing that it is technically possible to achieve a reduction of GHG emissions while keeping an annual growth rate of added value (VNCI, 2018).
- McKinsey, an international consultancy company, published a report on decarbonisation options for a variety of industrial sectors. In the chapter regarding ethylene, bio-based feedstock technologies were described as promising decarbonisation solutions (McKinsey&Company, 2018).

A quick search on Google Scholar and Science Direct also proved the presence of pilot projects, life cycle analysis, and scientific literature to supply the data necessary for this research, thus supporting the choice of focusing on bio-based and recycling technologies.

For completeness, other available decarbonisation options were to be listed with a short description, but without an extensive analysis of economic and environmental indicators. Having defined the scope, an extensive review of scientific and grey literature, technical reports, case studies, decarbonisation strategies written by industry organisations and/or public institutions, was carried out. All the collected information was then discussed with the representatives of the involved polyolefins producers (Dow, SABIC, Ducor), with the MIDDEN colleagues from TNO and PBL, and with other stakeholders (such as the business developers of the Chemelot Industrial Park in Limburg).

All the collected information about the decarbonisation options has become part of the MIDDEN dataset, which explicitly lists the following indicators (if available):

- Technology.
- Year.
- Technology description.
- Capacity [kt/year].
- Investment per unit of capacity [€].
- Annual O&M costs per unit of capacity [€].
- Technical lifetime.
- Electricity per unit of capacity [TJ].
- Heat per unit of capacity [TJ].
- Feedstock per unit of capacity [kt].
- Output per unit of capacity [kt].
- CO₂-eq emissions per unit of capacity (polymerisation) [kt].
- CO₂-eq emissions per unit of capacity (cradle-to-gate) [kt].

For bio-based decarbonisation options, some data processing was also necessary. As most of the information about the bio-ethanol and bio-methanol production processes comes from Brazil, it was necessary to transform the data to make it coherent with the Dutch and European techno-economic situation. Besides making the conversion of all costs from US dollars to euros, it was necessary to increase the levelized cost of bio-ethylene according to the price difference between Brazilian and European feedstock. A scientific paper shows that 70% of the bio-ethylene cost is due to feedstock

costs (Oliveira et al., 2020), and the current price of ethanol in Europe is 50% higher than the same product in Brazil (GlobalPetrolPrices, 2020). Therefore, the price was adjusted as it follows:

$$LC_{bio-ethylene,EU} = LC_{bio-ethylene,BR} \cdot (1 + 0.7 \cdot 0.5) \qquad \left[\frac{\epsilon}{t \ ethylene}\right]$$

Regarding the environmental impact, two cases were considered: a best-case in which all the carbon dioxide absorbed by the plants is permanently sequestered from the atmosphere (only possible if 100% of the bio-polyolefins are used for long-use objects and/or get fully recycled), and a more realistic case in which only 40% of the bio-polyolefins stay in circulation (thus preventing 40% of the absorbed carbon to be released again in the atmosphere). This value was chosen because the current plastic recycling rate in the Netherlands is 36%, and the share is likely to increase in the following years thanks to improvements in the collection system and innovations of recycling technologies. The following formulas were applied:

$$GHG_{best-case} = GHG_{process} - GHG_{absorbed} \qquad \left[\frac{t \ CO_2 eq}{t \ ethylene}\right]$$
$$GHG_{40\%-case} = GHG_{process} - 0.4 \cdot GHG_{absorbed} \qquad \left[\frac{t \ CO_2 eq}{t \ ethylene}\right]$$

Finally, a break-even carbon price was calculated for both cases, showing the value necessary to make bio-based polyolefins as economically convenient as their fossil counterparts.

$$Carbon \, price_{break-even} = \frac{LC_{fossil} - LC_{biobased}}{GHG_{biobased} - GHG_{fossil}} \qquad \left[\frac{\notin}{t \, CO_2 eq}\right]$$

All the indicators were then collected together in a table to make the comparison with fossil-based polyolefins easier. A calculation was also made to show how much land would be required if the Netherlands were to produce all the bio-PE starting from Dutch sugar beets:

Share of arable land =
$$\frac{kt PE \cdot Ml of ethanol per kt PE \cdot Land per Ml of ethanol}{Dutch arable land}$$

4. RESULTS

4.1 Polyolefin products and applications

This section answers the first research question: "What are the main applications of polyolefins, and what is the volume demand of each consumer sector in the Netherlands?". In the Netherlands, the annual demand for plastics is equal to 2.2 million tons, of which about half are polyolefins such as LDPE, LLDPE, HDPE, and PP (PlasticsEurope, 2019). Packaging is the biggest sector overall, and although plastics only account for 18% of all packaging materials (by weight) in the Netherlands, it makes up 43% of the total revenue, which was over 6 billion euros in 2016 (ABN AMRO, 2017). The following sub-section introduces the main characteristics and applications of each polyolefin, and present treemap graphs depicting the volume demand of the main Dutch consumer sectors in 2018, also providing an approximation of the market value.

4.2.1 Low-density Polyethylene

Low-Density Polyethylene (LDPE) is a flexible and translucent plastic with good resistance to electricity and chemicals like alcohols, acids, and dilute alkalis (Omnexus, 2020a). Table 1 shows an overview of its physical properties.

Table 1: Density, tensile yield strength, shore hardness and maximum continuous service temperature (CST) of low-density polyethylene (Omnexus, 2020b).

Density	Tensile yield strength	Shore hardness	Max CST
(g/cm³)	(MPa)	(D scale)	(°C)
0.92-0.94	10-20	40-50	80-100

Although it is susceptible to stress cracking and it cannot sustain high temperatures, its low cost, lightweight and transparency make LDPE a very good material to manufacture containers and bags by using extrusion or injection moulding (Omnexus, 2020a). A practical application of LDPE is shown in Figure 8.



Figure 8: Blown film extruded low-density polyethylene bags (LDPE Bags, 2020).

Linear low-Density Polyethylene (LLDPE) is a variant of LDPE, being flexible, light, and translucent, but having higher physical and chemical resistance, as shown in Table 15.

Table 2: Density, tensile yield strength, shore hardness and maximum continuous service temperature (CST) of linear low-density polyethylene (Omnexus, 2020b).

Density	Tensile yield strength	Shore hardness	Max CST
(g/cm³)	(MPa)	(D scale)	(°C)
0.92-0.95	10-30	55-65	90-110

Just like LDPE, LLDPE is lightweight and has a low production cost. The most common manufacturing process in extrusion (e.g. for blow and cast films), but injection and roto-moulding can also be used (Omnexus, 2020a). Moreover, LLDPE sheets are physiologically harmless, making it the perfect material for food packaging and agricultural film, as shown in Figure 9.



Figure 9: Linear low-density polyethylene agricultural film (Mulch Film, 2020).

LDPE and LLDPE together make 17.5% of the plastics demand in Europe, for a total of 8.96 million tonnes (PlasticsEurope, 2019). Assuming the ratio is the same in the Netherlands, the volume of LDPE and LLDPE would be equal to 385,000 tonnes. Considering a price between 0.98 and 1.08 €/kg (PlasticPortal, 2020), the Dutch market would be worth 375-515 million euros per year.

LDPE and LLDPE are used especially in the packaging sector (single-use food film, reusable bags and containers), agriculture sector (single-use agricultural film), building and construction sector, and are marginally used in the electrical and automotive sectors (PlasticsEurope, 2019). The shares of LDPE/LLDPE for the main consumer sectors in The Netherlands are shown in Figure 10.



Figure 10: Market demand (in kt/y) of LDPE/LLDPE used by the main consumer sectors in The Netherlands in 2018. Made with estimates from European data extracted from (PlasticsEurope, 2019).

4.2.2 High-density Polyethylene

High-density polyethylene (HDPE) is a flexible and translucent plastic with excellent resistance to electricity and solvents, and good resistance to heat, alcohols, dilute acids and alkalis (Omnexus, 2020a). Table 3 shows an overview of its physical properties.

Table 3: Density, tensile yield strength, shore hardness and maximum continuous service temperature (CST) of high-density polyethylene (Omnexus, 2020b).

Density	Tensile yield strength	Shore hardness	Max CST
(g/cm ³)	(MPa)	(D scale)	(°C)
0.94-0.97	25-30	60-70	100-120

HDPE is usually processed by injection moulding, extrusion, roto-moulding, and blow moulding. This grade of PE is heavier, sturdier, harder and can resist to higher temperatures than both LDPE and LLDPE, making it suitable for rigid packaging, household and consumer goods, fibres, textiles, pipes and fittings (Omnexus, 2020a). Practical applications of HDPE are shown in Figure 11 and Figure 12.



Figure 11: Example of blow moulded high-density polyethylene bottles (Indiamart, 2020).



Figure 12: Example of high-density polyethylene corrosion-resistant pipes (Acme Plastics, 2020).

HDPE makes 12.2% of the plastics demand in Europe, for a total of 6.25 million tonnes (PlasticsEurope, 2019). Assuming the ratio is the same in the Netherlands, the volume of HDPE would be equal to 268,400 tonnes. Considering a price range between 1.05 and 1.08 €/kg (PlasticPortal, 2020), the Dutch market would be worth 280-290 million euros per year.

HDPE is used especially in the packaging sector (milk bottles and shampoo bottles), for houseware and toys, and in the building and construction sector to make pipes (PlasticsEurope, 2019). The shares of HDPE in each sector are shown in Figure 13.



Figure 13: Market demand (in kt/y) of HDPE used by the main consumer sectors in The Netherlands in 2018. Made with estimates from European data extracted from (PlasticsEurope, 2019).

4.2.3 Polypropylene

Polypropylene (PP) is a semi-rigid and translucent plastic with high resistance to heat, fatigue, electricity and chemicals like alcohols, acids, alkalis and oils (BPF, 2020). These properties can be altered by adding additives or by changing the share of co-monomers during the polymerisation process, making PP a very versatile material with a vast range of applications. The PP homopolymer contains only propylene and is a general-purpose grade plastic, while the PP Random Copolymer features ethene units (up to 6% by mass) randomly incorporated into the polymer chain, making it very flexible and optically clear, and the PP Block Copolymer incorporates ethene (5-15% by mass) arranged in a regular pattern, making the polymer less brittle. Finally, the PP Impact Copolymer also incorporates randomly ethylene, reaching a share of 45-65% and thus making it lighter while providing good resistance to impact (Omnexus, 2020c). Table 4 shows an overview of its physical properties.

Table 4: Density, tensile yield strength, shore hardness and maximum continuous service temperature (CST) of polypropylene (Omnexus, 2020b).

Grade	Density (g/cm³)	Tensile yield strength (MPa)	Shore hardness (D scale)	Max CST (°C)
Homopolymer	0.90-0.91	35-40	70-83	100-130
Copolymer	0.90-0.91	20-35	70-80	100-130
Impact modified	0.88-0.91	11-28	45-55	90-115

Polypropylene is harder and more resistant to heat than most of the polyethylene compounds. Moreover, the tensile strength of PP makes it the perfect material to manufacture living hinges, i.e. thin and flexible connections between two relatively rigid section, potentially being able to be flexed more than 900,000 times without breaking (MIT-Edu, 2020). A practical application of a PP living hinge can be seen in Figure 14.



Figure 14: Example of a polypropylene butterfly living hinge used on the dispensing cap of a ketchup bottle (Davis, 2015).

PP makes 19.3% of the plastics demand in Europe, for a total of 9.88 million tonnes (PlasticsEurope, 2019). Assuming the ratio is the same in the Netherlands, the volume of PP would be equal to 424,600 tonnes. Considering a price range between 1.08 and 1.21 €/kg (PlasticPortal, 2020), the Dutch market would be worth 450-510 million euros per year.

PP is used especially in the packaging sector for flexible packaging (e.g. snack wrappers, thin films for clothing and tobacco) and rigid packaging (e.g. stackable crates, detergent and condiments bottles), in the household sector for reusable items (e.g. microwave food containers, toys, and gardening tools), in the building and construction sector (e.g. pipes), and has many applications in the automotive sector (e.g. interiors and bumpers) (BPF, 2020; PlasticsEurope, 2019). The shares of PP in each sector are shown in Figure 15.



Figure 15: Market demand (in kt/y) of PP used by the main consumer sectors in The Netherlands in 2018. Made with estimates from European data extracted from (PlasticsEurope, 2019).

4.2 Polyolefin producers in the Netherlands

This section answers the second research question: "Who are the main producers of polyolefins in the Netherlands, and what are their production volumes, energy use, and carbon emissions?". As explained in the Methodology chapter, Table 5 only shows the aggregated values for the Netherlands, without explicating the contribution of each company due to confidentiality reasons.

Table 5: Aggregated capacity, electricity consumption and heat consumption from p	polyolefins production in the Netherlands
in 2019 (Confidential data from Dow, SABIC and Ducor, 2020).	

Polyolefin product	Nominal capacity [kt/y]	Electricity consumption [GWh/y]	Heat consumption [TJ/y]
LDPE	730	678	-645
LLDPE	800	282	552
HDPE	310	105	450
РР	730	253	286
Total	2,570	1,318	643

As explained in the methodology, it is difficult to quantify the GHG emissions for polyolefins production. If only the polymerisation process is considered, the aggregate emissions of the three companies are calculated as 794 kt CO₂-eq, while using the cradle-to-gate method (which includes the emissions caused while producing the feedstock necessary to manufacture polyolefins) the value reaches 4,495 kt CO₂-eq.

To understand the scale of the values shown in Table 5, they represent 8.5% of the polyolefins produced (30,282 kt/y) in Europe (PlasticsEurope, 2019), and 1.2% of the electricity consumed (113,382 GWh/y) in the Netherlands (CBS, 2020a), while the GHG emitted by the whole Dutch industrial sector are 34,984 kt CO_2 -eq/year (CBS, 2020b).

4.2.1 Dow Chemical Company

Dow Chemical Company (from now on, Dow) is the third-largest chemical producer in the world (C&EN, 2019), with headquarters in the United States of America. Dow was founded in 1897 by chemist Herbert Henry Dow and always had the tradition of diversifying its product line, ranging from agricultural chemicals and plastics resins, to plutonium during World War II and napalm during the Vietnam War. In 2011, Dow divested its global polypropylene business to Braskem, the largest petrochemical company in Latin America, in order to better focus on improving the performance of its polyethylene production (Business Wire, 2011). In 2017, Dow merged with DuPont, becoming the largest chemical producer in the world, but two years later the company was reorganised and split into three separate publicly traded companies focusing on materials science (Dow Inc.), agriculture (Corteva), and speciality products (DuPont) (C&EN, 2019).

Dow is present in Europe since 1955 and in the Netherlands since 1964, when it opened its first factory in Terneuzen. The industrial cluster in the province of Zeeland also houses other chemical companies, like Yara and Arkema, and good transport connections via land and water (VNCI, 2018). The company expanded in the last decades, and Terneuzen is currently the second-largest Dow production site in the world (Dow, 2020b), with a yearly turnover of more than 2 billion euros (Dow, 2017). For what

concerns polyethylene production, the site hosts a production line for LDPE, built between 1968 and 1975, and three production lines for LLDPE, built in 1980, 1986 and 2000 (*Internal communication with Dow Terneuzen, 2020*), as seen in Figure 16.



Figure 16: Aerial view of Dow chemical plants in the Terneuzen site (Dow, 2020b).

Electricity and steam are supplied to Dow by the "Electricity and Steam Association" (ELSTA) near Terneuzen, which has been owned by Dow since 2018. The cogeneration facility is equipped with one steam turbine and three gas-fired turbines with a connected heat recovery boiler, and can generate a total of 460 MW of electricity and 850 ton/hour of steam at a pressure of 90 bar, supplying both Dow and the public grid (ELSTA, 2020).

4.2.2 SABIC

SABIC is the fourth largest chemical producer in the world (C&EN, 2019), and the second-largest public company in the Middle East and in Saudi Arabia, where its headquarters are based. SABIC was founded in 1976 under the will of the Saudi government to covert the oil into polymers and other chemicals, and later expanded in the fertilizers and metals subsectors. In 2019, 70% of SABIC's shares were acquired by Saudi Aramco, the state-owned energy company of Saudi Arabia (SUSTG, 2019). In 2002, SABIC acquired several factories from the Dutch petrochemical company DSM, officially starting operations in Europe (Kapner, 2002). The company is present in the Netherlands through its subsidiaries SABIC Europe, SABIC Limburg B.V., SABIC Petrochemicals B.V., and SABIC Innovative Plastics B.V. (SABIC Limburg, 2020). SABIC is part of the Chemelot cluster in Geleen, an industrial area characterized by a strong integration of utilities and services, grouping more than 50 factories and R&D facilities (VNCI, 2018). SABIC infrastructures constitute about half of the space at Chemelot, including two naphtha crackers (NAK3 and NAK4) for the production of plastic monomers, three LDPE production

lines (Id-PE), two HDPE production lines (hd-PE), and two PP reactors (*Internal communication with SABIC, 2020*), as shown in Figure 17.



Figure 17: Aerial view of SABIC chemical plants at Chemelot industrial park, showing the naphtha crackers NAK3 and NAK4, and the production lines of PP, HDPE and LDPE (SABIC Limburg, 2020).

Electricity, steam and technical gases are supplied by Utility Support Group (USG), a joint venture between SABIC and Sitech Utility Holding CV with approximately 200 employees (USG, 2020a). USG supplies many companies within the Chemelot industrial park thanks to its 150 MW cogeneration plant, plus other 90 MW purchased externally. USG also collects around 620 tons/hour of waste steam produced by the factories and produces itself an additional 330 tons/hour to satisfy the steam demand of the companies (USG, 2020b).

4.2.3 Ducor Petrochemicals

The polypropylene manufacturing facilities in Rozenburg were built in 1979 by Basell, but in 2007 the company merged with Lyondell and the agreement involved some capacity divestment requirement (ChemEurope, 2001). The Rozenburg plant was acquired by DOMO, a joint venture between the Belgian Domo Group and Carmel Olefins, a subsidiary of BAZAN Group, the biggest petrochemical company of Israel. The joint venture changed name in 2011, becoming Ducor Petrochemicals (PlastEurope, 2011). The manufacturing site in Rozenburg operates within the boundaries of Rotterdam-Rijnmond, the largest chemical and petrochemical cluster in the Netherlands, which is characterised by excellent infrastructures and connections, i.e. roads, pipelines and railways (VNCI, 2018). Ducor is also part of "CIRCLE", a consortium of international plastic companies and academia aiming to reduce industrial waste and increase the amount of recycled material in plastic products (Ducor, 2020b).

Ducor Rozenburg has 80 employees (Port of Rotterdam, 2016) and three production lines that can produce different PP resins at the same time or be combined to bear big production loads in a short

time. The strategic location into the logistic hub of the Rotterdam port also means that Ducor is connected with an efficient system of pipelines for the supply of utilities and feedstock (Ducor, 2018). Part of the infrastructures of Ducor Rozenburg can be seen in Figure 18.



Figure 18: View of Ducor chemical plants in the Rozenburg site (Ducor, 2020b).

Utilities such as electricity, steam and industrial gases are supplied to Ducor by Air Liquide, which is also part of the Rotterdam Port industrial cluster and adopt gas-fired cogeneration turbines.

4.3 Polymerisation processes

The monomers necessary for polyolefin production, namely ethylene (also called ethene, chemical formula: C_2H_4) and propylene (also called propene, chemical formula: C_3H_6), are produced by steam crackers. In the European Union, around 63% of the cracker feedstock is made up by naphtha coming from petroleum refineries, with the remaining share being LPG and other natural gas liquids (Petrochemicals Europe, 2020). In the Netherlands, between 5,000 and 6,000 ktoe of naphtha are produced every year (IEA, 2020), accounting for around 10% of the cracker feedstock in the European Union (Petrochemicals Europe, 2020).

During the chain polymerisation process, the chemical bonds of thousands of monomers are forced to open and link to each other, creating a saturated chain macromolecule whose properties can vary depending on the polymerisation process and technique (European Commission, 2007). As seen in Table 6, four main polymerisation process technologies are applied in the production of polyolefins, and all of them will be described and analysed in the following sections.

Table 6: Most common processes applied in polyethylene and polypropylene production. Made with information from (European Commission, 2007; PlasticsEurope, 2014a, 2014b).

POLYMERISATION PROCESS	LDPE	LLDPE	HDPE	РР
High-pressure	Х			
Solution		Х	Х	
Suspension/slurry			х	х
Gas-phase		х	х	х

At Dow, LLDPE is produced under the brand name of Dowlex with a double reactor solution process (Townsend Solutions, 2017), while HDPE is produced under the brand name of Continuum with UNIPOL II process technology, which corresponds to a double reactor gas-phase process (Dow, 2020a). At SABIC, LDPE is produced by a high-pressure tubular reactor, HDPE is produced using a slurry process with loop reactor, while PP is produced using a gas phase polymerisation process (*Internal communication with SABIC, 2020*). At Ducor, PP is produced under the brand names DuPure and DuClear using Novolen technology (Ducor, 2020a), which employs a stirred gas phase polymerisation reactor and a nitrogen degassing vessel (McDermott, 2020).

4.3.1 High-pressure polymerisation

Low-Density Polyethylene (LDPE) is a low crystalline and high amorphous polymer with an average density of 0.91-0.94 g/cm³ and a melting point of 105-115°C. It is composed of 4,000-40,000 carbon atoms with a high degree of short and long side-chain branching (Omnexus, 2020a), as shown in Figure 19.



Figure 19: LDPE structure with the main carbon chain in black and the side branches in blue and red (Graziano et al., 2019).

LDPE is produced through high-pressure polymerisation, which is a very standardized process following the steps shown in Figure 20.



Figure 20: LDPE high-pressure polymerisation process scheme. Made with information from (European Commission, 2007; PlasticsEurope, 2014a; SABIC Limburg, 2020).

After compressing the gaseous ethylene to 20-30 MPa together with the unreacted gas of previous cycles, the polymerisation takes place in an autoclave reactor, operating with a temperature of 180-300°C and pressure of 100-250 MPa, or in a tubular reactor, operating with a temperature of 140-340°C and a pressure of 200-350 MPa (European Commission, 2007). LDPE polymerisation usually requires the use of initiators, namely oxygen or organic peroxides with a mass fraction concentration of 0.1-0.5%, and sometimes the use of polar modifiers (e.g. ketones) or aliphatic hydrocarbons to influence the molecular weight distribution (PlasticsEurope, 2014a). The resulting polymer and unreacted gases are then separated in a high-pressure separator (15-30 MPa) and low-pressure separator (0.15 MPa), where the unreacted gases are recycled back into the process and part of the residual heat is recovered and used to produce steam (European Commission, 2007). As the polymerisation of ethylene is a highly exothermic process (3300-3600 J/g), the manufacture of LDPE is a net steam producer (Burdett & Eisinger, 2016). If required by the application of the product, specific additives (e.g. stabilizers or fire retardants) are added to the polymer melt to obtain the desired properties, and the polyethylene is then extruded into granules. Finally, the LDPE is dried, blended, and degassed to remove the residual ethylene before being transported to storage silos or packed and sent to the costumers (European Commission, 2007).

In 1999, the European average energy demand for LDPE production was equal to 3.9 MJ/kg of direct energy, or 9.4 MJ/kg of primary energy (assuming an efficiency of 40% for electricity and 90% for steam), while the average of the top 50% companies was equal to 2.6 MJ/kg of direct energy, or 7.5 MJ/kg of primary energy (European Commission, 2007). More than one decade later, the values are not so different, as in 2014 the production of LDPE still required an average of 7.9 MJ/kg of primary energy (assuming an efficiency of 40% for electricity and 90% for steam, see Appendix A). As shown in Table 7, the polymerisation of 1 kg of LDPE causes the emission of 0.32 kg of CO₂-eq, considering the Global Warming Potential (GWP) for 100 years.

Table 7: Energy demand and global warming potential (GWP) for the polymerisation of 1 kg of LDPE. Average values from literature (European Commission, 2007; PlasticsEurope, 2014b) *and internal communication with the companies involved.*

Product [1 kg]	Electricity demand [MJ]	Steam demand [MJ]	GWP [kg CO2-eq]
LDPE	3.45	-0.67	0.32

As explained before, the value for steam demand is negative because LDPE polymerisation is a net producer of heat, which is recovered and used for other processes (European Commission, 2007). If the boundaries of the life-cycle analysis are extended to the extraction of fossil and mineral resources, the production of raw materials (e.g. monomers and solvent), and the production of electricity, the primary energy demand for the production of 1 kg LDPE is equal to 82.9 MJ and the emissions are equal to 1.87 kg of CO₂-eq (PlasticsEurope, 2014a). Regarding the input materials, 1 kg of LDPE required 1.02 kg of monomer in 1999 (European Commission, 2007), and the value was unchanged in 2014 (PlasticsEurope, 2014a), showing no improvement in the efficiency of the process.

4.3.2 Solution polymerisation

Low-Density Polyethylene (LDPE) is a semi-crystalline polymer with an average density of 0.91-0.94 g/cm³ and a melting point of 105-115°C. It has a linear structure with short and uniform side-chain branches (Omnexus, 2020a), as shown in Figure 21:



Figure 21: LLDPE structure with the main carbon chain in black and the side branches in blue (Graziano et al., 2019).

LLDP can be produced through a solution process (which is also used to produce HDPE) or a gas phase process (which is also used to produce HDPE and PP). In the solution process, the ethylene is diluted with the co-monomer (such as hexene-1 and octene-1) and a hydrocarbon solvent in the range of C6 to C9, which are all purified by passing throughout beds of absorbent material (European Commission, 2007). As catalyst, both Ziegler-Natta and Metallocene-types can be applied (PlasticsEurope, 2014a). The solution is then sent to the polymerisation reactor, which operates at a temperature higher than 100°C and with pressure up to 20 MPa (European Commission, 2007). The unreacted ethylene and the residual solvent are separated by the evaporator, then the polymer passes through the standard steps of extrusion (including the addition of additives), drying, blending, degasification, and storage (European Commission, 2007), as shown in Figure 22.



Figure 22: LLDPE solution polymerisation process scheme. Made with information from (European Commission, 2007; PlasticsEurope, 2014a; SABIC Limburg, 2020).

In 1999, the European average energy demand for LLDPE production was equal to 2.4 MJ/kg of direct energy, or 4.1 MJ/kg of primary energy (assuming an efficiency of 40% for electricity and 90% for steam), while the average of the top 50% companies was equal to 2.1 MJ/kg of direct energy, or 2.9 MJ/kg of primary energy (European Commission, 2007). More than one decade later, the values are not so different, as in 2014 the production of LLDPE still required an average of 3.9 MJ/kg of primary energy (assuming an efficiency of 40% for electricity and 90% for steam, see Appendix A). As shown in Table 8, the polymerisation of 1 kg of LLDPE causes the emission of 0.27 kg of CO₂-eq, considering the Global Warming Potential (GWP) for 100 years.

Table 8: Energy demand and global warming potential for the polymerisation of 1 kg of LLDPE. Average values from literature (European Commission, 2007; PlasticsEurope, 2014b) and internal communication with the companies involved.

Product	Electricity demand [MJ]	Heat demand [MJ]	GWP [kg CO2-eq]
LLDPE (1 kg)	1.27	0.69	0.23

If the boundaries of the life-cycle analysis are extended to the extraction of fossil and mineral resources, the production of raw materials (e.g. monomers and solvent), and the production of electricity, the primary energy demand for the production of 1 kg LLDPE is equal to 79.2 MJ and the emissions are equal to 1.87 kg of CO₂-eq (PlasticsEurope, 2014a). Regarding the input materials, 1 kg of LLDPE required 1.03 kg of monomer in 1999 (European Commission, 2007) and 1.02 kg of monomer in 2014 (PlasticsEurope, 2014a), showing a small improvement in the efficiency of the process.

4.3.3 Slurry polymerisation

High-Density Polyethylene (HDPE) is a high crystalline and low amorphous polymer with an average density of 0.94-0.97 g/cm³ and a melting point of 120-140°C. It has a linear structure with a low degree of short side-chain branches (Omnexus, 2020a), as shown in Figure 23.



Figure 23: HDPE structure with the main carbon chain in black and the side branches in blue (Graziano et al., 2019)

HDPE is mainly produced through a suspension/slurry process (which is also used to produce PP), but also through a solution process (which is also used to produce LLDPE) or a gas phase process (which is also used to produce LLDPE and PP). Moreover, two different reactors can be applied in the suspension/slurry process: a stirred tank reactor (STR) or a loop reactor (PlasticsEurope, 2014a). The flow diagram for HDPE production using a suspension process and a loop reactor is shown in Figure 24.



Figure 24: HDPE suspension/slurry polymerisation process scheme. Made with information from (European Commission, 2007; PlasticsEurope, 2014a; SABIC Limburg, 2020).

The process starts with the purification and the mixing of the reagents: besides the ethylene as main feedstock, HDPE also requires a co-monomer (such as butene-1 or hexene-1), a catalyst (usually Ziegler-Natta organo-metallic compounds), and a solvent in the C5-C8 hydrocarbon group (PlasticsEurope, 2014a). If the loop reactor is used, Philips and Metallocene catalysts can also be applied, coupled with isobutane as solvent (European Commission, 2007). The mixture is fed to the reactor, which operates under 80-90°C and 0.5-1 MPa conditions. The resulting polymer is insoluble and precipitates in a fine suspension that can be separated from the solvent by centrifugation (PlasticsEurope, 2014a). The concentration of HDPE in the slurry is dependent on many process parameters, but it generally ranges between 30 and 45% by weight (European Commission, 2007). After the centrifuge, the polymer faces the standard steps of extrusion in granules (including the addition of additives to obtain the desired properties), drying, blending, degasification of residual monomer, and storage in silos (PlasticsEurope, 2014a; SABIC Limburg, 2020).

In 1999, the European average energy demand for HDPE production was equal to 2.5 MJ/kg of direct energy, or 5.1 MJ/kg of primary energy (assuming an efficiency of 40% for electricity and 90% for steam), while the average of the top 50% companies was equal to 2.1 MJ/kg of direct energy, or 4.2 MJ/kg of primary energy (European Commission, 2007). More than one decade later, the values are not so different, as in 2014 the production of HDPE still required an average of 5.4 MJ/kg of primary

energy (assuming an efficiency of 40% for electricity and 90% for steam, see Appendix A). As shown in Table 9, the polymerisation of 1 kg of HDPE causes the emission of 0.27 kg of CO_2 -eq, considering the Global Warming Potential (GWP) for 100 years.

Table 9: Energy demand and global warming potential for the polymerisation of 1 kg of HDPE. Average values from literature (European Commission, 2007; PlasticsEurope, 2014b) *and internal communication with the companies involved.*

Product	Electricity demand [MJ]	Heat demand [MJ]	GWP [kg CO ₂ -eq]
HDPE (1 kg)	1.50	1.50	0.27

If the boundaries of the life-cycle analysis are extended to the extraction of fossil and mineral resources, the production of raw materials (e.g. monomers and solvent), and the production of electricity, the primary energy demand for the production of 1 kg HDPE is equal to 80.2 MJ and the emissions are equal to 1.80 kg of CO_2 -eq (PlasticsEurope, 2014a). Regarding the input materials, 1 kg of HDPE required 1.03 kg of monomer in 1999 (European Commission, 2007) and 1.02 kg of monomer in 2014 (PlasticsEurope, 2014a), showing a small improvement in the efficiency of the process.

4.3.3 Gas-phase polymerisation

Polypropylene (PP) is a semi-crystalline and low amorphous polymer with an average density of 0.90-0.91 g/cm³ and a melting point of 210-290°C (BPF, 2020). It has a linear structure with a low degree of unsaturation, and thus of short side-chain branches, as shown in Figure 25.



Figure 25: PP structure with the main hydrocarbon chain in white and the methyl groups in black (BPF, 2020).

PP can be produced with a gas-phase process (which is also used to produce LLDPE and HDPE) or with a suspension/slurry process (which is also used to produce HDPE). In the gas phase process, the first step is to mix and compress the gaseous propylene, co-monomer, hydrogen and catalyst together before sending them in a steady flow to the polymerisation reactor under 2-4 MPa and 70-90°C conditions (European Commission, 2007). For many companies adopting the Borstar loop-reactor or Unipol II fluidised bed reactor technologies, it has become common to have a setup of two or more reactor in sequence. This so-called multistage configuration increases the capital cost of the production line but adds flexibility and widens the portfolio of produced polyolefins (Kanellopoulos & Kiparissides, 2019). which is For PP production, catalyst based on titanium and aluminium are usually chosen, including Ziegler-Natta, Phillips-types, and Metallocene-types (PlasticsEurope, 2014b). The resulting polymer powder is extracted at the bottom of the reactor and moved to a degassing vessel to deactivate the catalyst and remove the residual feedstock, which is cooled and recycled back into the cycle (PlasticsEurope, 2014b), as seen in Figure 26.



Figure 26: PP gas phase polymerisation process scheme. Made with information from (European Commission, 2007; PlasticsEurope, 2014b; SABIC Limburg, 2020).

The resulting PP is sent to the granule extruder, where the necessary additives are also added to the polymer mix. Finally, the product faces the standard steps of drying, blending, degasification of the residual monomer and storage in silos (European Commission, 2007).

The European average energy demand for PP production was not reported in 1999, but it could be considered equivalent to comparable HDPE processes, which had an average consumption of 5.1 MJ/kg of primary energy (European Commission, 2007). More than one decade later, the value seems to have improved, as in 2014 the production of PP required an average of 3.8 MJ/kg of primary energy (assuming an efficiency of 40% for electricity and 90% for steam, see Appendix A). As shown in Table 10, the polymerisation of 1 kg of PP causes the emission of 0.20 kg of CO₂-eq, considering the Global Warming Potential (GWP) for 100 years, making PP more environmentally friendly than LDPE, LLDPE, and HDPE.

Table 10: Energy demand and global warming potential for the polymerisation of PP. Average values from literature (European Commission, 2007; PlasticsEurope, 2014b) and internal communication with the companies involved.

Product	Electricity demand [MJ]	Heat demand [MJ]	GWP [kg CO ₂ -eq]
PP (1 kg)	1.26	0.61	0.20

If the boundaries of the life-cycle analysis are extended to the extraction of fossil and mineral resources, the production of raw materials (e.g. monomers and solvent), and the production of electricity, the primary energy demand for the production of 1 kg PP is equal to 77.9 MJ and the emissions are equal to 1.63 of CO₂-eq (PlasticsEurope, 2014b). Regarding the input materials, 1 kg of PP required 1.03 kg of monomer in 1999 (European Commission, 2007) and 1.02 kg of monomer in 2014 (PlasticsEurope, 2014b), showing a small improvement in the efficiency of the process.

4.4 Decarbonisation options

This section answers the fourth research question: "Which decarbonisation options are considered as most promising by the stakeholders, and to what extent are they technically, economically, and environmentally comparable to the current technologies?", describing two bio-based polyolefins processes and four innovative recycling technologies for monomers and polymers.

4.4.1 Sugar-based olefin monomers

The most established way to produce ethylene (and to some extent, propylene) from bio-based sources is by dehydrating ethanol obtained through the fermentation of sugar-rich crops. The typical plant is the sugarcane, which grows mostly in Brazil (39% of the global production), India (20%), China, Thailand, Pakistan and Mexico (FAOSTAT, 2018). Braskem, the biggest chemical company in Brazil and the global leader of bio-ethanol and bio-plastics production, collects sugarcane from 65,000 hectares (about 0.2% of the total arable land in Brazil) to produces 200 kt of bio-PE every year (Braskem, 2020). As seen in Figure 25, the typical first-generation biorefinery involves the extraction of the sugarcane juice and its fermentation using microorganisms (e.g. yeast, bacteria or molds), followed by a distillation process to separate the ethanol. The leftover fibrous residue of the sugarcane, which is called bagasse, is burned in a cogeneration plant to supply the biorefinery processes and possibly produce surplus electricity (Braskem, 2020; Gotro, 2013).



Figure 27: Process steps of bio-ethylene production, including the amounts of sugarcane necessary to produce 1 kg of final product. Made with information from (Braskem, 2017, 2020; Gotro, 2013; Machado et al., 2016).

The dehydration process is an endothermic chemical reaction, and it is usually carried out by heating the ethanol to 300-500°C together with a catalyst (e.g. concentrated sulfuric acid or activated alumina) in a fluidised bed reactor, causing the separation of bio-ethylene and water (Mohsenzadeh et al., 2017; M. Zhang & Yu, 2013). Figure 28 shows the dehydration process, while the chemical reaction happening inside the reactor is $C_2H_5OH \rightarrow C_2H_4 + H_2O$.



Figure 28: Dehydration process of ethanol to ethylene (M. Zhang & Yu, 2013).

Although the production of bio-ethanol and the de-hydration into ethylene have an energy consumption comparable to the production of ethylene from naphtha cracking (Ghanta et al., 2014), the absorption of CO₂ during the growth of the crops makes bio-based polyethylene (bio-PE) significantly better than his fossil counterpart when fighting climate change. Moreover, most of the energy necessary for the fermentation and distillation of the bio-ethanol can be produced through onsite cogeneration using the sugarcane bagasse as fuel, thus making the production process less reliant on fossil fuels (Machado et al., 2016). The cradle-to-gate life-cycle analysis (LCA) of bio-ethylene, starting from the cultivation of sugarcane and ending after the polymerisation process, shows that the overall process is a net-absorber of greenhouse gases, removing 3.09 kg of CO₂-eq for every kg of PE produced (Braskem, 2017). As the LCA does not cover that gate-to-grave cycle of the plastics, the CO_2 released when the plastic waste is burned for energy recovery (a common process in many European countries), is not considered. Would that be included, then 3.14 kg of CO₂- would be released in the atmosphere for every kg of PE incinerated, thus yielding an almost net-zero carbon balance. Another LCA presents a significantly smaller value, having calculated that every kg of PE produced from biobased sources causes the net absorption of 0.75 kg of CO_2 -eq from the atmosphere (Tsiropoulos et al., 2015). Even though the exact carbon balance of bio-PE varies from case to case, the benefit is still clear, especially when considering that each kg of fossil-based PE causes the emission of around 1.86 kg of CO₂-eq from cradle to gate (PlasticsEurope, 2014b).

Finally, one study shows that when considering the whole cradle-to-grave life cycle, each kg of bioethanol used as precursor for bio-PE has a net positive emission (1.3-2.0 kg of CO₂-eq), which in any case is around half of the emissions caused by its fossil-based counterpart (3.7 kg of CO₂-eq) (Muñoz et al., 2014). Seeing how great the impact of the materials end-life on GHG emissions is, it is extremely important to establish a circular economy system, which will be discussed in Section 4.4. Lastly, bio-PE has some major downsides compared to fossil-PE, namely high values of ozone and water depletion, land use, and terrestrial and aquatic eutrophication (Braskem, 2017), making necessary to evaluate the sustainability of each case and to carry out a multi-criteria environmental analysis assigning a weight to the different impacts on the local ecosystem and society. A comparison of fossil-based PE and bio-PE from sugarcane can be observed in Figure 29: Comparison of the cradle-to-gate environmental impact of fossil-based PE and sugarcane-based PE. Normalised values extracted from (*Braskem, 2017*). .



Figure 29: Comparison of the cradle-to-gate environmental impact of fossil-based PE and sugarcane-based PE. Normalised values extracted from (Braskem, 2017).

In Europe the cultivation of sugarcane is not common, therefore 43% of the European bio-ethanol is produced from corn, 26% from wheat, 21% from sugar beet, and 10% from other sources like lignocellulosic residues (ePURE, 2020). More than 80% of the European bio-ethanol is blended with gasoline to produce biofuel (usually with a 5-10% share of ethanol), and the market is expected to rapidly grow in the next years (ePURE, 2020), potentially competing with the bio-ethanol demand for bioplastics production. A Japanese study calculated that the added value and GHG reduction of using bio-ethanol for bioplastics production are greater than when bio-ethanol is used as fuel (Kikuchi et al., 2017), while an American study calculated that the GHG reduction achieved is indeed the same, but the economic potential is better when the bio-ethanol is used as a substitute for gasoline (McKechnie et al., 2015). Finally, according to a European study the use of sugar beet to produce bio-ethanol and then bio-PE in Europe is considered at least as economically convenient as producing ethylene from fossil sources, with a margin of uncertainty of ±10% due to the price volatility of the feedstocks (Posada et al., 2013). It is thus difficult to clearly evaluate the economic convenience of investing in bio-PE, but all the studies agree that scaling-up the production would have positive effects and make the production of bio-plastics more financially attractive.

In the Netherlands, sugar beet cultivation has a very high yield and it is thus possible to produce 7,200 litres of bio-ethanol for each hectare, which is the same value obtained by Brazilian sugar cane (Langeveld et al., 2014). The energy intensity in the Netherlands, however, is way higher: while sugar cane has a net energy balance ratio of 8, the Dutch sugar beet only reaches 1.3, although it could be improved up to 2.8 by using CHP systems for the distillation and reducing the amount of fertilizers used for the crops (Langeveld et al., 2014). Since the production of bio-ethanol only requires the sugar component of the beet, the remaining part can be used as animal feed: for every litre of bio-ethanol, the fermentation process also produces almost 1 kg of co-products (Dammer et al., 2017). According to this data, from one hectare of sugar beet it would be possible to produce 3 tons of bio-PE and 7 tons of animal feed. In that sense, it could be said that bio-ethanol production can make a positive contribution and do not be in contrast with food and feed security (Dammer et al., 2017). And some research even shows that a fair supply chain for bio-plastics may stabilize food prices and provide an additional income to farmers, thus making food production more sustainable (Van Den Oever et al., 2017).

If Dow Terneuzen and SABIC Limburg were to completely substitute their current production of 1,840 kt/year fossil-based PE with its bio-based counterpart, the required bio-ethanol would be equal to 4 billion litres. If all of it was to be produced from sugar beets grown in The Netherlands, that would require around 613,000 hectares of land, which is over 7 times more than the current area and is equal to 59% of the arable land in the country (FAOSTAT, 2018). Apart from being unrealistic from a socio-technical point of view, many studies show that the import of the required 4 billion litres of bio-ethanol from other countries (e.g. Brazil) would be economically more convenient (Althoff et al., 2013).

4.4.2 Biowaste-based olefin monomers

As an alternative to the biochemical route involving fermentation and distillation, bio-based ethylene and propylene can also be produced with a thermochemical process. As seen in Figure 30: Steps of the gasification process of biomass into methanol. Made with information from (Brachi et al., 2014; De-León Almaraz & Azzaro-Pantel, 2017; Ragaert et al., 2017)., the process starts with the gasification of lignocellulosic biomass (e.g. wood chips) or bio-waste (e.g. forest and agricultural residues, or byproducts of the paper industry), although the feedstock may need to be pre-treated to reduce the amount of moisture and increase the energy density (Nouri & Tillman, 2005). Gasification is the incomplete combustion (or partial oxidation) of the biomass feedstock in order to break down the molecules and produce a mix of syngas, char, and tars. If catalysts are used, the reaction takes place at 750-900°C (De-León Almaraz & Azzaro-Pantel, 2017), otherwise the temperature needs to be in the 1200-1500°C range (Ragaert et al., 2017). The unreformed syngas and biomass chars are combusted to supply heat to the gasification reactor, while the tars are reformed into useful syngas using a catalytic reaction (Foust et al., 2009). The resulting syngas needs to be cleaned and conditioned to remove acid gases and impurities, before being compressed and sent to the methanol synthesis reactor (Brachi et al., 2014). Quite often, the syngas needs to undergo a water-gas shift (WGS) reaction to maximize the hydrogen content (Speight, 2019). The conversion reaction is facilitated by a commercial methanol catalyst, ensuring a selectivity of 99.8% (Xiang et al., 2015).



Figure 30: Steps of the gasification process of biomass into methanol. Made with information from (Brachi et al., 2014; De-León Almaraz & Azzaro-Pantel, 2017; Ragaert et al., 2017).

Methanol (CH₃OH) is the simplest chemical in the alcohol category and it can be used as a precursor to other chemicals, or as fuel for transportation and energy production (Shamsul et al., 2014). Methanol-To-Olefins (MTO) is a well-known conversion process, whose chemical reactions have been studied for more than two decades (Nouri & Tillman, 2005). As seen in Figure 31, the bio-methanol faces a catalytic conversion in a fluidised-bed reactor at 350°C and 30 atm (Ragaert et al., 2017), often

coupled with a catalyst regenerator. The resulting gaseous product is sent to a purification unit to separate the olefins, with a final yield of 49-55% for ethylene and 25-33% for propylene (Xiang et al., 2015)



Figure 31: Methanol-to-olefins (MTO) process (Xiang et al., 2015).

Overall, the production of 1 kg of light olefins requires the input of 4 kg of biomass feedstock and 8 MJ of energy, which is used mainly for the gasification process (Xiang et al., 2015). In comparison, the production of ethylene and propylene from naphtha cracking consumes around 16 MJ of primary energy (PlasticsEurope, 2012), so it could be assumed that the carbon emissions of bio-based ethylene are also halved compared to fossil-based ethylene. Although gasification and MTO conversion follow a production route that is quite different from the fermentation and dehydration of bio-ethanol, the production of bio-polyolefins using thermochemical and biochemical process seem to have very comparable economic and environmental impacts (Foust et al., 2009). Considering that they require two different types of biomass feedstock, they complement well each other, and both have the potential to make a positive contribution to the Dutch decarbonization strategy.

4.4.3 Techno-economic indicators and current projects on bio-based polyolefins

Using data from (Oliveira et al., 2020) adjusted for the European market, Table 11 presents a comparison between the standard production of naphtha-based ethylene using steam crackers, the production of sugar-based bio-ethylene through fermentation of sugar beets, and the production of methanol-based ethylene from the gasification of biowaste. As the production of bio-based ethylene is more expensive than when using naphtha crackers, a price on carbon emissions is necessary to make the process convenient. It is assumed that 40% of the PE gets recycled at the end of its life, thus keeping the carbon sequestered from the atmosphere (in the Netherlands 32% of the plastics is already recycled, and the share will most likely increase in the next years thanks to improvements in mechanical and chemical recycling processes). In addition, the "cheap feedstock" case is made using the price of Brazilian ethanol, which is roughly 50% cheaper than the European one (GlobalPetrolPrices, 2020).

Table 11: Economic indicators and break-even carbon price necessary to make the production of bio-ethylene as economically convenient as its naphtha-based counterpart. Based on data from (Oliveira et al., 2020) adjusted for the European market (More information in Appendix A).

Indicator	Naphtha-based olefins	Sugar-based olefins	Biowaste-based olefins	
Levelized costs	1 070 80	1 672 57	1 792 04	
(€ ₂₀₁₇ /t olefins)	1,070.80	1,072.57	1,792.04	
GHG best-case	1.02	2.07	-2.72	
(tCO ₂ -eq/t olefins)	1.02	-2.57		
Break-even carbon	0	150.92	192.84	
price (€/ tCO₂-eq)	0	130.82		
GHG 40%-case	1.02	-1.00	-0.84	
(tCO ₂ -eq/t olefins)	1.02	-1.05	-0.04	
Break-even carbon	0	20E 7/	288 60	
price (€/ tCO₂-eq)	0	203.74	568.00	

To summarise, the production of bio-PE and bio-PP from ethanol or methanol is certainly one of the most promising options to decarbonise the Dutch plastic sector. The implementation of a large-scale supply chain for bio-based polymers, however, would require a joint effort from both companies and governments. From a technological and logistical point of view, the conversion technology for lignocellulosic materials must improve, and the production of sucrose or starchy crops needs to be expanded without competing with food production (Broeren, 2013). On the other hand, European and Dutch policies also need to change to increasing the tax for carbon emissions and removing subsidies to fossil fuels, which are equal to almost 40 billion euros (Hayer, 2017). Other measures beneficial for bio-plastics would include the incentive schemes and the removal of tariffs on imported bio-ethanol (IRENA, 2013). The combination of all these measures could make bio-based PE and PP as convenient as their fossil-based counterparts, facilitating the development of sustainable production facilities by the chemical companies and the achievement of the national and European decarbonisation targets.

There are many bio-based polyolefins projects in development in the Netherlands, such as:

- The Chemelot Industrial Park has developed many bio-based chemistry processes, including the production of polymers from lignin (i.e. woody) biomass (Brightlands, 2020). The production of bioplastics from biomass and bio-waste is an important part of Chemelot's decarbonisation strategy for 2050 (Chemelot, 2020).
- Dow Terneuzen announced a partnership with the Finnish biorefinery UPM, which produces renewable naphtha from residues of paper pulp production. The feedstock is called BioVerno and can be used by Dow Terneuzen to produce bio-based LDPE, potentially cutting in half the emissions of CO₂ compared to the standard fossil-based process (Dow, 2019a). The production of bio-ethanol from lignocellulosic materials is made more difficult because the lignin molecules create a barrier around the cellulose, thus making it necessary to pre-treat the feedstock with a physicochemical process. The cellulose is then hydrolysed into simple sugars, and finally fermented and distilled just like the bio-ethanol obtained from sugarcane and sugar beet (Mohsenzadeh et al., 2017).
- Borealis announced a cooperation with the Finnish company Neste, which produces renewable propane from a mix of residue oils and other fat waste streams. The propane will be produced in Neste's facilities in the Rotterdam Port industrial cluster, and then

dehydrogenated into propylene in Borealis' Belgian plant of Beringen, where it will be used for the polymerisation of bio-based PP (BioPlastic Magazine, 2019). The production of biopropanol uses Neste's proprietary technology NEXBTL to process animal fat waste, used cooking oil (UCO), and other fat residues (e.g. from palm oil production), but the company is also looking for ways to use plastic waste as feedstock (Neste, 2020).

 SABIC announced its intention to create a co-continuous blend of PE and thermoplastic starch to obtain a packaging film resistant against both oxygen and water. The inclusion of the biopolymer, produced from potatoes, improves the carbon balance of the plastics and is especially suited for printable multilayer films. The company also announced its plan to produce bio-PE and bio-PP with bio-based feedstock produced from waste oils and other biomass not in competition with the food chain (Vachon, 2019).

4.4.4 Mechanically-recycled polyolefins

As seen in Figure 32, the main technological processes for the end-of-life management of the postconsumer plastic waste are mechanical recycling, feedstock recycling (also called chemical recycling), incineration (possibly with energy recovery), and landfill. After a brief description of mechanical recycling technologies, this section will focus on innovative chemical recycling process able to increase the share of recycled plastic waste.



Figure 32: Possible treatments for plastic waste, in order of priority from highest (mechanical recycling) to lowest (landfill). Made with information from (Al-Salem et al., 2017; Rigamonti et al., 2014).

Mechanical and chemical/feedstock recycling are the two only options that allow for the recovery of the material and its re-inclusion in the circular economy cycle and are thus the preferred technologies for the treatment of plastic waste. Moreover, other options such as incineration and landfill have a significantly higher environmental impact (Lazarevic et al., 2010; Rigamonti et al., 2014). In the Netherlands, the annual demand for plastics is equal to 2.2 million tons, and around 32% of the post-consumer plastic waste is mechanically recycled, while the remaining part is incinerated in energy recovery plants (PlasticsEurope, 2019).

Mechanical recycling is the most common method for the material recovery of plastic waste, although it requires the waste to be sorted and divided into single-polymer plastic streams (Al-Salem et al., 2009). As seen in Figure 33, after the collection of the plastic waste the process starts with the separation of the plastic products based on chemical composition, density, colour, size and shape. The

waste is then washed to remove contamination (often organic, e.g. food waste, or chemical, e.g. glue and labels), ground into flakes, and finally milled and compounded into pellets and granules (Ragaert et al., 2017; Singh et al., 2017).



Figure 33: Mechanical recycling process. Made with information from (Al-Salem et al., 2009; Ragaert et al., 2017; Singh et al., 2017).

Sorting and separating the different polymers are the most delicate steps of the mechanical recycling process, and can be done by a combination of FT-NIR (Fourier Transform Near Infrared), optical colour recognition, a ballistic separator, float-sink techniques, and manual sorting by trained operators (Ragaert et al., 2017). Other possible sorting methods are triboelectric, X-rays, and high-speed separation (Al-Salem et al., 2009). The carbon footprint of mechanical recycling has been measured to be equal to -0.5 kg of CO₂-eq/kg input waste, compared to the production of virgin plastics for naphtha crackers (CE Delft, 2019). From a life cycle perspective, mechanical recycling is thus the preferred option to manage plastic waste, provided that the plastic stream is well defined, there is little organic contamination and the substitution ratio with virgin plastics is close to 1:1. For hard-to-recycle plastic waste, feedstock recycling becomes the preferable option (Lazarevic et al., 2010). Another factor that could make chemical recycling a better choice than mechanical recycling is the thermal-mechanical degradation faced by the polymers during the shearing and melting processes (Ragaert et al., 2017).

4.4.5 Dissolution-recycled polyolefins

Solvent-based purification, also known as dissolution, is a chemical recycling process able to recover high-quality polymers and separate them from additives and impurities. Although the method could technically be applied to mixed plastic waste, current setups are only able to handle homogeneous flows of polymers (Zero Waste Europe, 2019). When talking about polyolefins, however, an experiment involving the solvent-based purification process of a 50/50 mix of PE and PP has been proved successful, with a recovery rate of 99% for both polymers (Pappa et al., 2001). If the plastic waste contains a mix of LDPE, HDPE, PP, PS, and PVC, a preliminary separation by floatation in water is necessary to separate the polyolefins from the heavier polymers (PS and PVC) before processing them with solvent-based recycling (Pappa et al., 2001).

As seen in Figure 34, the process starts by cutting the polyolefins and cleaning them to remove organic contaminants. The polymers are then dissolved with a solvent (S) and filtrated to remove the unsolved particles (e.g. other polymers or impurities), then precipitated using an anti-solvent (AS), filtered from the A/AS liquid, washed and dried (Pappa et al., 2001). The solvent and anti-solvent are separated by distillation and reused in the next cycle. For the chemical recycling of PE and PP, the most common

selective solvent is xylene, while propanol, ACE or n-hexane can be used as anti-solvent for the precipitation step (Zhao et al., 2018).



Figure 34: Solvent-based purification process. Made with information from (Pappa et al., 2001; Zero Waste Europe, 2019; Zhao et al., 2018).

The main advantage of dissolution recycling is that it removes all the additives and unsolvable contaminants, resulting in recycled polymers with properties comparable to virgin products. On the long term, however, the stress generated by the process may affect the quality of the polymers, meaning that solvent-based purification cannot be used as perpetual chemical recycling method (Zero Waste Europe, 2019). Other disadvantages are the relatively high technical requirements and costs (Zhao et al., 2018), so improvements in the process could be necessary to facilitate the deployment of this technology on a national-scale level in the Netherlands. One example of solvent-based PE and PP recycling process at the industrial level is the APK chemical recycling plant, located in Germany and active since 2018 (APK AG, 2020). Outside of Europe, Unilever has co-developed a solvent-based process designed to recycle sachets and since 2018 has been operating a pilot plant in Indonesia with a capacity of 3 ton/day, with the ambition of developing a 30 ton/day commercial plant as soon as possible (Unilever, 2020).

4.4.6 Pyrolysis-recycled olefin monomers

Pyrolysis is the thermal conversion process at elevated temperatures in the absence of oxygen that allows the recovery of the monomers from the polymer plastics waste (Ray & Thorpe, 2007). Its advantage is that, unlike mechanical recycling, it can handle highly heterogeneous mixtures of plastics, such as the modern multi-layer packaging materials (Ragaert et al., 2017). A variety of researchers have demonstrated the feasibility of the pyrolysis of plastic waste composed by a mix of PE, PP and PS (Demirbas, 2004; Donaj et al., 2012; Kaminsky et al., 1996), although PET and PVC need to be separated and removed to guarantee the quality of the final product (Anuar Sharuddin et al., 2016). The usual pyrolysis product is a hydrocarbon liquid called pyrolysis oil, which can be used as a heavy fuel oil substitute (Fivga & Dimitriou, 2018). However, if the process reaches high-temperatures (650-850°C) secondary cracking of the gas phase occurs, resulting in the production of a wide spectrum of smaller hydrocarbons (Ray & Thorpe, 2007). This allows pyrolysis to directly produce ethylene and propylene, thus avoiding the need to run the pyrolysis oil into the steam crackers. Since the pyrolysis process requires high energy, using catalysts to reduce the optimal temperature helps to reduce the overall cost, and it also improves the production of olefins. The most common catalysts used in plastic pyrolysis are zeolites, FCC, and silica-alumina catalysts (Anuar Sharuddin et al., 2016).

As seen in Figure 35, the process starts with the pre-treatment of the mixed plastic waste, which usually consists just in the shredding of the polymers to a size of 1-5 mm (Donaj et al., 2012). As explained before, it is extremely important to use only polyolefins as feedstock, thus excluding PET and PVC (Anuar Sharuddin et al., 2016). The pyrolysis reaction takes place in a fluidised bed reactor and the products are then separated, with some of the by-products (e.g. methane) being combusted to supply the heat to the pyrolysis reactor (Fivga & Dimitriou, 2018). The resulting ethylene and propylene can then be sent to the PE and PP polymerisation processes described in Section 2 of this report.



Figure 35: Pyrolysis process. Made with information from (Donaj et al., 2012; Ray & Thorpe, 2007).

The pyrolysis of polyethylene to produce lower olefins has been tested since more than two decades: experiments have shown that LDPE can be chemically recycled in a fluidized bed reactor, obtaining a max yield of 27% for ethylene gas and 19% for propylene gas with a temperature of 700°C. When considering also the production of methane and other by-products, the overall yield reached 73% (Williams & Williams, 1999). The pyrolysis of HDPE in a fluidized bed reactor has also been proven successful, obtaining a max yield of 42% for ethylene gas and an overall yield of 86% with a temperature of 780°C and residence time of 1.34 seconds (Mastral et al., 2002). Dozens of studies also investigate the pyrolysis of plastic waste in combination with biomass, reporting that the thermal conversion is facilitated, and the yield is higher than the sum of the two independent processes (Xue et al., 2015). Some economic indicators for a commercial pyrolysis plant are given in Table 12.

Table 12: Comparison of the economic indicators for a pyrolysis plant with a capacity of 100 kg/h, 1,000 kg/h, and 10,000 kg/h (Fivga & Dimitriou, 2018). Values adjusted for inflation, more information in Appendix A.

Indicator	Capacity of 100 kg/h	Capacity of 1,000 kg/h	Capacity of 10,000 kg/h	
CAPEX [M€ ₂₀₂₀]	1.24	3.84	11.72	
OPEX [M€ ₂₀₂₀ /year]	0.53	1.50	2.76	
Payoff period [years]	Never	3.6 1.2		
NPV in 20 years [M€ ₂₀₂₀]	-1.4	13 220		
Additional comments	Negative revenue	Economically convenient	Lowest cost per unit	

From an environmental point of view, using pyrolysis to recover ethylene and propylene from plastic waste reduces the need to produce them from virgin oil and natural gas, thus reducing the emissions of carbon monoxide and CO₂ (Al-Salem et al., 2017). Recent simulations based on LCA studies estimate a reduction of 0.33 kg of CO₂-eq for each kg of PE and PP recycled through pyrolysis, compared to the reference emissions caused by the production of virgin material from naphtha cracking (*Internal communication with TNO*). The result is the same range of another analysis, which found out an emissions reduction up to 0.20 kg of CO₂-eq/kg input waste (CE Delft, 2019). Today, pyrolysis is mainly applied to produce crude diesel for power plants or ship fuel, making policy intervention a necessary step to ensure that the technology is used to close the plastic-to-plastic loop instead (Zero Waste Europe, 2019).

4.4.7 Gasification-recycled olefin monomers

As seen in Section 4.4.2, lignocellulosic materials and biowaste can be gasified into syngas, which is then converted into methanol and then into olefins. The same process can be applied to hard-torecycle plastic waste and municipal solid waste (MSW), thus increasing the circularity of the polymers supply chain (Arena et al., 2011). As seen in Figure 36, the process starts with the gasification of the plastic waste, although the feedstock may need to be pre-treated to reduce the amount of moisture and increase the energy density (Nouri & Tillman, 2005). Gasification is the incomplete combustion (or partial oxidation) of the feedstock in order to break down the molecules and produce a mix of syngas, char, and tars. If catalysts are used, the reaction takes place at 750-900°C (De-León Almaraz & Azzaro-Pantel, 2017), otherwise the temperature needs to be in the 1200-1500°C range (Ragaert et al., 2017). When MSW is included in the feedstock mix, the optimal temperature for catalytic gasification seems to be 900°C in order to minimize the quantity of tars produced (Guan et al., 2009). The unreformed syngas and carbon chars are combusted to supply heat to the gasification reactor, while the tars are reformed into useful syngas using a catalytic reaction (Foust et al., 2009). The resulting syngas needs to be cleaned and conditioned to remove acid gases and impurities, before being compressed and sent to the methanol synthesis reactor (Brachi et al., 2014). Quite often, the syngas needs to undergo a water-gas shift (WGS) reaction to maximize the hydrogen content (Speight, 2019). The conversion reaction is facilitated by a commercial methanol catalyst, ensuring a selectivity of 99.8% (Xiang et al., 2015).



Figure 36: Gasification of plastic waste into methanol. Made with information from (Brachi et al., 2014; De-León Almaraz & Azzaro-Pantel, 2017; Ragaert et al., 2017).

Methanol (CH₃OH) is the simplest chemical in the alcohol category and it can be used as a precursor to other chemicals, or as fuel for transportation and energy production (Shamsul et al., 2014). Methanol-To-Olefins (MTO) is a well-known conversion process, whose chemical reactions have been studied for more than two decades (Nouri & Tillman, 2005). As seen in Figure 37, the bio-methanol faces a catalytic conversion in a fluidised-bed reactor at 350°C and 30 atm (Ragaert et al., 2017), often coupled with a catalyst regenerator. The resulting gaseous product is sent to a purification unit to separate the olefins, with a final yield of 49-55% for ethylene and 25-33% for propylene (Xiang et al., 2015).



Figure 37: Methanol-to-olefins (MTO) process (Xiang et al., 2015).

Many studies investigate the possibility of combining biomass and plastic waste as feedstock in order to achieve a better performance of the system. When polymeric waste (in quantities exceeding 20% of mass share) is co-processed with biomass, the resulting syngas has such quality that does not require the WGS conditioning to become suitable for methanol synthesis, thus simplifying the system configuration and reducing the projected costs (Brachi et al., 2014). Another successful syngas process managed to achieve 98% of energy conversion when mixing 60% of PE plastic waste with wood waste in the form of pine chips (Pinto et al., 2002). Promising results have been obtained also when using a mix of polyethylene waste and biomass in a 0.3 mass ratio (Moghadam et al., 2014). In conclusion, gasification of plastic waste (potentially in combination with biomass) seems to be the most promising technology to process hard-to-recycle plastic waste, reducing the need for virgin plastics and increasing

the share of recovered carbon content, potentially leading to emissions reduction up to -0.25 kg CO_2 - eq/kg input (CE Delft, 2019).

4.4.8 Techno-economic indicators and current projects

The chemical recycling industry is still at its infancy and still need further research. Most pyrolysis and gasification plants for the treatment of plastic waste are in a pilot stage and the deployment of these technologies on a commercial scale can only be expected after 2025. It is thus very difficult to show precise values regarding the economic and environmental indicators of these recycling technologies. Table 13 shows the available data, collected from multiple sources.

Table 13: Costs of the mechanical (Gradus et al., 2017) and chemical recycling options (Fivga & Dimitriou, 2018) and CO2 emissions reduction compared to the production of virgin material from naphtha crackers (CE Delft, 2019; Ligthart et al., 2019).

Indicator	Mechanical	Solvent-based	Pyrolysis	Gasification
Costs (€ ₂₀₂₀ /t input waste)	767	n.a.	330	n.a.
GHG emissions reduction (tCO ₂ -eq/t input waste)	-0.50	n.a.	-0.20	-0.25

Among the chemical recycling projects in The Netherlands, the following news has been gathered:

- Dow Terneuzen announced a partnership with the Dutch company Fuenix Ecogy Group for the supply of pyrolysis oil, which will be used by Dow to produce new polymers. The collaboration will help Dow Terneuzen in its goal to produce at least 100 kt of recycled plastics (around 9% of their current LDPE and PP production) by 2025 (Dow, 2019b).
- SABIC started a collaboration with Renewi and Plastic Energy to realize a chemical recycling
 plant in the Chemelot site to process low-quality mixed plastic waste to use as feedstock for
 their steam crackers. The plant will use pyrolysis technology to convert the plastic waste into
 feedstock, thus reducing both the flow of low-quality waste to incineration and the demand
 for fossil naphtha. The project is expected to be operational by 2021, helping SABIC in its goal
 to process 200 kt/year of recycled plastics by 2025 (Chemelot, 2018; Renewi, 2018).
- Recycling is already one of the most interesting options for plastics producers in the Netherlands, and both mechanic and chemical recycling are included in the circular economy strategy of the Rotterdam Port, the biggest industrial cluster of the country (Port of Rotterdam, 2019b).
- "Pyrolyseproeftuin Moerdijk" is a pyrolysis project in the Southern part of the Netherlands supported by various private and public parties. Among other experimental projects, the company Teknow Systems developed a pilot set-up for the recycling of plastic waste (Recycling Netherlands, 2019).

4.4.9 Other decarbonisation options

Biomass fuel for cogeneration: the electricity and steam required to produce polyolefins are usually generated by combined-heat-power (CHP) plants, which normally consist of natural gas-fired turbines paired with a steam boiler. To reduce the GHG emissions caused by the combustion of natural gas, the producers could choose to use bio-based fuels. As the CO₂ absorbed by the vegetation during its growth is almost the same as the one emitted when burning it, biomass is usually considered carbon neutral if the areas harvested are reforested (Sebastián et al., 2011). Some case studies suggest that the payback period of bio-CHP is the 5-11 years range (Ciric & Kuzmanovic, 2014), while the actual environmental performance depends on the supply chain, the

type of biomass and the competition for soil with edible crops, but in the best-case scenario the overall emissions can be ten times lower than when using natural gas (US EPA, 2007).

- Heat pumps: Industrial Heat Pumps (IHPs) can recover the waste heat of an industrial process and increase its temperature to make it useful for an adjacent process, thus reducing the overall energy consumption and relative GHG emissions (J. Zhang et al., 2016). Although many heat pumps for residential and district heating can only reach temperatures around the 50-90°C range, there are now IHP systems able to supply heat at temperatures higher than 100°C (IETS TCP, 2014). Within polyolefins production, IHPs could be used to prepare the feedstock for polymerisation and to dry the resulting polymer. Especially for the drying process, IHPs could be convenient thanks to their ability to control the gas temperature and humidity while extracting the latent heat of condensation (Chua et al., 2010), potentially consuming between 30 and 50% less energy than conventional steam drying cycles and adopting low GWP refrigerants like R-1234yf and R-744 (IETS TCP, 2014).
- Hydrogen fuel: Hydrogen has the potential to be a clean and renewable fuel used for transportation, industries, heating, and power generation, and in the last decades the industry has been rapidly growing (Gupta, 2008). Hydrogen is mainly produced through the reformation of natural gas and coal, or from water splitting using electrolysis, thermolysis, or photoelectrolysis, and there are many other processes under development, although not all of them are economically competitive (Holladay et al., 2009). The current market is currently equal to 0.8 Mt/year but has the potential to grow tenfold by 2030, leading to a yearly emission reduction of 5 Mt of CO₂ (CE Delft, 2018). Looking at current and future projects in the Netherlands, the Port of Rotterdam is planning to invest 2 billion euros in the development of production infrastructures and related value chain to produce hydrogen to use as fuel, carrier, or raw material in industrial processes. The estimated production will be able to supply up to 20% of the heat and power required by the industries part of the Rotterdam Port, leading to a CO2 emission reduction of 16% (Port of Rotterdam, 2020).
- **Process design**: The key concept of process design improvements is to change the manufacturing process of polymers to reduce the energy consumption and GHG emissions, while at the same time preserving the productivity and quality of the products. Ever minor energy efficiency improvements could have big impacts over time. It is estimated that the total primary energy losses related to the production of polyolefins in Western Europe are exceeding 100 PJ for PE and 50 PJ for PP (Neelis et al., 2007). Example of technologies and process that could help to achieve this goal are: (1) Design optimisation (e.g. reduced leakage) and energy efficiency improvements (e.g. power factor) for the industrial compressors used during the polymerisation process (Ekradi & Madadi, 2020; Hu et al., 2019; Mascarenhas et al., 2019). (2) Use of better catalysts to lower the temperature and pressure requirements for the polymerization process and increase the copolymerisation performance (Zhu et al., 2011). (3) Renewal of existing process parts, integration of multi-functional equipment, and superstructure design optimization (Harmsen, 2004).
- **Carbon capture**: The key concept of this decarbonisation option is not to reduce the GHG emissions, but to capture the carbon dioxide before it is released into the atmosphere and store it in protected reservoirs. In short, the term CCS (Carbon Capture and Storage) refers to technologies which capture carbon dioxide from processes such as gasification and power generation. The CO2 is then pressurised to 100 bar or more, transported, and stored underground (e.g. in a depleted natural gas reservoir) (Boot-Handford et al., 2014). An example of CCS project in the Netherlands can be found in the Rotterdam Port industrial cluster, where petrochemical companies such as Shell, ExxonMobil and Air Liquide have signed an agreement with the project organisation Porthos for the realisation of a CCS system. The carbon emitted by the companies will be captured, transported, and stored beneath the North Sea by the end of 2023 (Port of Rotterdam, 2019a).

5. DISCUSSION

5.1 Limitations to the research

The first restraint of this study was the limited time frame, which only allowed to focus on two categories of decarbonisation options, namely bio-based feedstock and recycling technologies. If more time was allocated for the research, other technologies such as CCS (Carbon Capture and Storage) or electrification (e.g. heat pumps) powered by RES (Renewable Energy Sources) may have been analysed in detail. This issue, however, is partially solved by the fact that CCS, RES, and many other technologies that are non-specific for polyolefins, have been already included in the MIDDEN dataset by other researchers over the last two years. The MIDDEN report on the PVC industry, for example, evaluates a cost of $150 \notin$ /ton CO₂ captured and stored (Semeijn & Schure, 2020), while the total abatement potential for the Dutch petrochemical industry is estimated to be 11.4 Mton per year (VNCI, 2018).

There were also two barriers concerning the gathering of data. On one side, polyolefins producers are reluctant to share all the information about the technologies adopted in their production sites, the energy consumption, and their plans for the future. Although they agreed to share some of the data for the MIDDEN dataset, they did not agree to show it to the public, and they kept the details of their future strategies hidden. This lack of transparency led to some missing data (e.g. the yearly energy consumption of one production site), which had to be estimated using the available information (e.g. installed capacity and technologies adopted), attaining an approximation to reality. On the other side, many decarbonisation options are still under development, making it difficult to get accurate data regarding their techno-economic and environmental indicators. Especially when analysing thermochemical recycling processes, it is currently difficult to assess the feasibility and convenience of their deployment. Hopefully, however, more research and pilot-projects will make the performance improve in the next years, meaning that the data presented in this thesis still has value as a "pessimistic" scenario.

Finally, the scope of this research is the producers' side only, completely ignoring the consumers' side. This is due to the nature of the MIDDEN project, which is focused on finding decarbonisation options for the Dutch industrial sector, but it is clear the supply and demand influence each other, and that acting on consumers' behaviour would have a great impact on the amount of carbon emissions caused by polyolefins. It is thus recommended for future studies to include products regulation and consumers' awareness within their research boundaries. In the last few years, the demand for polyolefins in Europe has been stagnating, and the European Union is working on measures to further reduce the consumption of single-use plastics (e.g. banning disposable cutlery and plates), but the growth of Asian, Middle-Eastern, and African countries may keep the global demand (and consequently the supply) steady. Either way, studying the international market and forecasting the future of plastic production would be helpful to guide the change of the Dutch polyolefins industry.

5.2 Impact on the results on the future of the Dutch polyolefins industry

Even considering all the aforementioned limitations, this study is considered a valuable contribution to the MIDDEN project. The results are supported by both scientific literature and interviews with the polyolefins producers, meaning that they are as precise and relevant as possible to the situation of the Dutch industry. The Netherlands can thus use the MIDDEN dataset to develop decarbonisation strategies and achieve its national targets for 2050, in accord with the "klimaat-akkord".

This research also highlights the need for economic support for decarbonisation options such as biobased feedstock, which is not competitive unless a carbon price between 150 and 400 \notin kg CO₂-eq is applied to the market. The price range is wide due to the influence of the recycling rate: the more plastic waste is recycled and kept in the circular economy loop, the more it will be the abatement potential of bio-based sources, thanks to the absorption of CO₂ during the crops' growth. Import taxes are another factor to consider when evaluating the feasibility of bio-based feedstock, as producing ethanol in Europe is currently 50% more expensive than producing in abroad (e.g. Brazil). If the tariffs were lowered and the Netherlands could import cheap bio-ethanol from abroad, the cost of biopolyolefins could be almost 30% lower. Cost-learning and innovation will probably decrease the cost of bio-polyolefins while scaling-up, but regulations and financial help (e.g. subsidies, reduced taxes, public investments) are essential to foster the initial deployment of these technologies in the Netherlands.

When considering circular economy options, it appears that collaboration between companies is crucial, as one plant's waste can become another plant feedstock. A good example in the Netherlands is the pilot project between Renewi (a plastic recycling company), Plastic Energy (holder of a patented thermochemical conversion process) and SABIC (a polyolefins producer), which is only possible thanks to the combination of technical expertise. The knowledge of the MIDDEN reports and dataset provides valuable tools for the Dutch government and industry to foster collaboration and innovation, and eventually re-shape the sector to fight the global climate change.

6. CONCLUSION

This research started with the goal of answering the research question: "What is the current state of polyolefins production in the Netherlands, and what are the potential decarbonisation options to mitigate its impact on climate change?". The first step was to investigate the yearly consumption of polyolefins in the Netherlands, which altogether is equal to over 1,000 kt, or around 62 kg per capita. The analysis of the Dutch polyolefin market revealed that over two-thirds of LDPE (Low-density Polyethylene) and LLDPE (Linear Low-density Polyethylene) is used by the packaging sector, while HDPE (High-density Polyethylene) and PP (Polypropylene) have a more diversified market, still dominated by packaging applications but also including the automotive, construction and household sectors. Moving to the Dutch industry, the production sites of Dow in Terneuzen (Zeeland), SABIC in Geleen (Limburg), and Ducor in Rozenburg (South-Holland) have been investigated. The aggregate polyolefins production of these three manufacturers is 2,570 kt/year, which is equal to 8.5% of the total production of polyolefins in Europe, while their electricity consumption is equal to 1.2% of all the Dutch electricity. Dow, SABIC, and Ducor adopt four main technologies to produce polyolefins: highpressure polymerisation in a tubular reactor, solution and slurry polymerisation in a fluidised bed reactor, and gas phase in a loop reactor, with the GHG emissions ranging from 0.20 to 0.32 kg CO₂eq/kg of product.

The final part of the thesis concerned the techno-economic and environmental analysis of decarbonisation options for the Dutch polyolefins industry, focusing on bio-based feedstock and innovative recycling technologies. Olefin monomers such as ethylene and propylene can be produced from the dehydration of bio-ethanol, which can be made from the fermentation of sugar-rich crops (e.g. sugar beets). The resulting bio-plastics have the same properties of the fossil-based ones, but the overall carbon emissions are way lower, as the crops absorb CO₂ while they grow. Bio-based olefins can also be produced from the gasification of bio-waste from forestry, agriculture, the paper industry, and the food industry (e.g. cooking oils). The same process can be applied to mixed plastic waste, even in combination with biomass, although the technology is not yet ready for large scale deployment. Other recycling technologies still under innovation are dissolution (also called purification), which directly recycles the olefin polymer, and pyrolysis, which thermochemically splits the plastic waste into its monomer components. Both processes are considered promising by industry and experts and could contribute to dealing with the huge problem of plastic waste, but they are not expected to be commercially ready before 5 to 10 years. Therefore, it still needs to be proven if they are the best options, or if more sustainable end-of-life processes will be discovered and developed.

As energy can be produced from a multitude of renewable sources, the final recommendation is to focus on the material side of the plastics issues and stimulate a plastic-to-plastic circular economy loop that recycles as much olefin monomers as possible, relying on bio-based technologies to satisfy the demand for virgin polyolefins. On top of reducing the impact of the industry on the global climate change and the diffusion of plastic waste in the natural environment, this model has the additional benefit of reducing our dependency to oil and natural gas. Of course, the full decarbonisation of the Dutch polyolefins industry can only be achieved by collaborating together and deploying all the other available decarbonisation options, such as energy efficiency, heat recovery, green hydrogen, and industrial heat pumps. Many other reports confirm the importance of integrating multiple decarbonisation options when developing a strategy for the industrial sector, with the main technologies being bio-based materials, electrification powered by renewable sources, and CCS (Boulamanti & Moya Rivera, 2018; McKinsey&Company, 2018; VNCI, 2018). As most of the emissions are caused by the production of feedstock and by the end-of-life of the products, though, this research has focused on the decarbonisation technologies shown in Figure 38, which presents a possible

integrated model between bio-based olefin productions and recycling of the plastic waste. In conclusion, this thesis has achieved its goals of answering the research questions, and the knowledge collected will hopefully be used to provide valuable insight to the scientific community and the Dutch government, leading the national decarbonisation strategies and re-shaping the polyolefins industry.



Figure 38: Combination of decarbonisation options for the Dutch polyolefins industry. The blocks connected with dotted lines (energy efficiency and renewable energy supply) are the improvements that could potentially be applied to every other industrial process

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