THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

Defossilization of Refineries and Petrochemical Industries

Towards a Framework for Early-Stage Screening of Defossilization Pathways

JUDIT FORTET CASABELLA

Department of Environmental and Energy Sciences

CHALMERS UNIVERSITY OF TECHNOLOGY

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Acknowledgments and similar personal statements in this thesis reflect the author's own views.

Department of Environmental and Energy Sciences
Chalmers University of Technology
SE-412 96 Gothenburg
Sweden
Telephone +46(0)31-772 1000

Cover: Watercolor painting painted by the author, illustrating the transition towards defossilization of refinery and petrochemical industries, inspired by the research conducted in this thesis. This painting reflects the idea of shifting refineries and petrochemical industries from a fossil-based production toward the use of recycled (waste), biogenic (forest) and electrical (grid) feedstocks.

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Abstract

Defossilization of refineries and petrochemical industries requires replacing virgin fossil carbon feedstock with alternative carbon sources to produce carbon products that are hard to substitute, such as plastic building blocks (e.g., olefins) and transport fuels for hard-to-electrify applications (e.g., sustainable aviation fuel (SAF)). However, alternative carbon feedstock, such as waste and biomass, is heterogeneous and contains high levels of oxygenates, contaminants and unsaturated compounds, increasing process complexity, hydrogen demand and uncertainty in production pathway selection.

This thesis contributes to the development of an early-stage screening framework for defossilization technologies for refineries and petrochemical industries using thermodynamic analysis under technical, feedstock, and regulatory constraints. The framework evaluates feedstock carbon recovery potential, energy and exergy efficiency, and implications of European Union (EU) regulatory frameworks. The analysis includes two case studies focused on (i) SAF production through methanol as an intermediate fuel, focusing on production of renewable fuels of non-biological origin (RFNBO), and (ii) production of olefins and BTX for plastic through thermochemical recycling of waste using a fluidized bed steam cracking technology (FBSC) with Fischer-Tropsch synthesis for carbon recovery.

The results show that a high level of carbon recovery in FBSC systems is necessary to reduce on-site CO_2 emissions and improve resource efficiency. Increasing carbon conversion to plastic building blocks from 52% (without carbon recovery) to 90% (with full carbon recovery) is feasible while maintaining the overall energy efficiency between 60–70%. This corresponds to 30 percentage points higher energy efficiency than process pathways based on combustion in waste-to-energy plants with carbon capture and utilization. In contrast, current RFNBO requirements favor SAF production based on energy-depleted carbon sources (CO_2), resulting in an exergy efficiency of 58%, 10–26 percentage points lower than pathways based on energy-containing carbon feedstock.

The thesis highlights the importance of flexible technologies that can convert heterogeneous carbon feedstocks into essential carbon products. The proposed framework supports comparison of alternative production pathways to help decision makers identify key limitations related to carbon efficiency, energy efficiency, exergy losses and costs during early-stage technology screening.

Keywords: *industrial defossilization*, *alternative carbon feedstock*, *carbon recovery*, *exergy analysis*, *renewable fuel of non-biological origin (RFNBO)*, *sustainable aviation fuel (SAF)*, *olefins*, *thermochemical recycling*.

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Resum Simplificat

Desfossilitzar les refineries i la indústria petroquímica requereix substituir el petroli cru per fonts alternatives de carboni, com els residus i la biomassa, per continuar produint productes difícils de substituir, com els components bàsics dels plàstics (olefines) i combustibles per al transport difícil d'electrificar, com el combustible d'aviació sostenible (SAF). Tanmateix, aquestes fonts alternatives són heterogènies i contenen compostos oxigenats i insaturats que augmenten la complexitat dels processos i la demanda energètica. A més, la manca de tecnologies madures i la incertesa dels marcs reguladors dificulten l'avaluació de noves vies de producció basades fonts alternatives de carboni.

Aquest estudi proposa les bases per desenvolupar un marc d'avaluació preliminar de tecnologies de desfossilització per a refineries i la indústria petroquímica mitjançant una anàlisi termodinàmica simplificada. El marc considera aspectes tècnics, disponibilitat de matèries primeres i regulacions de la Unió Europea. L'anàlisi inclou l'estudi de dos casos (1) la producció de combustible d'aviació sostenible, utilitzant metanol com a producte intermediari, amb especial atenció als requisits europeus per als combustibles renovables d'origen no biològic (RFNBO), i (2) la producció d'olefines i compostos aromàtics per a plàstics a partir de residus mitjançant reciclatge termoquímic amb un cracker de vapor de llit fluïditzat (FBSC) i recuperació de carboni amb síntesi Fischer-Tropsch.

Els resultats mostren que una alta recuperació de carboni és essencial en sistemes FBSC per reduir emissions de CO_2 i millorar l'eficiència dels recursos. Augmentar la conversió de carboni cap a productes plàstics des del 52% al 90% manté una eficiència energètica entre el 60 i el 70%, 30 punts superior a altres vies basades en combustió de residus amb valoració energètica i captura de carboni. En canvi, la producció de SAF compatible amb RFNBO necessita utilitzar CO_2 com a font de carboni, fet que redueix l'eficiència exergètica del procés (58%) en comparació d'altres vies de producció que arriben al 84%.

L'estudi destaca la importància de instal·lar tecnologies flexibles capaces de processar diferents tipus de residus i biomassa per produir productes difícils de substituir. El marc proposat permet comparar un ampli ventall de vies de producció i identificar limitacions clau relacionades amb l'eficiència energètica, l'ús del carboni i els costos en fases inicials de desenvolupament tecnològic.

Paraules clau: *refineries i indústria petroquímica, desfossilització industrial, fonts alternatives de carboni, recuperació de carboni, marcs reguladors, anàlisi exergètica, combustibles renovables d'origen no biològic (RFNBO), combustible d'aviació sostenible, olefines, reciclatge termoquímic.*

List of publications

The thesis is based on the following appended papers, which are referred to in the text by their assigned Roman numerals:

I. J. Fortet Casabella, J. Beiron, T. Roshan Kumar, M. Sundén, S. Harvey & H. Thunman. "Do RFNBO Regulations Promote Exergetic Efficiency in Sustainable Aviation Fuel Production? A Comparative Exergoeconomic Analysis", *Energy Conversion and Management: X* (2026), https://doi.org/10.1016/j.ecmx.2026.101948.

II. J. Fortet Casabella, J. Beiron, T. Roshan Kumar, V.R. Reddy Marthala, S. Harvey & H. Thunman. "Towards full carbon recovery in a fluidized bed steam cracker for thermochemical recycling of plastic waste". Working manuscript.

Authors contributions

Judit Fortet Casabella is the principal author of **Papers I–II** and performed the modeling, method development, data processing, analysis and writing of the papers. Dr. Johanna Beiron is a co-supervisor of the academic work and contributed to method development, discussions and editing of **Papers I–II**. Dr. Tharun Roshan Kumar contributed to method development, discussions and editing of **Papers I–II**. Dr. V.R. Reddy Marthala contributed to the discussions for the modeling work of **Paper II**. Dr. Moa Sundén contributed to the discussions of **Paper I**. Professor Henrik Thunman is the main academic supervisor and contributed with discussions and editing of **Papers I–II**. Professor Simon Harvey contributed to the discussions and editing of **Papers I–II**.

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Declaration of the Use of Generative AI

During the preparation of **Papers I–II** and the introductory essay, the generative AI *ChatGPT* was used in order to improve the language and readability. It did not create original ideas, scientific data or conclusions. After using this tool/service, the text has been reviewed and edited as needed. The main author takes full responsibility for the manuscript's accuracy and integrity.

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I am very grateful to all my friends, in Sweden, Catalonia or wherever in the world, for supporting me and handling my ups and downs. For all the hikes, bars, trips, dances and unforgettable moments along the way and the ones to come.

I would also like to thank *dance*. Dancing has always been part of my life, but during this PhD, I truly understood how necessary it is for me.

To my family (mama, papa, and Anna), thank you for all the constant support and trust in me, always showing that you are very proud of my achievements and of who I am becoming. Special thanks to my sister for being herself and showing me that everything is possible.

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Judit Fortet Casabella
Göteborg, 17th May 2026

Abbreviations

ASF	Anderson-Schulz-Flory	**KPI**	Key Performance Indicators
ASR	Automotive Shredded Residues	**LHV**	Low Heating Value
ASU	Air Separation Unit	**MEA**	Monoethanolamine
ATR	Autothermal Reformer	**MPW**	Mixed Plastic Waste
BECCS	Bioenergy with Carbon Capture and Storage	**MtG**	Methanol-to-Gasoline
BFB	Bubbling Fluidized Bed	**MtJ**	Methanol-to-Jet
BTX	Benzene Toluene Xylene	**MtO**	Methanol-to-Olefins
CAPEX	Capital Expenditures	**OPEX**	Operational Expenditures
CBAM	Carbon Border Adjustment Mechanism	**PE**	Polyethylene
CCGT	Combined Cycled Gas Turbines	**PEM**	Proton Exchange Membrane
CCS/U	Carbon Capture and Storage/Utilization	**POX**	Partial Oxidation
CDR	Carbon Dioxide Removal	**PSA**	Pressure Swing Adsorption
CFB	Circulating Fluidized Bed	**RCF**	Recycled Carbon Fuels
CHP	Combined Heat and Power Plant	**RED**	Renewable Energy Directive
DACCS	Direct Air Carbon Capture and Storage	**RED III**	Renewable Energy Directive
DFB	Dual Fluidized Bed	**RFNBO**	Renewable Fuels of Non-Biological Origin
EED	Energy Efficiency Directive	**r-WGS**	Reverse Water Gas Shift
ETS	Emission Trading System	**SAF**	Sustainable Aviation Fuel
EU	European Union	**SC**	Steam Cracker
FBSC	Fluidized Bed Steam Cracker	**SMR**	Steam Methane Reformer
FCC	Fluid Catalytic Cracker	**SOEC**	Solid Oxide Electrolyzer Cell
FEED	Front-End Engineering Design	**TRL**	Technology Readiness Level
FT	Fischer-Tropsch	**UCO**	Used Cooking Oil
GHG	Greenhouse Gas	**WGS**	Water Gas Shift
GHR-POX	Gas Heated Reformer-Partial Oxidation	**WtE**	Waste to Energy

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1. Introduction and Aim

To align with climate-neutrality goals to limit global warming to 1.5 °C by 2050 (IPCC, 2018), future production of basic chemicals must achieve substantial reductions in greenhouse gas (GHG) emissions and dependence on virgin fossil resources, primarily crude oil. Refineries and petrochemical industries rely on carbon both as an energy carrier and as a material feedstock for the production of essential carbon products[1], i.e., fuels, chemicals, and plastics, that cannot be substituted by direct electrification. Consequently, the sector cannot *decarbonize* by only eliminating carbon use but must instead replace virgin fossil-based carbon with alternative carbon sources, a transition commonly referred to as *defossilization*[2] (Gabrielli et al., 2020). This transition increasingly relies on alternative carbon feedstocks, typically categorized as biomass, waste-derived feedstocks, recycled carbon streams, and CO_2-derived intermediates (Manalal et al., 2025; Renewable Carbon Initiative, 2025), which are often heterogeneous and resource-constrained (Slade et al., 2014). This shift is crucial since embedded carbon accounts for approximately two thirds of the greenhouse gas (GHG) emissions of most building blocks in the chemical industry (Kähler et al., 2022). The efficient use of these limited circular and renewable carbon resources is a central challenge for achieving both climate neutrality and resource efficiency in the chemical industry.

Existing refineries and petrochemical infrastructure are optimized for processing large and continuous flows of relatively homogeneous virgin fossil feedstocks of a specific feedstock quality. Hence, repurposing this infrastructure for alternative carbon feedstock introduces a system mismatch. Waste and biogenic-derived feedstocks are geographically constrained, highly heterogeneous, and of unstandardized quality, containing elevated levels of oxygenates, contaminants, and unsaturated compounds. Processing these feedstocks in existing industrial infrastructure requires additional upgrading and pretreatment steps, leading to high hydrogen and energy demands. Furthermore, contaminants may lead to operational challenges throughout the downstream system, such as catalyst deactivation and coke formation.

A wide range of emerging technologies[3] have been developed to convert and upgrade alternative carbon feedstocks into fuels and plastic building blocks. These include conversion technologies, e.g., through thermochemical recycling, and carbon recovery processes based on, e.g., Fischer-Tropsch or Methanol-to-X synthesis (Bube et al., 2024; Manalal et al., 2025). These technologies differ significantly in terms of carbon recovery[4], energy efficiency, external hydrogen demand, product flexibility, GHG emissions, and economic performance.

[1] Essential carbon products are defined in this work as products which are currently produced using virgin-fossil feedstock and that are considered hard to replace with electricity, mainly focused on the transportation and material sector: maritime fuel, aviation fuel, olefins and BTX (building blocks for plastic production).

[2] "Defossilization" is used throughout the thesis to indicate the transition from virgin-fossil-based feedstock to alternative carbon sources while minimizing plant-level fossil CO_2 emissions, without eliminating the carbon embedded in the final products.

[3] Technology readiness level (TRL) < 7.

[4] In this work, carbon recovery is defined as the yield of feedstock carbon converted to essential carbon products (targeted products).

Existing assessment approaches often require detailed process simulations, which limit their use for rapid comparison of multiple pathways at early stages (Bube et al., 2024; Manalal et al., 2025; Roshan Kumar, 2025). Although detailed process models are valuable for evaluating specific technologies and sites, their level of detail is not always required for a broad early-stage comparison of multiple defossilization pathways. A simplified screening approach is therefore needed to compare pathways consistently in terms of carbon recovery, energy and exergy efficiency, cost, and regulatory constraints.

To unlock alternative carbon feedstock for carbon-dependent industries such as refineries and petrochemicals, energy production systems must decarbonize to shift away from using carbon-containing resources as fuels. Producing carbon-free[5] electricity enables the substitution of carbon as an energy carrier, making carbon-rich waste or biomass available for material production. Additionally, producing intermediates such as hydrogen via carbon-depleted routes, for example, electrolysis powered by carbon-free electricity, is desirable to avoid the consumption of carbon resources and minimize carbon emissions. Combustion of fuels for energy production in, for example, combined heat and power (CHP) plants releases CO_2, with no energy value. Carbon capture and utilization (CCU) can be applied to capture the CO_2 and produce essential carbon products by the addition of hydrogen, leading to high hydrogen consumption and thus, high electricity demand.

Regulatory frameworks play a central role in shaping the competitiveness and deployment of defossilization technologies. In the European Union (EU), regulatory frameworks such as the Renewable Energy Directive (RED III), Energy Efficiency Directive (EED), and Waste Directive affect how carbon flows, renewable energy use, emissions reductions, and waste management are accounted for, thereby influencing the attractiveness of different conversion pathways (European Commission, 2008, 2023b, 2023c). Since these regulatory frameworks prioritize different objectives, they may favor production pathways with substantially different carbon recovery, hydrogen demand, energy efficiency, and cost. Consequently, technology assessment for industrial defossilization must consider not only technical performance, but also the influence of regulatory constraints on pathway viability.

Existing studies often focus on detailed defossilization pathway simulations focusing on single technologies and isolated case studies, such as specific integration processes, feedstocks, and case studies. For example, the literature includes refinery and petrochemical cluster studies investigating the integration of alternative carbon feedstocks and carbon recovery pathways under site-specific conditions (Manalal et al., 2025; Roshan Kumar et al., 2025; Stepchuk et al., 2025), as well as comprehensive combinations of thermodynamic assessment methodologies to identify optimal defossilization strategies, using multi-criteria or exergy-based evaluation (Roshan Kumar, 2025; Silva Ortiz et al., 2024). However, existing detailed simulations are often computationally intensive and time-consuming as they are highly case-specific, and focused on individual technologies or feedstocks, limiting their applicability for broad comparative assessment during early-stage decision-making. Furthermore, limited

[5] Carbon-free electricity meaning electricity produced without direct CO_2 emissions, e.g., wind, hydro and solar power.

integration of technical performance and regulatory constraints is found in the literature, particularly regarding the implications of carbon recovery, hydrogen demand, and pathway selection under evolving regulatory frameworks (Beiron et al., 2026; Prussi et al., 2025). Consequently, there is a need for simplified early-stage screening frameworks to compare a wide range of defossilization technologies to capture key trade-offs related to resource efficiency, economics, and regulatory frameworks.

1.1 Towards a framework for early-stage production pathway screening

This work presents the initial basis of a framework that aims to enable process screening of early-stage production pathways to defossilize carbon-intensive industries under technical, feedstock, and regulatory constraints. The methodological framework is driven by the need to be resource efficient in a resource-constrained and carbon-dependent system that requires maximizing carbon recovery into essential carbon products while minimizing exergy losses. *Essential carbon products* are defined in this thesis as products that are currently produced using virgin-fossil feedstock for applications that are considered hard-to-electrify, mainly focused on the transportation and material sector: maritime fuel, aviation fuel, olefins, and BTX (building blocks for plastic production). Each production pathway for defossilization is characterized by different mass and energy flows, thereby influencing carbon flows, energy requirements, emissions, and overall economics. Therefore, the framework relies on combining known methodologies to thermodynamically group defossilization technologies to enable screening of a wide range of production pathways. External factors such as regulatory frameworks, technology maturity, and feedstock availability are discussed to evaluate the implications of the implementation of the technologies. The framework encompasses methods and Key Performance Indicators (KPI) used and discussed in Section 4 and **Paper I–II**.

1.2 Aim

The aim is to develop and apply an early-stage screening framework to refinery and petrochemical defossilization pathways. The introductory essay builds upon the appended papers to present a structured methodology and to:

- Identify and compare defossilization production pathways that can be integrated into refineries and petrochemical industries.
- Evaluate how carbon recovery affects carbon utilization, energy efficiency and exergy losses.
- Define key performance indicators for consistent early-stage technology comparison.
- Analyze whether regulatory frameworks act as drivers or barriers to the deployment of resource-efficient defossilization technologies.

The following research questions are addressed:

RQ1: How can alternative production pathways be evaluated and compared to support the defossilization of refinery systems?

RQ2: How does the integration of carbon recovery processes affect carbon utilization and energy efficiency in refinery and petrochemical systems? (Mainly discussed in **Paper II**)

RQ3: How do regulatory frameworks influence the selection and performance of defossilization pathways, particularly in terms of trade-offs between energy efficiency, carbon recovery, and economics? (Mainly discussed in **Paper I**)

1.3 Scope

This work focuses on the defossilization of virgin-fossil-based refineries and petrochemical industries, specifically steam cracker plants. The scope of this work is limited to the production of essential carbon products for society for applications that are hard-to-electrify, including monomers for plastic production (olefins and BTX) and aviation fuels. Other carbon-intensive industries, e.g., pulp and paper, steel, and cement, are outside the scope of this thesis.

Waste is considered an alternative carbon feedstock for carbon recovery. The scope of recycling analysis is limited to the thermochemical recycling techniques discussed in the appended paper. Additionally, only mixed plastic waste and biomass residues are considered in the work. Thus, for example, textiles, metals, and hazardous waste are not included.

The framework emphasizes the possibility of integrating technologies within the same industry or across industries and utilizing byproducts within the site. Waste collection, power grid infrastructure extension, and electricity supply are out of scope.

The methodology is designed for low-detail process pathway comparisons aimed at identifying key resource efficiency limitations rather than detailed process design or equipment-level evaluation. The level of detail is chosen to align with the research questions, which focus on pathway feasibility and comparative assessment. In cases where external constraints, such as regulatory limitations or feedstock availability, dominate the outcome, detailed process simulations would provide limited additional insights. Thus, the use of simplified models enables efficient technology screening and consistent comparison of transition pathways.

1.4 Outline of the thesis

The thesis consists of an introductory essay, and two appended papers. Chapter 1 introduces and contextualizes the introductory essay and appended papers, presenting the aim, the scope, and the outline of the work. Chapter 2 provides the background to this work, including studied technologies and industries, and relevant regulatory frameworks. Chapter 3 presents an overview of the proposed framework, the methodology used, key performance indicators analyzed, and introduces the case studies used in this work. Chapter 4 presents a summary of the selected results from the appended papers and discusses them within the current European Union context. Chapter 5 presents the overall conclusions. Finally, Chapter 6 contains consideration for future work.

Figure 1 illustrates the scope of the appended papers and the methodological framework presented in the introductory essay, highlighting the overlap of topics between **Paper I**, **Paper II**, and the framework for early-stage process screening. Contributions of each paper to the overall aim of the thesis:

Paper I provides an early-stage assessment of defossilization technologies through exergy analysis, including step-by-step exergy losses, and techno-economic analysis. The paper analyzes sustainable aviation fuel (SAF) production pathways to understand how they are affected by regulatory frameworks framed today in the European Union. It sets the basis for setting a framework, enabling rapid and robust screening of emerging fuel pathways to understand regulatory risks, limitations, and inconsistencies.

Paper II provides an evaluation of the addition of a carbon recovery section to valorize unconverted carbon-containing byproducts. It explores the impact of increasing carbon recovery on energy efficiency from fluidized bed steam cracking using Fischer-Tropsch (FT) synthesis as a carbon recovery section. The work builds on the analysis performed by Thunman et al (2019) which assumed a methanol-to-olefins (MTO) process. The work contributes to understanding the potential of deploying FT as a technology to enhance carbon recovery in a steam cracker plant.

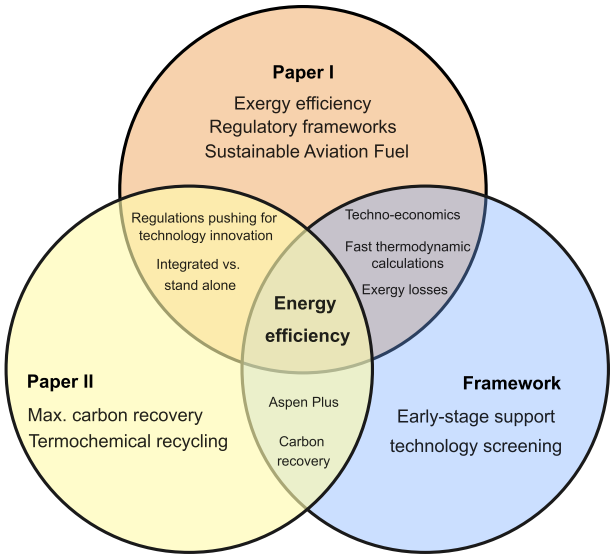

Figure 1: Scope of the introductory essay and appended papers.

2. Background

This chapter focuses on refinery and petrochemical systems, the limitations of existing fossil-based infrastructure, the conversion technologies considered in the appended papers, and the EU regulatory frameworks relevant to pathway screening. The section is divided into two main topics: Section 2.1 presents the relevant production processes in refineries and petrochemical industries, including virgin-fossil based and alternative carbon production-based, and Section 2.2 describes the main relevant regulatory frameworks for this work, specifically from the European Union.

2.1 Relevant production processes for Refineries and Petrochemical industries

2.1.1 Current set-up and limitations

Petrochemical industries and refineries have in common the use of virgin crude oil-based feedstock in their processes. Figure 2 shows how the feedstock and targeted product vary between the two industries. Refineries traditionally refine virgin crude oil using a crude oil distillation column, which separates the different crude oil fractions based on boiling points, which typically correlate with carbon numbers. Most of the distilled fractions are used for transportation fuels, while the naphtha fraction, from ~C5–C10, is sent to petrochemical industries to be cracked into chemicals or plastic building blocks, such as light olefins (ethylene and propylene) and aromatics (benzene, toluene, xylene (BTX)). Throughout the process, both industries require heat, which is produced through the combustion of byproduct streams, leading to CO_2 emissions.

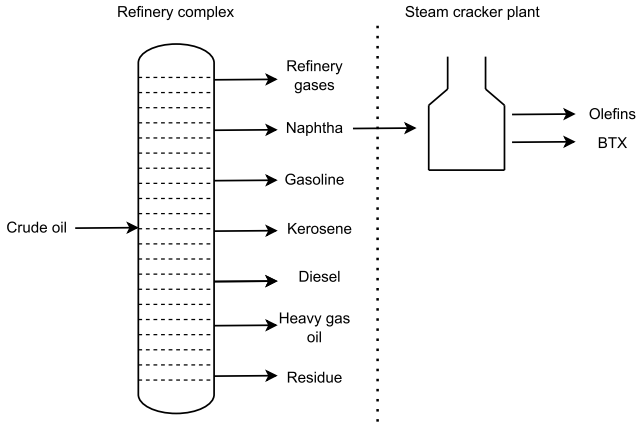

Figure 2: Refineries and petrochemicals using conventional feedstocks and products.

Current refineries and petrochemical industries are optimized for feedstock with very specific properties. For refineries, a certain crude oil type is chosen based on product fractions, which determines the proportion of light gases over residue, and on sulfur content, which determines the processing complexity for sulfur removal (Alfke et al., 2007). For steam crackers, a certain carbon range such as ethane, propane, butane, naphtha, gas oil and residue is commonly used (Zimmermann & Walzl, 2009). Specific technologies can be more flexible in terms of feedstock, for example, there are flexible steam crackers that can handle different carbon ranges, such as the Linde-Pyrocrack Furnace (Zimmermann & Walzl, 2009). The products obtained are typically characterized by very high purities and specific properties in refineries, such as boiling point, cloud point, density or viscosity (Alfke et al., 2007).

In the context of defossilization, jet fuel and plastics are particularly relevant as they represent substantial demands for carbon-containing products. In the EU, jet fuel consumption in 2022 was ~865,000 bpd (~1,740 PJ/year), of which roughly 70% was produced within the EU (~1,218 PJ/year) and the rest imported from the Middle East (Euronews, 2026; World Bank, 2024). At a national level, Sweden has a near balance between the plastic raw material production (~1046 kton/year) and consumption (~1077 kton/year), allowing assessment of the relationship between waste generation (~905 kton/year) and feedstock demand (Naturvårdsverket, 2023). Currently, ~50% of plastic waste is directed to energy recovery (Naturvårdsverket, 2023), emitting the carbon to the atmosphere.

Both industries aim to defossilize their production by substituting the feedstock used for an alternative carbon source, generally as a solid, such as biomass or waste, and to decarbonize by minimizing scope 1 emissions from the plant, through, for example, direct and indirect electrification and carbon capture and storage (CCS). Refineries and petrochemical industries have started defossilising by introducing specific biogenic feedstock. For refineries, lipid-based feedstock (vegetable oil from crops such as rapeseed, palm or soy, used cooking oil (UCO) and animal fats) and biogas (Transport and Environment, 2023), and for steam cracker plants, bio-naphtha and biodiesel mainly from UCO (De Bruycker et al., 2016). The largest producer of renewable diesel and SAF is currently Neste, located in Porvoo (Finland), Rotterdam (the Netherlands) and Singapore, with a combined production of 2.9 Mton/year based on NEXBTL technology (TRL 9) using vegetable oils (IEA Bioenergy & ARTFuels, 2020). VAROPreem in Sweden produces HVO, renewable diesel and bio-LPG from UCO and animal fats predominantly extracted from Europe, with a production capacity of 354,000 m^3/year (Preem AB, 2024). A recent expansion using Topsoe HydroFlex (TRL 9) hydrotreater technology adds 900,000 m^3/year to the production capacity (Preem AB, 2024; Topsoe, 2026a). Renewable raw materials such as bio-propane and bio-naphtha, co-produced in the HVO process, have been used in steam crackers operated by SABIC, Dow, INEOS and Borealis (Borealis AB, 2023; De Guzman, 2021). The described technologies are highly focused on lipid-based feedstock, which is limited by collection availability. In the EU, approximately 80% of the refined lipid-based feedstock is imported, mainly from countries such as China and Malaysia (Transport and Environment, 2023, 2024).

Lipid-based feedstock is highly oxygenated and olefinic. Steam cracker plants and refineries are limited with respect to their capability to process olefinic and oxygenated feedstocks (Alfke et al., 2007; Kusenberg et al., 2022) since such feedstocks typically lead to coking and contamination of the downstream equipment, requiring downstream process steps, such as hydrogenation and deoxygenation, to decontaminate the feed, leading to high hydrogen demand[6]. Hydrogen production for decontamination is one of the largest sources of CO_2 emissions in refineries that produce biofuels (Biermann et al., 2022; Rambech, 2025).

[6] 9 moles of H_2 are required to saturate each mole of triolein ($C_{57}H_{104}O_6$) and remove all the oxygen (olefinic saturation, 3 moles, and oxygen removal, 6 moles). From each mole of triolein, 3 moles of n-paraffins and 1 mole of propane are obtained.

Thus, the use of other alternative carbon feedstocks may lead to the following limitations:

- Technologies and processes in place in the refinery and petrochemicals are highly optimized for specific quality feedstock, which does not match the homogeneity of waste streams, requiring new technologies that can handle heterogeneous feedstock.
- Biomass contains large concentrations of oxygen, leading to contamination in the downstream processes, implying that oxygen needs to be removed before entering the refinery or have technologies that can handle the oxygen levels.
- Waste and biomass contain high concentrations of unsaturated chains, leading to large hydrogen demands for saturating them; hence, a large capacity for low-emission hydrogen production units is required, e.g., electrolysis using renewable electricity.
- Alternative carbon feedstock is limited, meaning that technologies that achieve high resource efficiency or that can recover the carbon through other processes are of interest.
- Renewable electricity is a limited source, especially due to low capacity in the grid connections to the refinery. Thus, energy efficiency processes are key to limiting its use.

2.1.2 Alternative carbon processing technologies studied

Processes capable of handling heterogeneous and contaminated solid feedstocks with high resource efficiency are required to enable efficient carbon recovery, minimize emissions, and enhance product yield. Examples of alternative carbon feedstock that are heterogeneous are municipal solid waste, mixed plastic waste or biomass waste. A two-step conversion or a direct conversion can be used to process the feedstock. A two-step conversion is typically applied for the conversion of heterogeneous feedstock, where step (i) is to convert any hydrocarbon molecule to liquid, similar to pyrolysis oil, or gas, similar to syngas, and step (ii) is to convert syngas into essential products through Fischer-Tropsch (FT) or alcohol-to-olefins (such as MTO) or pyrolysis oil via steam cracking or distillation of the fractions. A direct conversion is focused on the conversion of solids or liquids to gaseous molecules of high value through technologies such as fluidized bed steam cracking (FBSC) or pyrolysis. The different conversion pathways analyzed in the introductory essay are discussed in the following subsection for a clear understanding of the main results. The thesis focuses on the technologies and production processes investigated in the appended papers.

Primary conversion technologies

Figure 3 shows combustion, reforming and gasification technologies for hydrocarbon feedstock, including their reactor set-up, product produced and oxygen supply. Similarly, Table 1 contains the process conditions for the different technologies utilized in this work.

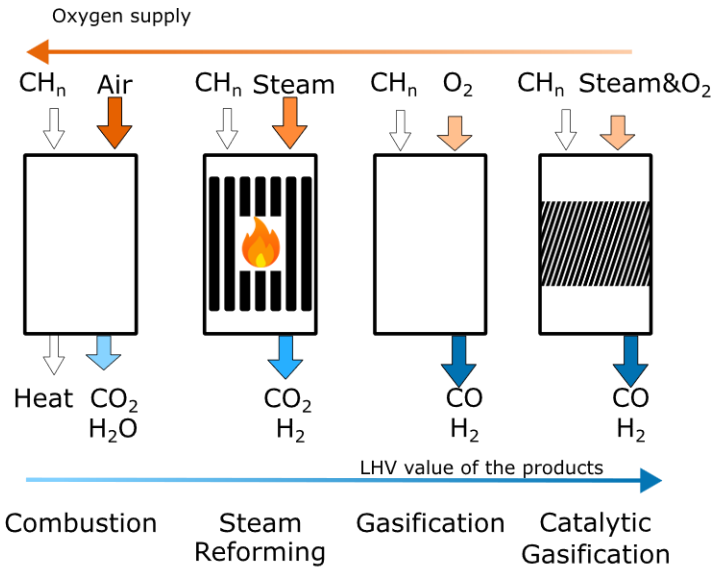

Figure 3: Broad conversion technologies to convert hydrocarbon feedstock into CO_2, H_2 and syngas (CO and H_2). The technologies are combustion, steam reforming, gasification and catalytic gasification. Inspired from (Ribun et al., 2023).

Combustion is the simplest technology in which air, or pure oxygen, is fed into a combustion chamber to produce energy, following reaction R1. Combustion is widely used to produce heat and electricity in, for example, combined cycled gas turbines (CCGT) when using gaseous fuels, and combined heat and power plants (CHP) when using solid feedstock, such as coal, waste or biomass. Heat, electricity and flue gases are obtained through the process. CO_2 can be captured using carbon capture and storage (CCS) from the flue gases either with a benchmark MEA post-combustion carbon capture unit or using pure oxygen as a combustion medium to minimize contaminants in flue gas, leading to CO_2 and H_2O. CO_2 does not contain usable chemical energy thus, its further synthesis to hydrocarbons requires the addition of hydrogen. Some CCS projects are undergoing construction, such as Hafslund CHP plant in Oslo (Norway), which uses the technology from Aker Solutions AS and SLB Capturi Norway AS, aiming to capture 350 kton/year of CO_2. The facility is expected to be operational by 2029 (Hafslund, 2025). Additionally, Stockholm Energi (Sweden) is aiming to capture 800 kton CO_2/year using the technology CapsolEoP provided by Capsol Technologies using hot potassium carbonate, and it is planning to be operational in 2028 (Capsol Technologies, 2026; Stockholm Exergi, 2026).

An *electrolyzer*, such as alkaline, proton exchange membrane (PEM) or solid oxide electrolyzer (SOEC), can be used to produce low-carbon hydrogen using renewable electricity. The alkaline and PEM electrolyzers operate at low temperatures (60–80 °C), leading to efficiencies of electricity-to-hydrogen of ~58%, while SOEC operates at higher temperatures (above 650 °C), where its efficiency strongly depends on system integration and boundary definitions (Beiron & Thunman, 2024). Topsoe's SOEC reports values up to 97% electricity-to-hydrogen efficiency under specific system assumptions, including steam supply at 2 bar (Topsoe, 2024a), while Sunfire reports approximately 89% efficiency under a different system boundary definition (Sunfire, 2025). These values are therefore not directly comparable, as differences may reflect both technological performance and the chosen system integration and efficiency definitions. A facility in Herning (Denmark) is in the installation and ramp-up phase toward

full stack production, designed to manufacture 500 MW of electrolyzer per year using Topsoe SOEC technology, with potential scaling to 1.4 GW per year by 2031 (Topsoe, 2024a).

Hydrocarbon *steam reforming* is typically performed using similar technology, such as a steam methane reformer (SMR), where steam is added in order to produce a mix of syngas, which can be shifted through water gas shift (WGS) to increase the production of hydrogen (following reaction R3), leading to H_2 and CO_2, according to reaction R2. SMR reactors are widely available at refineries for hydrogen production, accounting for the largest share of hydrogen generation and one of the main sources of CO_2 emissions in refineries (Biermann et al., 2022; Rambech, 2025). Repurposing such systems could constitute an attractive opportunity to valorize fuel gas from refineries.

A *gasification*-like system is typically performed in a pure-oxygen constrained environment where syngas is produced in a catalytic (autothermal reformer, ATR) or a non-catalytic (partial oxidation, POX) system, both following reactions R4–R5. Other reactor types, such as fluidized bed (FB), can reform solid feedstock into syngas (Thunman, Gustavsson, et al., 2019). Similarly, a system composed of two reactors in series, a gas-heated reformer and a partial oxidation (GHR-POX), can be used to achieve autothermal (Matthey, 2022). The GHR converts the hydrocarbons into mainly CH_4 and H_2 using steam. The heat required for the endothermic reaction in the GHR is supplied by the high-temperature products from the POX reactor, which converts the output of the GHR into CO and H_2 using pure oxygen. More information in **Paper II**.

When producing syngas, a syngas ratio of 2–2.2 (Eq. 1) is typically required for the production of hydrocarbons through FT and MTO. Pure oxygen can be produced from a cryogenic air separation unit (ASU) and as a by-product from the SOEC hydrogen electrolyzer, although the achievable oxygen purity depends on the stack configuration and sweep-gas strategy (Axelsson et al., 2024).

$$CH_4 + 2\,O_2 \rightarrow 2\,H_2O + CO_2 \qquad \Delta H = -800\ kJ/mol\ C \qquad \text{(R1)}$$

$$CH_4 + 2\,H_2O \rightarrow 4\,H_2 + CO_2 \qquad \Delta H = 164\ kJ/mol\ C \qquad \text{(R2)}$$

$$CO + H_2O \leftrightarrows CO_2 + H_2 \qquad \Delta H = -41\ kJ/mol\ C \qquad \text{(R3)}$$

$$CH_4 + \frac{1}{2}O_2 \rightarrow 2\,H_2 + CO \qquad \Delta H = -36\ kJ/mol\ C \qquad \text{(R4)}$$

$$CH_4 + H_2O \rightarrow 3\,H_2 + CO \qquad \Delta H = 205\ kJ/mol\ C \qquad \text{(R5)}$$

$$syngas\ ratio = \frac{H_2 - CO_2}{CO + CO_2} \qquad (1)$$

Table 1: Technology data assumed throughout the work

Technology	Parameters	Value	Comment/Reference
Combined Cycle Gas Turbine (CCGT)	Total exergy efficiency	0.88 $MW/MW_{fuel,LHV}$	Assuming steam production at 300 °C and 60 barg, expanded with a steam turbine to 4 barg (Danish Energy Agency, 2025)
	Energy efficiency steam production	0.43 $MW_{steam}/MW_{fuel,LHV}$	
Autothermal Reformer (ATR)	Temperature, Pressure	900–1,000 °C, 28 bar	(Halabi et al., 2011)
	Conversion rate	90%	
Steam Methane Reformer (SMR)	Temperature, Pressure	700–900 °C, 28 bar	(Y. Ding & Alpay, 2000)
	Conversion rate	80%	
Carbon Capture	CO_2 capture efficiency	90%	(Garðarsdóttir et al., 2018)
	Reboiler duty	3,600 kJ/kg of CO_2	
	Steam pressure provided to the reboiler	2 barg	
Pressure Swing Adsorption (PSA)	Electricity demand	5 kWh/kg H_2	(Song et al., 2015)
Gas Heated Reformer–Partial Oxidation (GHR-POX)	GHR: Temperature, Pressure	T = 975 °C, P = 20 bar	(Matthey, 2022)
	POX: Temperature, Pressure	T = 1,200–1,400 °C, P = 20 bar	
	POX: Oxygen to methane ratio (O_2/CH_4)	1.5	
Solid Oxide Electrolyzer Cell (SOEC)	Efficiency electrolyzer (Electricity to hydrogen)	$75\%_{LHV}$	(Danish Energy Agency, 2024)
	Steam demand for SOEC	5.21 kWh/kg H_2	Stoichiometric value (9 ton low pressure steam/ton H2) at 2 barg (Sunfire, 2025)
Cryogenic Air Separation Unit (Cryogenic ASU)	Electricity for cryogenic ASU unit	0.2 $kWh_{el}/kg\ O_2$	(Thunman, Berdugo Vilches, et al., 2019)
Methanol synthesis (MeOH synthesis)	Temperature	250 °C	With high recycling rate (Borisut & Nuchitprasittichai, 2019)
	Pressure (R8)	50 bar	
	Pressure (R9)	100 bar	
	Conversion rate (R8 and R9)	99%	
Methanol-To-Olefins (MTO)	Temperature, Pressure	400–550 °C, 1–3 bar	UOP/Hydro MTO (Gogate, 2019; Jasper & El-Halwagi, 2015)
	Conversion methanol to ethylene and propylene	80%	
Low-Temperature Fischer-Tropsch (LT-FT)	Temperature, Pressure	220 °C, 20 bar	With internal recycling of unconverted gases (Pondini & Ebert, 2013)
	Chain growth probability (α)	0.92	
	CO conversion	90%	
High-Temperature Fischer-Tropsch (HT-FT)	Temperature	290–360 °C	(De Klerk, 2011)
Fluidized Bed Steam Cracking (FBSC)	Temperature, Pressure	700–800 °C, 1 bar	(Mandviwala et al., 2024)
	Carbon-to-products	60–70%	Using PE as feedstock (Thunman, Berdugo Vilches, et al., 2019)
	Energy efficiency	~65%	

Carbon recovery technologies

Syngas and/or CO_2+H_2 can be used for the production of alcohol, olefins, methane and other larger hydrocarbons through different synthesis processes. This thesis considers carbon recovery technologies such as methanol production, methanol-to-olefins or methanol-to-jet, and Fischer-Tropsch. Table 1 contains the process conditions of the technologies described.

Methanol can serve as an intermediate product for the production of olefins, through methanol-to-olefins, and of longer hydrogenated hydrocarbon chains, through methanol-to-jet (MTJ). Methanol can be synthesized through direct synthesis, from CO_2 and H_2 according to reaction R6, (Borisut & Nuchitprasittichai, 2019; Marlin et al., 2018) or conventional synthesis, from syngas according to R7 (Dalena et al., 2018). The endothermic heat of reaction (R6) is assumed to be sufficient to separate the methanol from the obtained water in a distillation column (Battaglia et al., 2021; Halager et al., 2021; Johansson, 2013).

$$CO_2 + 3\,H_2 \rightarrow CH_3OH + H_2O \qquad \Delta H = -50\ kJ/mol\ C \qquad \text{(R6)}$$

$$CO + 2\,H_2 \rightarrow CH_3OH \qquad \Delta H = -91\ kJ/mol\ C \qquad \text{(R7)}$$

Methanol-to-olefins is conducted in a dual fluidized bed reactor, converting methanol into a ZSM-5 catalyst-filled reactor to obtain C2–C5 light olefins. The light olefins can be oligomerized to produce paraffins, aromatics and higher olefins (Elwalily et al., 2025). The UOP/Hydro MTO process can achieve a high yield of ethylene and propylene, where the specific ethylene to propylene ratio depends on the process parameters (Jasper & El-Halwagi, 2015). It uses a SAPO-34 zeolite type in an FB reactor, obtaining DME as a byproduct, which is recovered (Jasper & El-Halwagi, 2015). UOP/Hydro MTO (TRL 8–9) is provided by Honeywell, and it is commercially available at Wison Clean Energy located in Nanjing (China), designed for a capacity of 300 kton/year of ethylene and propylene (Honeywell UOP, 2026; Wison Clean Energy, 2013).

MTJ production is based on the oligomerization of olefins, and depending on process conditions, it is possible to change the chain growth probability, limiting it to larger hydrocarbon chains, jet fuel, or shorter, e.g., gasoline (Eyberg et al., 2024). Technologies are being commercialized, such as the methanol-to-gasoline (MTG) process from ExxonMobil, which has been studied since 1970s, and it is now used in the Haru Oni Project in Chile (ExxonMobil, 2026). The MTJet process, developed by Topsoe, is currently in the pre-construction phase in Houston (USA), where it is designed to process 30,000 ton/day of methanol (Nacero, 2023; Topsoe, 2026b).

$$CH_3OH \rightarrow [CH_2] + H_2O \qquad \text{(R8)}$$

The *Fischer-Tropsch* process can synthesize syngas to hydrocarbons ranging from shorter chains, such as methane, to longer chains, such as waxes, according to reaction R9. The intermediate product produced is synthetic crude oil, also known as syncrude, which can serve as feedstock for fuel production or for reforming further in a steam cracker plant. Figure 4 shows the Anderson-Schulz-Flory (ASF) chain growth probability (α) distribution,

differentiating between high-temperature (HT) and low-temperature (LT) FT. Chain growth probability is defined by operating temperature and pressure. Iron-based and cobalt-based catalysts can be used for this reaction, iron-based are mainly employed for HT-FT and cobalt-based for LT-FT (De Klerk, 2011).

$$nCO + (2n+1)H_2 \rightarrow C_nH_{2n+2} + nH_2O \qquad \text{R9}$$

LT-FT in a slurry bed using a cobalt (Co) catalyst operating at high pressures and low temperatures, promotes the formation of long paraffins and olefin chains, producing a hydrocarbon oil similar to plastics-derived pyrolysis oil (Bube et al., 2024). The olefinic production depends on the FT reactor technology used, either fixed bed or slurry bed, where slurry bed reactors produce a higher olefin-to-paraffin ratio (De Klerk, 2011). There are multiple commercially available FT production processes, for example, Sasol has been operating facilities since the 1950s using multiple reactor designs. Currently, it is operational in Sasolburg (South Africa), producing ~150,000 bpd (~8.7 million m^3/year) of FT crude from coal, using a slurry reactor Co-based LT-FT (De Klerk, 2011; Sasol, 2025). Additionally, Sasol and Topsoe are working to commercialize a SAF production plant using the G2L e-fuels technology (TRL 6–7), supported by demonstration projects such as FrontFuel (Denmark) and German Aerospace Center (Topsoe, 2026a).

In contrast, HT-FT yields a product distribution similar to MTO processes (see Figure 4), predominantly producing olefinic hydrocarbons smaller than C5 and high methane production. Separation of C2–C4 hydrocarbons from unconverted syngas is energy and capital-intensive because multiple distillation columns are required, which may become economically unattractive at a small scale (Gogate, 2019; Thunman, Berdugo Vilches, et al., 2019).

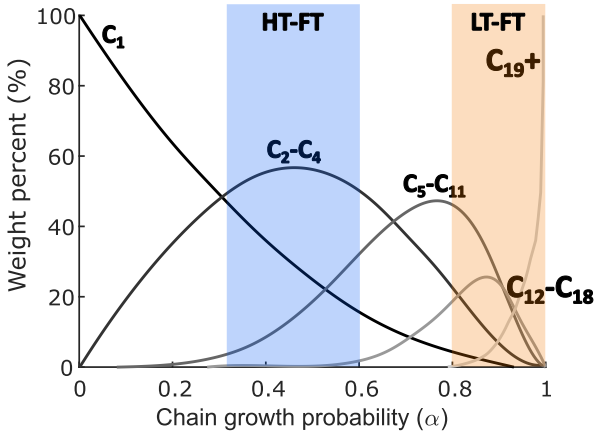

Figure 4: Anderson-Schulz-Flory (ASF) model for the prediction of Fisher-Tropsch reactor product distribution. The orange section shows the prediction for low-temperature Fischer-Tropsch (LT-FT) production, while the blue section shows the prediction for high-temperature Fischer-Tropsch (HT-FT) production.

The effect on chain growth probability can be seen in Figure 5, where the molar carbon fraction for each carbon number is shown. Figure 5a shows the molar carbon fraction distribution for α=0.92, representative of an LT-FT reactor, and Figure 5b shows the molar fraction for α=0.6, representative of an HT-FT reactor. According to the ASF distribution model, methane should not be in the mixture when α≥0.9. However, there have been studies concluding that methane

and lighter hydrocarbons are underestimated in the ASF distribution (Pondini & Ebert, 2013; Rane et al., 2012), more information on this effect is discussed in the Supplementary material for **Paper II**.

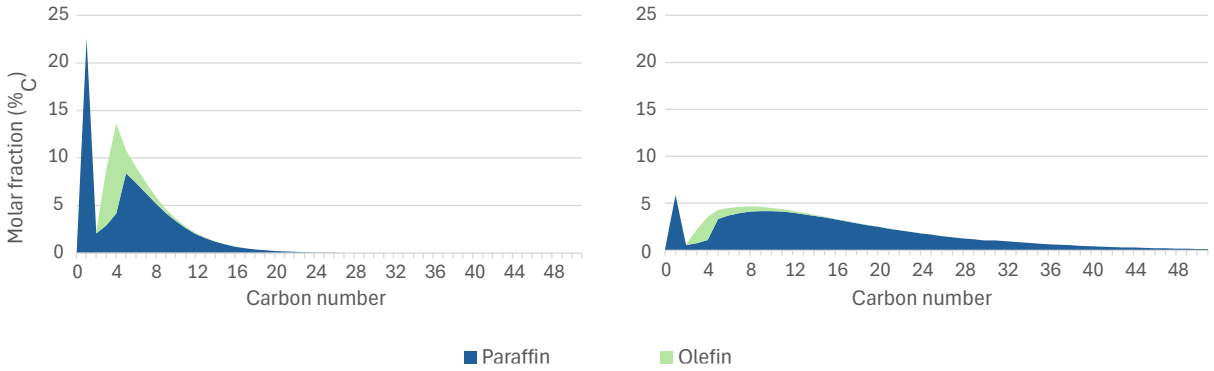

Figure 5: Molar carbon fraction in the FT production per carbon number, divided between paraffin and olefin, for different chain growth probability (α) to represent (a) High-temperature FT of α=0.7 and (b) low-temperature FT of α=0.92. Data obtained using the model utilized in **Paper II**.

FT syncrude cannot be reformed in a steam cracker plant or used as fuel directly due to the olefinic content and large amounts of methane and long hydrocarbon chains (see Figure 5). A number of published studies have explored the possibility of (i) hydrogenating the FT syncrude and using the naphtha range in a steam cracker, leading to high hydrogen demand (Karaba et al., 2021; Wang et al., 2010), (ii) separating and using only the paraffinic range, leading to high energy demand (De Klerk, 2011) , (iii) hydrocracking (De Klerk, 2007; Wang et al., 2010) or using a fluidized bed catalytic cracker (FCC) (Dupain et al., 2005; Kubička & Černý, 2012) followed by further refining to convert wax into hydrogenated lighter fractions for fuel production or as a steam cracker feedstock. Dement'ev et al (2020) converted unsaturated FT syncrude in an FCC to produce light olefins, obtaining yields of 56.8% of C2–C4 olefins, where 27.6% is propylene. Hydrogenation requires large quantities of hydrogen, approximately three times the stoichiometric hydrogen required, in order to fully hydrogenate the mixture, which makes the process economically challenging (De Klerk, 2011).

Integrated thermochemical conversion technologies

Thermochemical conversion technologies such as pyrolysis, gasification, and steam cracking enable the transformation of biomass and waste into valuable fuels and chemicals. *Pyrolysis* targets the production of liquid hydrocarbons, whereas *gasification* processes favor gaseous products, such as syngas, through higher temperatures. In this thesis, they are included only as contextual reference points, while the detailed discussion focuses on fluidized bed steam cracking (FBSC), which is evaluated in **Paper II** as a thermochemical recycling route for plastic waste.

Fluidized bed steam cracking (FBSC) technology has emerged as a promising thermochemical recycling technology, capable of converting heterogeneous solid and liquid waste streams into virgin-quality plastic building blocks by breaking down the molecular structure into constituent monomers (Forero-Franco et al., 2025; Mandviwala et al., 2024; Thunman, Berdugo Vilches, et al., 2019). FBSC technology builds on the work by Kaminsky et al (1995) and can produce a product distribution similar to conventional naphtha steam crackers, enabling integration with

existing downstream fractionation sections located in steam cracker plants and thereby providing the potential to substitute existing tubular steam crackers.

A DFB steam cracker semi-industrial reactor (TRL 6) is installed and operated at Chalmers University (4 MW_{th}, equivalent to 350 kg/h of pure PE) (Mandviwala, 2024). The reactor has been tested with a wide range of waste feedstocks (Mandviwala, 2024), as shown in Figure 6. The system uses a circulating fluidized bed boiler (CFB) as a combustor to provide heat, and a bubbling fluidized bed (BFB, ~700–800 °C) as a steam cracker reactor using steam as a fluidizing agent. Bed material circulates from the CFB to the BFB to provide heat for the endothermic reaction. Thunman et al (2019) showed that the DFB steam cracker processing pure polyethylene (PE) can achieve high carbon recovery (60–70%) with high overall energy efficiency (~65%). DFB can handle heteroatoms, such as oxygenates, as well as olefinic feedstock. This is enabled by the reactor configuration, where coke formed in the steam cracker side is continuously transferred with the bed material to the combustion side, where it is burned, enabling coke removal and heat transfer.

Other research institutes and industries are investigating similar technology. TNO has developed a technology based on an FCC, in which the cracking occurs in the riser. The technology has been demonstrated in pilot scale (25 and 800 kW_{th}) (Synova, 2026; Van Der Meijden et al., 2009). Additionally, the research center VTT has developed a similar technology based on CFB-BFB design, demonstrated at pilot scale (Nieminen, 2022).

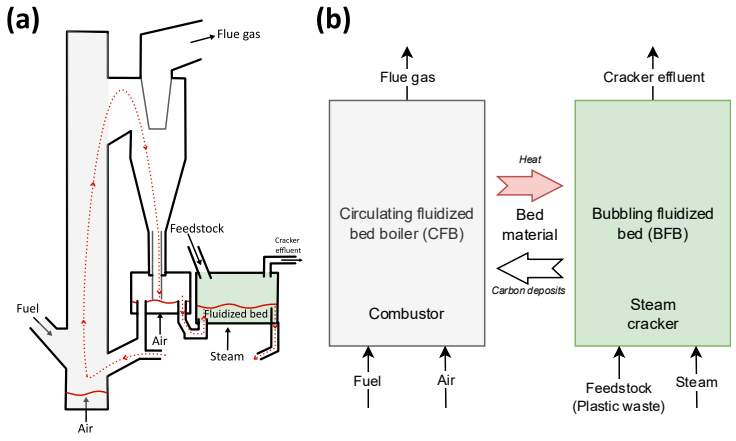

Figure 6: (a) Realistic representation of the DFB steam cracker installed at Chalmers University of Technology, from which the experimental data used in this work have been obtained. The red dotted lines show the circulation of the bed material. (n) Schematic of the interconnected CFB combustor and BFB reactor. In all figures, the green box represents the reactor performing the steam cracking reaction, while the gray box represents the reactor supplying energy for steam cracking (combustor). Adapted from Mandviwala (2024).

Integrated carbon recovery pathways

Studies on existing plants to implement carbon recovery technology in order to synthesize chemical products are ongoing. For example, *Waste to Energy (WtE) plants* can recover carbon contained in the boiler flue gases through a carbon capture and utilization (CCU) process, such as methanol-to-olefins (MTO). Recovering carbon through CCU requires a large additional energy input for hydrogen production, leading to low overall energy efficiency from feedstock to final products (35–45%$_{LHV}$) (Beiron et al., 2026; Cañete Vela et al., 2022; Li et al., 2025) with a potential carbon recovery of 70–85% (Witoon et al., 2023). *Gasification* technologies

for biomass, including the MTO process, can lead to higher energy efficiency (52–54%) (Arvidsson et al., 2016; Xiang et al., 2015), but due to the feedstock quality, lower carbon recovery can be achieved, 25–32% (S. Ding et al., 2026; Xiang et al., 2015). Similarly, gasification of municipal waste with high LHV content (~20 MJ/kg) has been developed by Enerkem and is being investigated for implementation by Repsol in the Ecoplanta project in Tarragona (Spain). The technology is based on steam-oxygen gasification conversion of municipal solid waste to syngas (TRL 8) to methanol, which, according to their system boundary, can reach ~70% carbon recovery with an energy efficiency of around 60% (European Commission, 2021a). The integration of MTO in a *DFB steam cracker* has previously been explored by Thunman et al. (2019), showing the potential for integration of carbon recovery-based processes, while also highlighting the importance of achieving sufficient economies of scale to avoid additional separation units. Energy efficiency of 69% when achieving a carbon recovery of 92% using PE as feedstock is reported using biomass combustion as an energy source on the combustor side.

2.2 Regulatory Frameworks

Regulatory frameworks play a critical role in shaping industrial practices, establishing clear guidelines and targets to drive innovation for efficient and sustainable production. Thus, defossilization of industry is driven by policy requirements that include energy efficiency requirements, use of alternative carbon feedstocks, emission reduction targets (e.g., EU-ETS), and carbon pricing or taxation of imported goods (e.g., CBAM).

National and international policies and targets are designed to establish clear guidelines that industries can use to support their decisions. There is currently no globally accepted common criteria for fuel definitions and targets (IEA, 2024) which can lead to inconsistencies in interpretation and regulatory complexity. Hence, regulatory frameworks can also act as a constraint for innovation, limiting specific feedstocks or technologies from being deployed despite their performance and availability. In this work, regulatory frameworks from the European Union (EU) are analyzed.

2.2.1 European Union regulations

The EU aims to achieve climate neutrality, net-zero greenhouse gas (GHG) emissions, by 2050 (European Commission, 2021b). The following directives are included that are relevant to this thesis.

Energy Efficiency Directive (EED) (2023/1791) aims to promote the use of energy more efficiently, contributing to reducing the EU's overall energy consumption. It establishes “energy efficiency first” as a fundamental principle for EU energy policy, meaning that energy efficiency must be considered by EU countries in all relevant policy and major investment decisions and “without which the full decarbonization of the Union’s economy cannot be achieved” (European Commission, 2023c). The aim is to double energy efficiency improvements by 2030 from 2% to 4% per year.

EU Emission Trading System (ETS) (2003/87/EC) puts a price on carbon emissions and lowers the cap on emissions from certain economic sectors every year (European Commission, 2020). Refineries and petrochemical industries are required to purchase allowances for emissions exceeding their allocated cap. 2030 targets aim to reduce emissions by 62% compared to 2005 levels, scale down free allocation of allowances, remove free allocation for the aviation sector and include new sectors such as buildings, road and transport in the EU ETS in ETS2 (European Commission, 2020).

Renewable Energy Directive (RED III) (2023/955) targets producing 40% of the EU energy supply from renewable sources in order to reduce GHG emissions from the energy sector (The European Commission, 2023b). RED III includes the prioritization of non-food-based feedstocks, limiting the approved feedstocks for biofuel production, in line with the directive's objective to maximize resource efficiency while ensuring sustainable resource management. **ReFuelEU Aviation** obliges fuel suppliers to blend increasing levels of sustainable aviation fuels (SAF), including synthetic aviation fuels, aviation biofuels and recycled carbon aviation fuels. To be classified as SAF, they must lead to at least 70% GHG emission savings compared to their fossil equivalent. ReFuelEU Aviation mandates that, starting from 2025, at least 2% of the fuel supplied at EU airports must be SAF, increasing to 70% by 2050. Additionally, within this target, a drop-in quota of 1.2% must come from synthetic aviation fuels, defined as Renewable Fuels of Non-Biological Origin (RFNBO), by 2030, rising to 35% by 2050 (European Commission, 2023d). The different fuel definitions and how they contribute to the targets described are shown in Table 2.

Waste Framework Directive (2008/98/EC) is designed to tackle problems related to the definition of waste, recycling and recovery, as well as waste hierarchy (prevention, preparing for re-use, recycling, recovery and disposal), to facilitate the transition to a circular economy system in an energy-efficient manner (European Commission, 2008). EU focuses on reducing pressure on natural resources, achieving climate neutrality by 2050 (European Commission, 2025). The long-term aim is to reduce the amount of waste generated, and when it is unavoidable to generate waste, to promote it as a resource and achieve higher waste recycling levels and disposal (Eurostat, 2024).

Thus, in order to comply with the mentioned regulatory frameworks, refineries and petrochemical industries need to minimize their carbon emissions and increase defossilization, e.g., increasing biogenic product production and using carbon dioxide removal technologies to offset residual emissions.

Additionally, to avoid shifting the emissions from within the EU to regions with less strict climate policies, the **Carbon Border Adjustment Mechanism** (CBAM, 2023/956) was introduced (European Commission, 2023a). CBAM applies carbon prices to a range of imported goods to prevent carbon leakage, ensuring that the European climate action does not result in an increase of emissions outside the EU. Currently, only cement, iron and steel, aluminum, fertilizers, electricity and hydrogen, however, organic chemicals, plastics, crude petroleum and refinery fuels are planned to be included in the second expansion of CBAM, after 2030. Additionally, it helps ensure that sustainably produced goods within Europe remain cost-competitive compared to imported products.

Table 2: Fuel definitions according to the European Union RED III regulatory framework and ReFuelEU Aviation, as well as the common definition in the fossil industry.

Broad category	**Fuel definition (incl. regulatory framework)**	**Definition used in this work**	**Contribution to the ReFuelEU Aviation objectives**
Sustainable aviation fuels (SAF) Defined by ReFuel EU in Article 3(7), including (i) Synthetic aviation fuels (RFNBO), (ii) Aviation biofuels, and (iii) Recycled carbon aviation fuels.	**RFNBO** Renewable liquid and gaseous transport fuels of non-biological origin. Defined in Article 3 (7) as synthetic aviation fuels and RED II Article 2 (36): Liquid or gaseous fuels which are used in the transport sector other than biofuels or biogas, the energy content of which is derived from renewable sources other than biomass.	Energy content of the fuel from renewable electricity contributes to 70% emission reduction from its fossil equivalent. Carbon source must have no energy content. Thus, CO_2. Combustion must not be intentional for CO_2 production. CO_2 agnostic (biogenic or fossil). Fossil CO_2 can be used until 2041, or 2036 if it is from a CHP plant.	SAF drop-in quota contribution.
	Advanced biofuels RED II article 2(34): From waste and residue (produced from feedstock listed in part A of annex IX of RED II).	Biofuels produced from feedstock listed in Part A of Annex IX of RED III.	SAF drop-in quota contribution.
	Biofuels RED II article 2(33): Produced from oils and fats and from feedstock listed in Part B of annex IX of RED II. Or defined in ReFuelEU Article 3(8c): Not using food & feed crops, which comply with the sustainability and lifecycle emissions savings criteria laid down in Article 29 of that Directive and are certified in compliance with Article 30 of that Directive.	Liquid fuel for aviation produced from biomass, including feedstock listed in Annex IX Part B of RED II. Comply with the sustainability and lifecycle emissions savings.	Lifecycle GHG emission reduction contribution.
	RCF (Recycled carbon fuels). RED II article 2(35): Liquid and gaseous fuels that are produced from liquid or solid waste streams of non-renewable origin, which are not suitable for material recovery, or from waste processing gas produced unintentionally.	RCF is not considered in this work since it depends on the carbon feedstock sourced.	SAF drop-in quota contribution.
Aviation fuels	Derived from crude oil.	Business as usual. Production through a crude distillation column.	No sustainability contribution.
	Fossil – low emissions production.	Using carbon capture and utilization techniques with fossil feedstock.	Plant-level GHG reduction contribution
Hydrogen to refinery, e.g., hydrogenate biofuels/fossil fuels. Not for hydrogen as a final product	**Fossil** – low emissions production. Not a policy definition.	Produced using biogenic feedstock connected to carbon capture and utilization techniques.	No sustainability contribution.
	Biogenic – low emissions production. Not a policy definition.		Plant-level GHG reduction contribution
	Low-carbon hydrogen. Defined in ReFuelEU	Produced using renewable electricity contributes to at least 70% GHG emission reduction from its fossil equivalent.	

3. Method

This chapter presents the methodology used to screen defossilization pathways for refinery and petrochemical systems. The framework is first described at a general level (Section 3.1), followed by the two case studies (Section 3.2), the selected technologies (Section 3.3), and the key performance indicators used for comparison (Section 3.4).

3.1 Methodology for early-stage screening of production pathways

Early-stage screening process for defossilization technologies is key to understanding the advantages and disadvantages of different process pathways and enabling an informed decision, minimizing time and resources. The methodology combines resource efficiency, including carbon recovery, energy efficiency and exergy losses, with preliminary technoeconomic analysis and external constraints such as regulatory frameworks and feedstock quality.

The framework thermodynamically groups defossilization technologies to compare a broad range of production pathways using a limited number of representative cases. The analysis uses a level of detail appropriate for early-stage screening, aiming to understand the main limitations of each production pathway in terms of potential resource utilization efficiencies or main exergy losses, rather than providing equipment-level design. The method is not intended to achieve a Front-End Engineering Design (FEED) study of the process pathways, which would require a detailed analysis of, for example, equipment sizes or integration into the steam network. Instead, it provides sufficient resolution for consistent pathway comparison.

The early screening of defossilization technologies methodology is shown in Figure 7. The selection of a defossilization technology alters the mass and energy flows of the production pathway, thereby influencing carbon flows, energy requirements, emissions, and overall economics. The framework consists of four steps: reference system definition, technology selection and system-boundary definition, screening-model development, and pathway comparison.

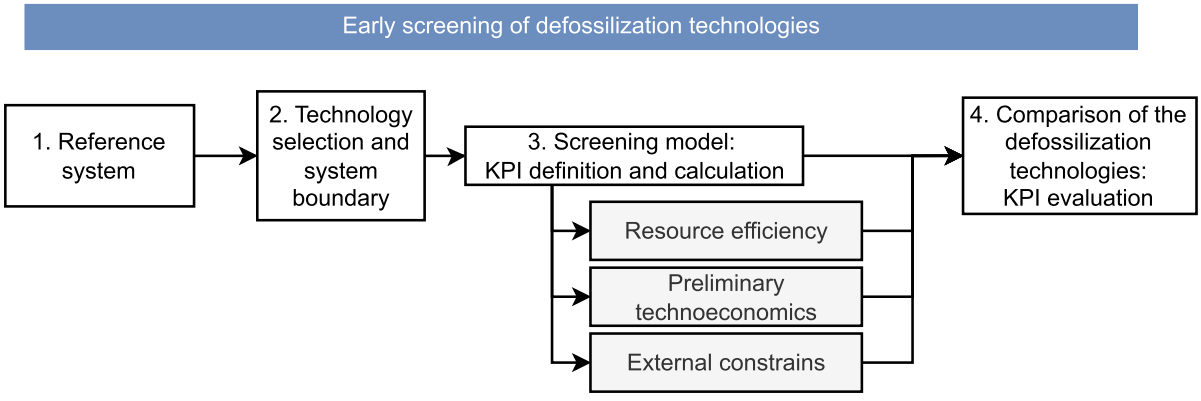

Figure 7: Visualization of the methodology for early-stage screening of production pathways of defossilization technologies. The numbers correspond to the bullet points described in Section 3.1.

1. **Reference system definition:** The reference system is defined through the specification of the industrial context, the targeted products and the potential feedstocks. Together, they determine the scope of the analysis. The selected case studies used in this thesis are shown in Section 3.2.
2. **Technology selection and system boundary:** a set of defossilization technologies are systematically identified and screened based on the product and feedstocks defined in the *reference system*. A preliminary literature review is then conducted to understand the key characteristics of each *technology*, highlighting key similarities and differences. Technologies with similar thermodynamic functions are grouped into representative pathway categories to broaden the screening without increasing complexity. When possible, this step is complemented by consultation with industrial stakeholders to validate assumptions and ensure industrial relevance. A *system boundary*, such as shown in Figure 8, is defined by identifying the relevant parts of the process to be investigated, such as feedstock, products and energy sources, ensuring a consistent basis for comparison.
3. **Screening model:** Simplified process screening models are developed for the selected pathway categories. The models include the main chemical reactions and energy requirements, analyzing *mass and energy balances* required to estimate *process performance*. The pathways are then evaluated using resource-efficiency indicators, including carbon, energy, and exergy efficiency. Where relevant, a *preliminary techno-economic assessment* can be performed using literature data. This provides an initial indication of the economic implications of each technology. *External constraints*, such as feedstock availability, regulatory frameworks, and technology maturity, are considered separately because they can strongly affect pathway feasibility. A description of the analyzed KPIs is provided in Section 3.4.
4. **Comparison:** the KPI outputs analyzed in Step 3 are integrated to support *technology screening and comparison*. Thus, the framework provides a discussion of trade-offs between technical performance, technoeconomic performance, and external constraints. The comparison and evaluation of the KPI are presented in Section 4.

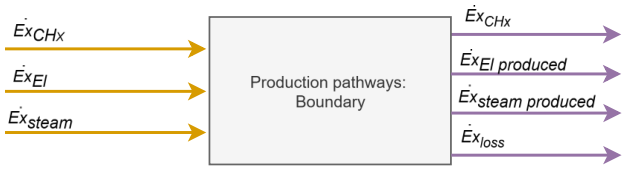

Figure 8: Example of a system boundary (Step 2) used for the exergy analysis in **Paper I**.

3.2 Reference system: Selected case studies

The methodology defined in Section 3.1 is exemplified using the following case studies investigated in **Papers I–II**.

In **Paper I** the perspective of defossilization of an oil refinery is addressed. The *reference system* addresses the production of SAF through different production routes. The paper evaluates three conceptually different *technology* process pathways that convert hydrocarbon feedstock into an intermediate product for the production of SAF. Due to the chemical complexity of the feedstock (biomass or waste) and the product (jet fuel), the technologies are compared using a simple hydrocarbon feedstock, where methane is used as a proxy hydrocarbon feedstock, and methanol is used as a proxy intermediate for SAF production. The *system boundary* is drawn from reforming the hydrocarbon feedstock to the production of methanol. Electricity and steam are supplied from an external source.

In **Paper II** the defossilization of steam cracker plants by using waste as feedstock is analyzed. The reference system addresses the effects of including a carbon recovery section using a fluidized bed steam cracking unit for thermochemical recycling of waste, to increase production of plastic building blocks: olefins and BTX. The paper builds on previous work by Thunman et al (2019) which uses methanol-to-olefins as a carbon recovery section by selecting Fischer-Tropsch as a carbon recovery technology for the recovery of carbon byproducts. The *technology selection* enables analyzing the effects of carbon recovery and of using the intermediate product in a steam cracker plant. The *system boundary* encompasses the conversion of waste into plastic building blocks, assuming that electricity is purchased to produce hydrogen, for reforming of byproducts, and oxygen, for oxyfuel combustion.

3.3 Technology selection: Process descriptions

In **Paper I**, the technologies are *screened* by calculating mass and energy balances to evaluate resource efficiency, based on exergy efficiency, energy efficiency and techno-economics. Exergy is prioritized over energy due to the combination of different energy levels, i.e., steam, electricity and chemical energy, see Section 3.4. *External constraints* are evaluated, such as the implications of the production routes on the fuel definitions defined in the RED III regulatory framework, specifically for RFNBO production. Additionally, simplified molecules are used to capture the fundamental characteristics and constraints of reforming hydrocarbons. Methane is used as a proxy for any biogenic feedstock to represent complex molecules, such as forest residues or municipal biogenic waste. Similarly, methanol is used as an intermediate product for SAF production, which can be upgraded via a MtJ process (see Section 2.1). The implications of using simplified molecules are discussed. Figure 9 shows a representation of the different technology pathways evaluated, including stand-alone and integrated into a refinery.

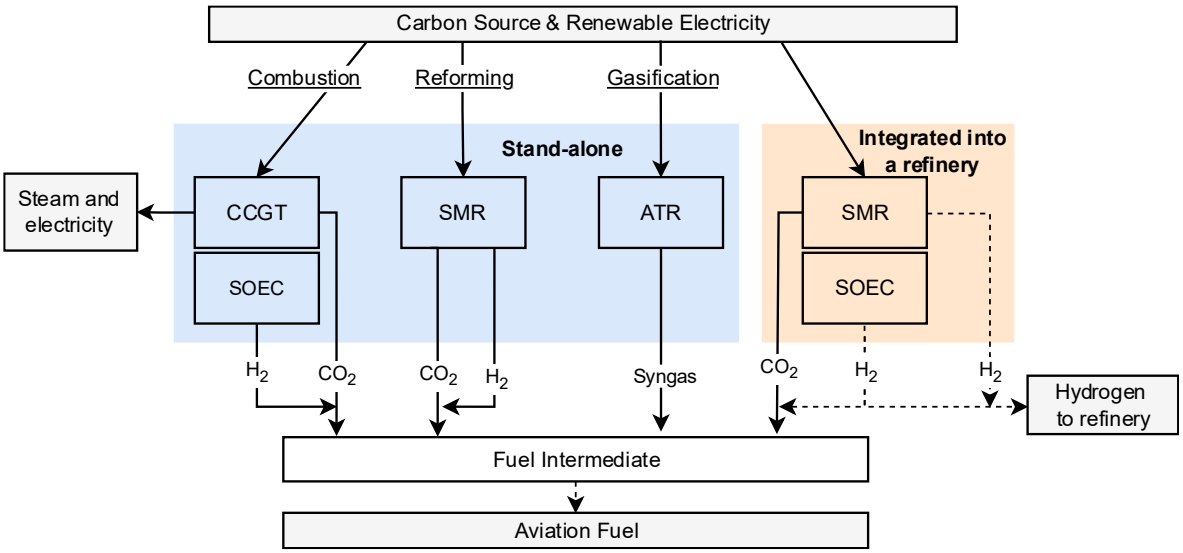

Figure 9: Overview of pathways using a cabron source and renewable electricity to produce methanol as an intermediate for aviation fuel. The pathways are presented at an aggregated level and therefore do not distinguish between specific feedstock types. Abbreviations: CCGT, combined cycle gas turbine; SOEC, solid oxide electrolyzer cell; SMR, steam methane reforming; ATR, autothermal reforming.

Three stand-alone production pathways are considered, based on combustion, reforming, and gasification technologies (see Section 2.1). Additionally, an integrated case is studied by integrating the reforming pathway into a refinery that requires significant volumes of hydrogen, e.g., for the hydrogenation of lipid-based feedstocks. In the integrated case, two configurations are possible: (i) *linear*, in which hydrogen and carbon are from the same source; and (ii) *hydrogen reallocation*, in which hydrogen production is decoupled from the carbon feedstock, leading to RFNBO methanol production. The combustion pathway (CCGT-SOEC) is used as the reference pathway throughout the work in **Paper I**, as it enables the production of RFNBO methanol in accordance with current EU directives.

In **Paper II,** the *screening* model uses Aspen Plus simulation software to generate mass and energy balances. The process is shown in Figure 10. The carbon recovery and energy efficiency of each scenario are analyzed, including the electricity demand for hydrogen and oxygen production. The impact of *external constraints* is analyzed through the quality of feedstock by comparing virgin polyethylene (PE) feedstock, representing the maximum achievable efficiency, with mixed plastic waste (MPW), representing a more realistic system. For more information, see **Paper II**.

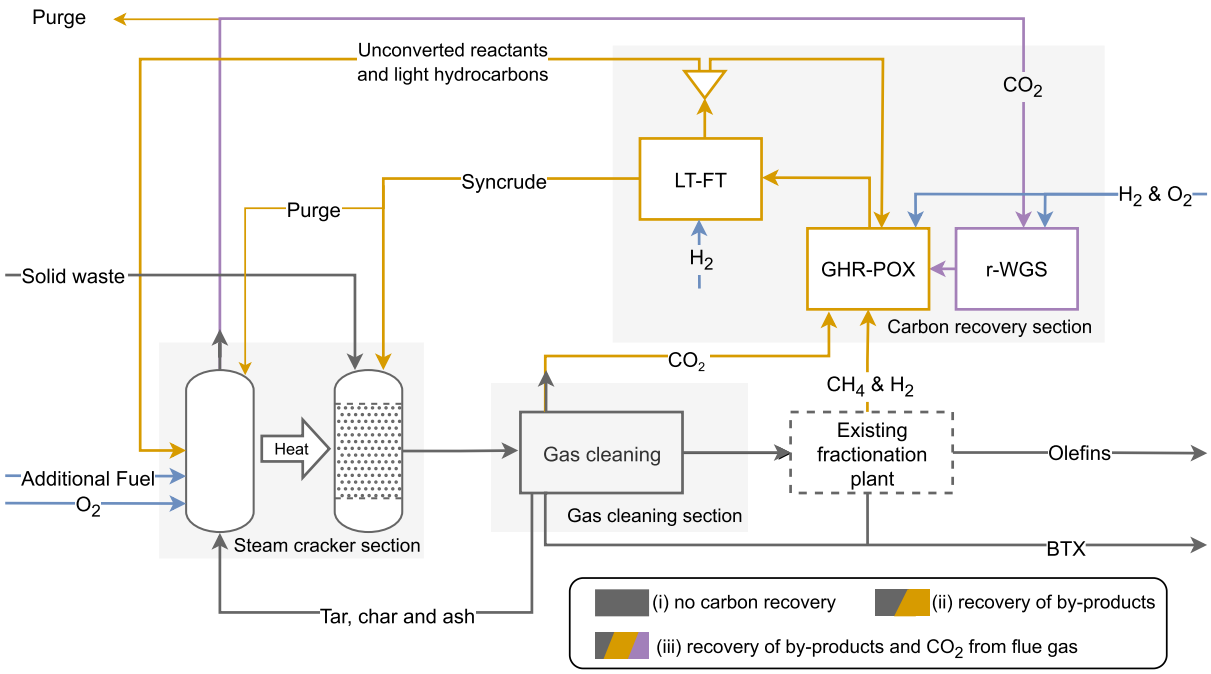

Figure 10: Process diagram of the steam cracker, gas cleaning and carbon recovery section. Hydrogen and oxygen-consuming units are marked. A purge stream is implemented downstream of the LT-FT reactor to ensure model convergence. Further description of the units and scenarios can be found in **Paper II**. GHR-POX: Gas heated reformer with partial oxidation, r-WGS: reverse water gas shift, LT-FT: low-temperature Fischer-Tropsch.

Solid waste is fed to a DFB steam cracker in which heat is provided through an oxyfuel combustor. The product gas from the steam cracker is first cleaned to remove solids and heteroatoms. The cleaned gas is then sent to a fractionation plant for separation of the products. The by-product gases (CO_2, CH_4 and H_2) separated in the gas cleaning and fractionation plant are sent to the carbon recovery section, where, through GHR-POX, they are converted to syngas and sent to an LT-FT reactor to produce a wide range of hydrocarbon chains (see Section 2.1). The FT light gases are separated from the syncrude and water and mainly recycled to the reformer. The syncrude is sent to the DFB steam cracker to be reformed and increase production of essential products. The CO_2 in the flue gas from the oxyfuel combustor can be recovered using a reverse water gas shift reactor (r-WGS) in the carbon recovery section to generate syngas and send it further to the LT-FT. Additional hydrogen and oxygen are required for the production of syngas and oxygen for oxyfuel combustion. An SOEC is assumed for emission-free hydrogen production, obtaining oxygen as a by-product, and an air separation unit (ASU) if additional oxygen is required.

Three different carbon recovery scenarios are selected, shown in Figure 10. Scenario *(i) no carbon recovery* (shown in black) implies a linear system, meaning no internal recirculation of by-product gases or residual CO_2. Scenario *(ii) recovery of by-products* (shown in black and orange) includes the reforming of the downstream by-products. Finally, Scenario *(iii) recovery of by-products and flue gases* or *full carbon recovery* (shown in black, orange and purple) includes the reforming of the CO_2 from the combustion side of the DFB steam cracker in addition to reformed by-products mentioned in Scenario (ii).

3.4 Screening model: Key performance indicators

Clearly defined key performance indicators (KPIs) are required to evaluate process performance and enable high-level comparisons of defossilization pathways. Technologies are assessed based on their contribution to resource efficiency, including carbon, energy and exergy efficiency, see Section 3.4.1, techno-economics, see Section 3.4.2, while also considering external constraints, such as regulatory frameworks and feedstock quality, see Section 3.4.3. The selected KPIs reflect the dual objective of maximizing resource efficiency while accounting for external constraints relevant to industrial implementation.

3.4.1 Resource efficiency:

Carbon recovery

Defossilization aims to shift from virgin fossil-based feedstock to recycled or renewable feedstock, which are difficult to collect and sort. As a result, resources used in the process should be converted as efficiently as possible. Therefore, carbon recovery is a central indicator of resource efficiency. Carbon recovery is defined in Eq. 2 as the percentage of the carbon in feedstock that is incorporated into essential carbon products (EP), as defined in Section 4.1.

$$Carbon\ recovery = \frac{\dot{C}_{EP}}{\dot{C}_{feedstock}}\ (\%_C) \tag{2}$$

Energy efficiency

Resource efficiency also depends on the amount and quality of energy required to achieve high carbon recovery. High carbon recovery requires heat and electricity, for example, for hydrogen production, and thus all candidate pathways must also be optimized with respect to energy use. Renewable electricity is considered a constrained resource and should be used efficiently and prioritized for applications where it provides the highest value. Energy efficiency of the process is defined by useful energy divided by energy expenditure, shown in Eq. 3. Useful energy includes the energy content in the product streams. Energy expenditure includes the energy content of the feedstock, steam demand and electricity demand. For example, electricity can be consumed by compressors, electrolyzers and air separation units. Steam demand is not included if the process gives a net positive heat balance (see **Paper II**).

$$\eta_{en} = \frac{\dot{m}_{products} \cdot LHV_{products}}{(\dot{m}_{feedstock} \cdot LHV_{feedstock}) + \dot{W}_{el} + \dot{Q}_{steam}} \tag{3}$$

Exergy efficiency

In heat-intensive processes, energy efficiency alone can be misleading, since differences in energy quality across energy streams are not accounted for. Exergy is defined as the "maximum useful work which can be extracted from a system as it reversibly comes into equilibrium with its environment" (Szargut et al., 1987). In simpler terms, it can be described as a metric that considers the *quality of energy*, reflecting its potential to be converted into *useful work*. Electricity is assumed to have an exergy efficiency of 100%, as it can theoretically be fully converted into useful work. Figure 11 shows an example of the impact between energy and

exergy as an example for a CHP plant, illustrating the effect of accounting for heat quality. Hence, exergy is used as a metric to provide a more appropriate basis for comparing energy use in production processes, particularly when multiple energy forms (chemical energy, electricity and steam) and temperature levels are involved. Energy flows are converted into exergy flows according to the methodology described in **Paper I** where the reference state for all the streams is atmospheric conditions (T_{ref} = 298 K and P_{ref} = 1.01 bar). Exergy efficiency is calculated according to Eq. 4.

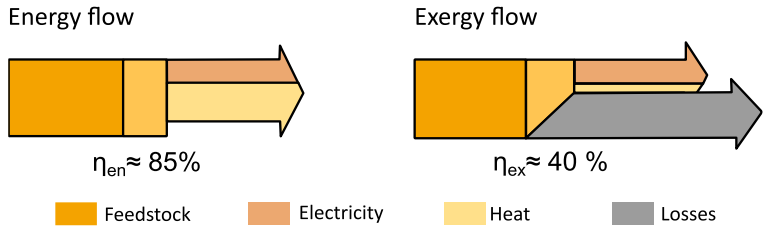

Figure 11: Energy vs exergy flows in a combined heat and power plant as an example. Representation inspired by (Wall, 2004).

Unlike energy, exergy is not subject to a conservation law, rather it is destroyed or consumed during irreversible processes due to thermodynamic inefficiencies. Hence, exergy loss is calculated (Eq. 5–6) to understand the exergy flow for each pathway *i* throughout the process *y* and thus identify the process with the smallest exergy losses.

$$\eta_{ex} = \frac{\dot{E}x_{products}}{\dot{E}x_{input}} \tag{4}$$

$$\varepsilon_{loss} = \frac{\dot{E}x_{input} - \dot{E}x_{products}}{\dot{E}x_{products}} = \frac{\dot{E}x_{loss}}{\dot{E}x_{products}} \tag{5}$$

$$\dot{E}x_{i,y} = \dot{E}x_{input\ i} - \dot{E}x_{loss\ i,y} \tag{6}$$

3.4.2 Techno-economics and OPEX cost assessment

Technoeconomic calculations are performed for each pathway to understand the impacts on each part of the process. Techno-economic calculations are not intended to provide realistic capital cost (CAPEX) and operational costs (OPEX), but rather to capture the relative differences between the compared pathways. A bottom-up approach is taken where the capital and operational costs are compared to determine the levelized cost of production of the essential products EP for each process pathway *i*, as defined in Eq. 7–8. The economic parameters required for CAPEX, OPEX calculation and sensitivity analysis are presented in **Paper I**.

$$C_{EP_i} = C_{CAPEX_i} + C_{Fixed\ OPEX_i} + C_{Feedstock_i} + C_{El_i} + C_{steam_i} + C_{CO_2\ emisssions} \left(\frac{€}{\text{year}}\right) \tag{7}$$

$$LCOEP_i = \frac{C_{EP_i}}{\dot{m}_{EP_i}} \left(\frac{€}{kg_{EP}}\right) \tag{8}$$

3.4.3 External constraints:

This subsection describes the external constraints influencing the evaluation of production pathways for their implications for real-world implementation. The external constraints chosen are regulatory frameworks and feedstock selection.

Regulatory frameworks

Analysis of the implications and advantages of applying new defossilization production pathways within current regulatory frameworks is key to understanding the incentives for industrial investment. Regulatory frameworks evolve to reflect technological development and societal needs. Therefore, scenarios are chosen to assess the impact of regulatory frameworks on the analyzed defossilization production pathways. The following procedure is applied when comparing regulatory framework requirements with the technical results obtained. An example of the procedure is shown in **Paper I**.

1. Identifying relevant regulatory frameworks for Europe and worldwide.
2. Review of the requirements and constraints of the regulatory framework that can affect the production pathway, such as feedstock requirements or economic compensation.
3. Evaluate if the installation of the analyzed production pathways goes in line with the regulatory frameworks in place or if it would not be incentivized.

Feedstock selection

Fundamental differences between pathways can be overshadowed by the use of a specific feedstock, requiring a high number of computational studies which explore the large variety of alternative carbon feedstock, ranging from different types of biomass, level of heterogeneity, contamination level, etc. In this work, the contamination level has been calculated according to Eq. 9, where $\dot{m}_{heteroatoms}$ is the mass of the heteroatoms, i.e., the atoms other than carbon or hydrogen, such as oxygen, nitrogen, ash and sulfur. Additionally, a comparison between low- and high- contamination feedstock to evaluate the impact of contaminants present in the system is performed in **Paper II**.

$$Contaminantion = \frac{\dot{m}_{heteroatoms}}{\dot{m}_{total,daf}} \ (\%_{daf}) \tag{9}$$

The use of simplified molecules as a proxy leads to (i) minimization of calculation complexity, (ii) simpler understanding of the effects of the production pathway itself without the background of the feedstock, and (iii) a methodology that is applicable to all sorts of hydrocarbons. Thus, the selection of the same feedstock for pathway comparison simplifies the calculation, enabling a consistent carbon and hydrogen balance among the pathways, while avoiding uncertainties related to specific feedstock, such as variable biomass composition and moisture content. The methodology of using methane as a proxy for biomass for SAF production is used in **Paper I**, improving the effectiveness of analyzing a regulatory framework in a generalized way, since the results apply to a wide range of feedstock and products.

4. Selected results and discussion

This section synthesizes selected results from **Papers I–II** to illustrate how the proposed screening framework identifies trade-offs between carbon recovery, energy and exergy efficiency, feedstock quality, techno-economic performance, and regulatory constraints. Section 4.1 discusses resource-efficiency trade-offs, while Section 4.2 evaluates how regulatory frameworks affect pathway selection and deployment.

4.1 The role of resource efficiency

4.1.1 Carbon recovery

Enhancing production of products and thus maximizing carbon recovery translates to a high increase in internal recycling loops. Figure 12 shows a Sankey diagram indicating the effects on carbon flows when increasing carbon recovery for mixed plastic waste (MPW).

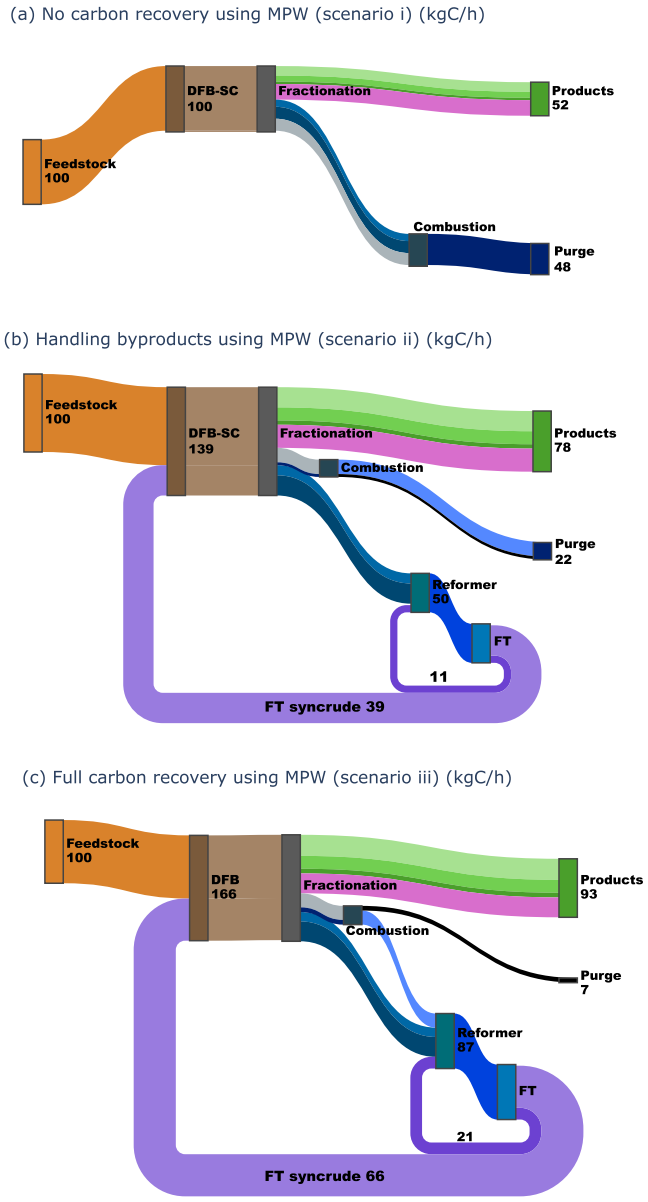

Figure 12: Carbon flows (kg C/h) for the different carbon recovery scenarios using mixed plastic waste as feedstock. Full description of the scenarios is shown in Section 3.3 and **Paper II**. Abbreviations: DFB, Dual Fluidized Bed; SC, Steam Cracker; FT, Fischer-Tropsch; MPW, Mixed Plastic Waste.

For the products to increase from 0.4 to 0.55 kg products/kg MPW, a recycling loop of 39% of carbon is required with an electricity demand of 2.8 MWh_{el}/ton of products. Similarly, for near-to-full carbon recovery (0.7 kg products/kg MPW), a recycling loop of 66% of carbon is needed with an additional total electricity demand of 8.6 MWh_{el}/ton of products for compressor work, oxygen demand and hydrogen demand, corresponding to 45% of the total energy input. Thus, an increase in carbon recovery translates into a shift of energy demand from chemical to electrical, i.e., shifting from hydrocarbon feedstock to renewable electricity, which is in alignment with defossilization goals. For more information, see **Paper II**.

The concept of carbon recovery can be extended to CCU pathways, such as methanol production, for integration into refineries. Figure 13 shows the exergy-based input and output flows for the case of integration into a refinery complex using methanol for SAF production, analyzed in **Paper I**. The utilized CO_2 is produced by minimizing combustion of the feedstock, thus leading to high exergy efficiency (~68%) and compliance with RFNBO definitions. In comparison, a stand-alone configuration (CCGT-SOEC) achieves an exergy efficiency of 58%. Thus, the integrated case shows the potential for efficient carbon recovery for fuel production using equipment installed in a refinery complex.

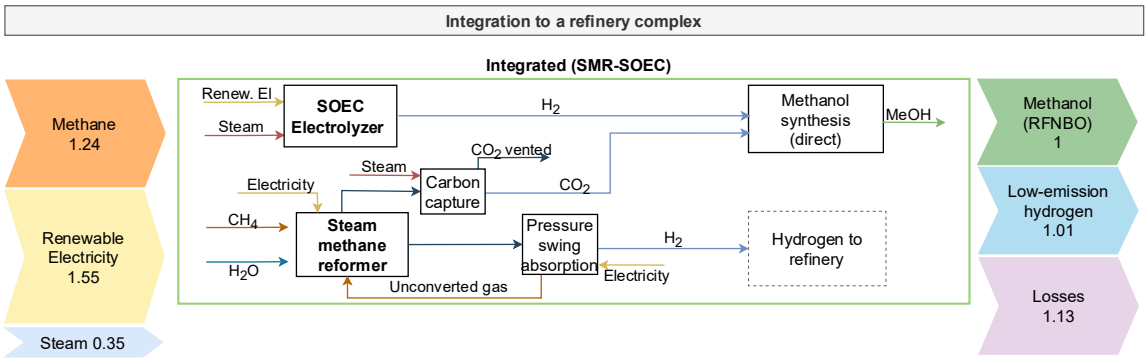

Figure 13: Exergy-based process flow diagram of the analyzed pathways. Overall flows are normalized to 1 GWh/year of methanol production. Abbreviations: SOEC, solid oxide electrolyzer cell; RFNBO, renewable fuel of non-biological origin.

4.1.2 Energy and exergy efficiency

Step-by-step exergy losses

All process steps lead to exergy losses, clear quantification of them helps the identification of the process limiting steps. Thus, step-by-step exergy loss analysis is used to understand the exergy losses in each step of the production pathway process, according to the methodology explained in Section 3.2–3.3. Figure 14 shows the exergy flows for each SAF production pathway throughout the process steps relative to the reference production pathway.

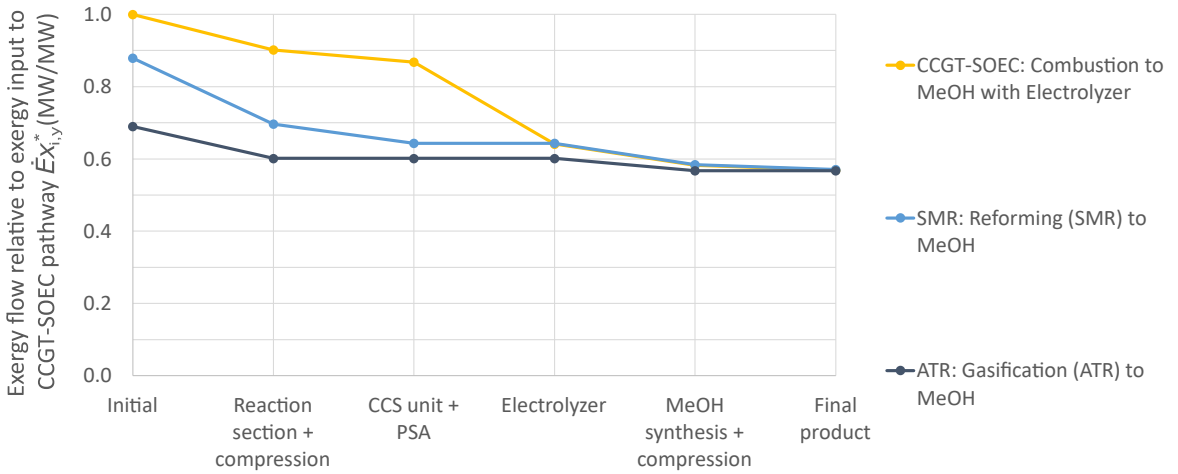

Figure 14: Representation on how to show the exergy losses for the stand-alone pathways from methane to methanol (MeOH), relative to the combustion-based pathway (CCGT-SOEC). The x-axis shows the successive process steps from the initial conversion reaction to the final methanol product. The y-axis shows how the exergy flow is lost throughout the process, normalized to the combustion-based pathway, illustrating the exergy added and retained through each step x and, consequently, the exergy losses along the pathway. Additional information can be found in **Paper I**. Abbreviations: CCGT, combined cycle gas turbine; SOEC, solid oxide electrolyzer cell; SMR, steam methane reforming; ATR, autothermal reforming; CCS unit, carbon capture unit; PSA, pressure swing adsorption.

Figure 14 shows that the largest exergy losses occur in the electrolyzer (decreased by 23 percentage points), placing the starting point of the combustion pathway above the other fuel production pathways. Additionally, most of the exergy losses occur during combustion, as it leads the fuel to a state of no chemical energy (CO_2). Furthermore, the reforming pathway exhibits a substantial exergy loss (a decrease of 15 percentage points) during the reaction due to its endothermic nature and the steam required for the process. The exergetic efficiency of the combustion (58%), reforming (68%) and gasification (84%) pathways differs from the energy efficiency for the same pathways (51%, 57% and 86%, respectively). Regulatory frameworks often mention energy efficiency, such as the Energy Efficiency Directive (EED), but do not fully capture the losses illustrated in Figure 14.

Implications of energy and exergy efficiency

The choice of technology directly influences the maximum range of carbon recovery and energy efficiency that can be achieved. The FBSC without carbon recovery can achieve a high carbon-to-products yield (52%) with a high energy efficiency (60%) when processing MPW. Lower quality feedstock, such as automotive shredded residues (ASR), would achieve a

carbon-to-product yield of around 21% (Thunman, Berdugo Vilches, et al., 2019). The processing of pure PE represents an upper bound for performance, achieving a maximum carbon-to-products yield (64%) and a maximum energy efficiency (~65%). Increasing the amount of carbon recovery using a FT reactor system can achieve a carbon recovery of 70–75% (max. 77–82%) with an energy efficiency of 70–72% (max. 73–75%) for MPW when emitting the CO_2 produced in the combustor side of the DFB reactor (scenario ii), where the maximum is represented for the conversion of pure PE. When achieving near-to-full carbon recovery (90%, max. 95%), an energy efficiency of 54–66% (max. 65–78%) is obtained.

Figure 15 provides an overview of some of the potential technologies for waste and biomass usage as feedstock for olefin production as a function of their energy recovery and carbon efficiency ranges. The boxes show approximate data extracted from literature for Waste to Energy plants and Gasification, mentioned in Section 2.1, while the dots show the results of the model studied in **Paper II** including FBSC technology using FT as carbon recovery for a range of feedstock qualities: from pure polyethylene to mixed plastic waste. **Paper II** discusses in detail the specific cabron recovery results and scenarios.

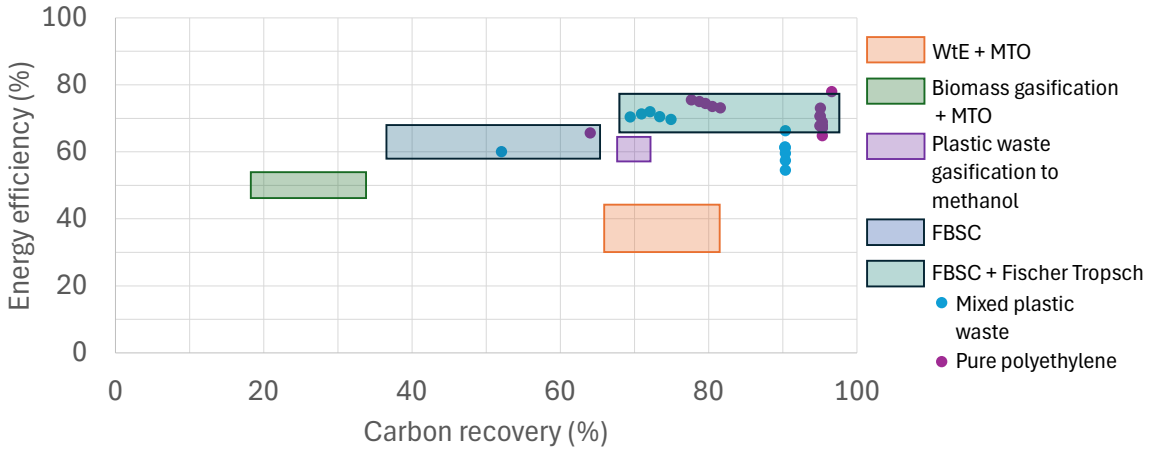

Figure 15: Carbon recovery vs energy efficiency of selected technologies for waste and/or biomass conversion to olefins for plastic production. Specific data extracted from the scenarios for the model performed for FBSC and Fischer-Tropsch as carbon recovery technology are represented as dots, while boxes are used for literature data and the average of the analyzed technology in **Paper II**. Abbreviations: WtE, waste-to-energy plant; MTO, methanol-to-olefins; FBSC, fluidized bed steam cracking.

Gasification can achieve high carbon recovery with moderate energy efficiency. However, the level of carbon recovery strongly depends on feedstock quality: higher biomass content translates to higher CO_2, thus lower carbon conversion. FBSC can achieve high carbon to product conversion even without carbon recovery sections, if high polyolefinic content is present in the feedstock. For increased biogenic content in the feedstock, higher oxygenates are produced, resulting in a lower carbon-to-products conversion rate. Combustion in WtE plants can achieve high carbon recovery, but it leads to an overall lower energy efficiency, due to the amount of hydrogen required for hydrocarbon production, in line with the findings in **Paper I**.

Figure 16 presents an analysis for the production of SAF through different production pathways, indicating the exergy input required per unit of exergy products, including different exergy inputs and produced fuel fractions. The production pathways include (i) fossil fuel production (i.e., crude oil reforming), (ii) RFNBO-fuel with methanol-to-jet (combustion

pathway (CCGT-SOEC) studied in **Paper I**), and (iii) biofuel technology currently implemented in refineries, i.e., conversion of used cooking oil (UCO) to jet fuel. This comparison illustrates the production of the different fuel fractions. Refineries can, however, optimize their product distribution, often at the expense of other fractions and with associated changes in energy demand.

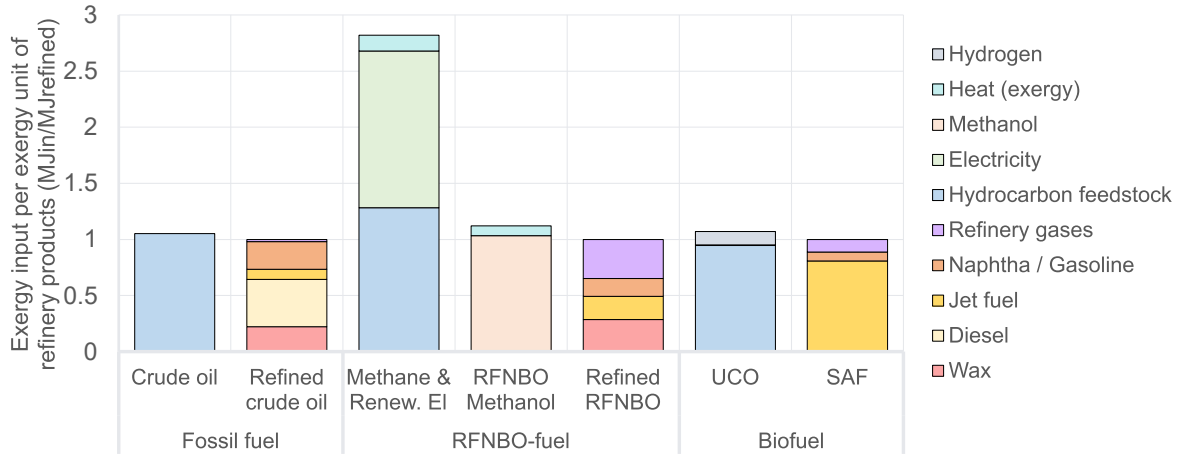

Figure 16: Exergy input per refinery product for known production pathways based on **Paper I**. The analysis is done for a European oil refinery using crude oil (IEA, 2025), a future refinery producing RFNBO (CCGT-SOEC pathway studied) (Eyberg et al., 2024), and for a UCO refinery, maximizing the yield of SAF (based on (Danish Energy Agency, 2024)). Abbreviations: RFNBO, renewable fuel of non-biological origin; UCO, used cooking oil; SAF, sustainable aviation fuel; Renew. El, renewable electricity.

To produce refined RFNBO fuel from the CCGT-SOEC (combustion) pathway studied, 2.77 MJ are needed, mainly due to the electricity requirement for hydrogen production. Integration into a refinery requires 3.23 $MJ_{in}/MJ_{refined,}$ but this pathway also produces 1.01 MJ of hydrogen, thus 1.61 $MJ_{in}/MJ_{products}$. The different production pathways generate by-products such as light hydrocarbons and waxes, which can be further reformed to increase their value and enhance the production of essential carbon-based products.

4.1.3 Impact of alternative feedstock type

Waste, either fossil or bio-based, is expected to play an increasing role as a feedstock to substitute crude oil. Sorted waste and biomass are limited feedstock, while mixed waste remains more available and easier to source. Figure 17 represents the alternative carbon sources used in the case studies, highlighting differences in composition in terms of the molar hydrogen-to-carbon (H/C) ratio as a function of the contaminant level (dry ash-free basis). The bubble size corresponds to the oxygen content in the feedstock.

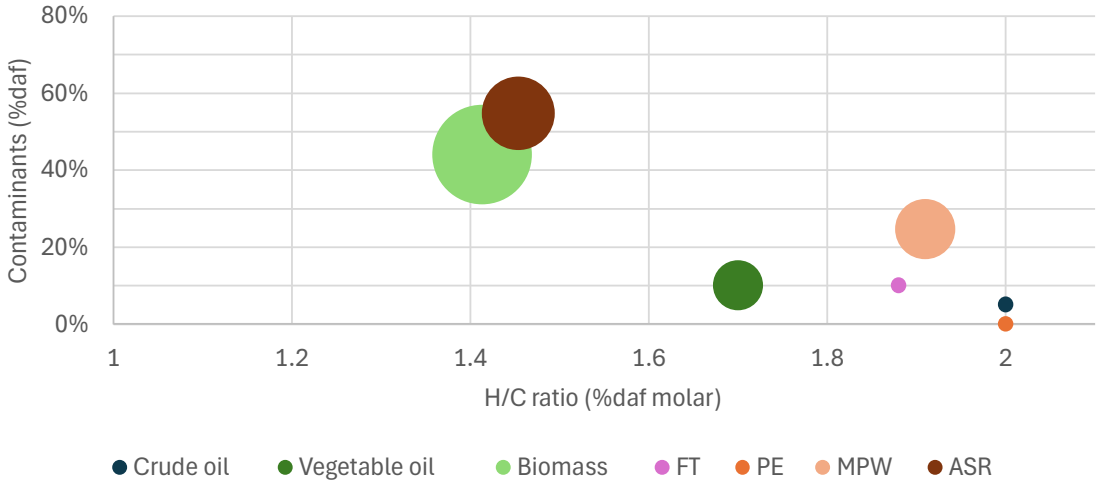

Figure 17: Hydrogen-to-carbon ratio (H/C) as a function of contaminant level for the feedstocks analyzed in this work on a dry ash-free basis. The bubble size corresponds to the oxygen content in the feedstock. Composition data for polyethylene, mixed plastic waste and FT are taken from **Paper II**. Other composition data is extracted from literature: for biomass (Fortet Casabella & Chehade, 2023), for ASR (Cañete Vela, 2024), for vegetable oil (Yang et al., 2024) and for crude oil (Alfke et al., 2007). Abbreviations: FT, Fischer-Tropsch; PE, polyethylene; MPW, mixed plastic waste; ASR, automotive shredded residues.

An H/C ratio close to 2 is similar to the feedstock currently used in petrochemical and refineries (crude oil), which contains a low grade of contaminants, mainly sulfur and nitrogen, and a low level of oxygen. Refineries need to adapt to renewable and recycled feedstock streams, which contain larger shares of contaminants and thus, a lower H/C ratio. Vegetable oil is part of the portfolio of defossilization measures currently implemented in refineries (see Section 2.1), leading to large hydrogen consumption due to the need for deoxygenation and hydrogenation measures. The implementation of more unrefined and heterogeneous feedstocks, such as biomass woodchips or, as an extreme, ASR, would require flexible technologies that can deal with decontamination. Flexible technologies that can handle different types of feedstocks are targeted to understand their effect. For example, to ensure compliance with regulatory frameworks over the entire project lifetime, industries should consider flexible technologies that can perform the transition from fossil to bio-based feedstock, including clean and waste-derived streams, without requiring significant process modifications.

RFNBO methanol production through the CCGT-SOEC pathway requires more electricity (52% of the total exergy input) than aviation biofuel methanol produced through the SMR pathway (12%). Furthermore, the cost of carbon feedstock has a smaller effect on the cost of producing RFNBO (34% of the cost) than the cost of producing aviation biofuel (70% of the cost). However, replacing feedstock with bio-based feedstock can significantly raise the

production costs. For example, methanol production based on biomethane can increase the production cost by ~20% compared to fossil-methane-based production. Therefore, the feedstock choice should be evaluated as a parameter in the feasibility study to account for its potential impact on production cost.

Similar increases in feedstock demand and energy consumption can be seen for plastic building block production through FBSC and FT processes, when switching from high plastic content feedstock (PE, 100% polyolefinic material) to higher biogenic content feedstock (MPW, 21% cellulosic material). Higher biogenic content is translated into higher oxygen content, which increases energy requirements. Nonetheless, for waste recycling, higher biogenic content translates into lower sorting rates, which translates into lower costs and lower conversion rates. Additionally, higher biogenic content allows claiming of biogenic content in emissions or products. Thus, maximizing carbon recovery directly enhances the incorporation of biogenic carbon into valuable products, which, through multiple recycling loops, can end up being considered fully biogenic.

When performing waste recycling in a DFB steam cracker, increasing carbon recovery of by-product (scenario ii) and including the recovery of the flue gas carbon (scenario iii) reduces plastic waste demand from 2.4 (scenario i) to 1.8 (scenario ii) and further to 1.44 (scenario iii) ton MPW/ton essential products. In order to achieve this reduction, the amount of electricity needed to produce the hydrogen required rises from 2.5 to 7.7 MWh_{el}/ton essential product (scenarios ii and iii). The total electricity required, accounting for compressors, hydrogen demand and oxygen demand, increases from 0.7 (scenario i) to 2.8 (scenario ii) and further to 8.6 (scenario iii) MWh_{el}/ton essential products.

To transform 10% of the Swedish production (~83 kton/year of products) from MPW would require 120–208 kton/year of MPW, depending on the carbon recovery scenario, which corresponds to 13–23% of the plastic waste generated in Sweden in 2023 (Naturvårdsverket, 2023). The reduction in plastic feedstock demand is achieved at the expense of increasing electricity demand, see **Paper II**, which translates to 61 (scenario i), 237 (scenario ii) and 717 (scenario iii) GWh_{el}/year, corresponding to 0.05–0.53% of total Swedish electricity consumption[7] (SKGS, 2024). This highlights the significant feedstock and electricity demand associated with large-scale implementation, even in a system characterized by relatively balanced production and consumption. However, combustion-based energy supply (e.g., biomass) represents an alternative to electricity-based energy supply.

The scale of the RFNBO requirement becomes clear when related to the EU jet fuel demand discussed in the background (see Section 2.1.1). A 1.2% synthetic aviation fuel share in 2030 corresponds to approximately 14.6 PJ/year of final fuel energy. Producing this amount of RFNBO jet fuel would require 52.5 TWh_{el}/year of renewable electricity, equivalent to approximately 39% of Swedish electricity consumption (SKGS, 2024), following the production pathway CCGT-SOEC in **Paper I**. If the same pathway were scaled to the 2050 target of 35% synthetic aviation fuel, the required RFNBO production would increase to

[7] The Swedish electricity consumption was ~135 TWh_{el}/year in 2023 (SKGS, 2024)

approximately 426.3 PJ/year, corresponding to an electricity demand of 1,531 TWh_{el}/year. This illustrates that RFNBO jet fuel production is constrained not only by carbon availability, but also by the very large demand for renewable electricity.

4.2 The role of regulatory frameworks

Regulatory frameworks can support the deployment of defossilization technologies by creating markets for otherwise economically challenging investments, either through incentives for low-emission products or penalties for less sustainable production pathways. However, policy support and mandates alone cannot eliminate the importance of resource efficiency. Building on Section 4.1, this section examines how selected EU regulatory frameworks influence the interpretation and deployment of pathways studied. The discussion focuses on sustainable aviation fuel (SAF) under RED III, ReFuelEU Aviation and EED (**Paper I**), and on waste recycling under the EU Waste Directive (**Paper II**).

4.2.1 RED III targeting less energy-efficient technologies

The pathways studied, shown in Figure 13 and **Paper I**, may raise concerns from a regulatory and sustainability perspective. Current regulatory frameworks designed by the EU set minimum blend-in quotas for SAF drop-in, which include advanced biofuel, biofuel, and RFNBO fuel definitions (see Section 2.2), with a planned increase over time.

Despite all production pathways studied yielding the same intermediate product (methanol), the classification as an RFNBO is not distributed equally. Figure 18 uses the costs calculated in **Paper I** and compares them to the production costs of conventional fossil jet fuel, including the costs of compensating for emissions with carbon credits. Offsetting GHG emissions by purchasing carbon credits does not exempt airlines from complying with the minimum-share requirements for drop-in of SAF and RFNBO. However, the analysis provides insights into when the costs of carbon credits will have a large enough impact on total costs to achieve cost break-even with SAF production costs, specifically RFNBO.

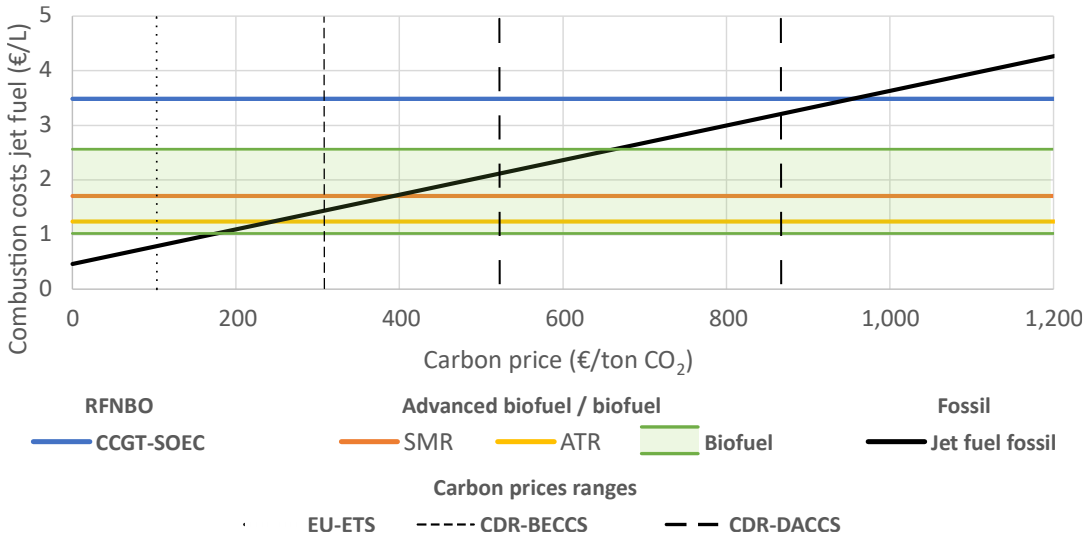

Figure 18: Combustion costs (€/L jet fuel) for different fuel production pathways (CCGT–SOEC, SMR, and ATR), compared with fossil and biogenic jet fuel. Costs include methanol-to-jet fuel conversion, assuming an exergy efficiency of 88% and a mass yield of 34% (Eyberg et al., 2024), using bio-based methane and electricity as inputs. Biogenic SAF prices are assumed to be 2–5 times higher than fossil jet fuel (Pavlenko et al., 2019), with literature ranges for animal fats and rapeseed oil pathways indicated (Danish Energy Agency, 2024; Neste, 2026; Pavlenko et al., 2019). Carbon prices reflect EU-ETS levels and voluntary market prices for BECCS and DACCS (cdr.fyi, 2025; Statista, 2025). For comparison and simplification, all fuel categories, i.e., refinery gas, naphtha, diesel, and wax, are assumed to be valued equally to jet fuel. Abbreviations: CCGT, combined cycle gas turbine; SOEC, solid oxide electrolyzer cell; SMR, steam methane reforming; ATR, autothermal reforming; EU-ETS, European Union emissions trading system; BECCS, bioenergy with carbon capture and storage; DACCS, direct air carbon capture and storage; CDR, carbon dioxide removal.

Figure 18Figure 18 shows that around ~400 €/ton CO_2 is required to make advanced biofuels-compliant SAF competitive with fossil-based jet fuel, similar to the voluntary market price for bioenergy with carbon capture and storage (CDR-BECCS) credits (cdr.fyi, 2025; Statista, 2025). However, for the production of RFNBO, approximately 1,000 €/ton CO_2 (above direct air capture (CDR-DACCS) voluntary market price) is required to become more cost-effective than offsetting fossil jet fuel emissions. RFNBO jet fuel costs are about 3.5–4 €/L, depending on the renewable electricity price, which is roughly eight times the price of fossil jet fuel (~0.5 €/L) and about two to three times the advanced biofuels cost (~1.3–1.7 €/L). Carbon emissions, rather than the underlying fuel product cost, become the primary cost driver for fossil-based jet fuel when the carbon price exceeds roughly 200 €/ton CO_2, which can promote the import of low-carbon fuel from outside the EU over domestic production through energy-efficient pathways.

Under RED III, this cost gap is addressed through a minimum blend-in quota, which creates a guaranteed demand for RFNBO-SAF and effectively reduces exposure to direct price competition with fossil jet fuel. Consequently, its deployment relies strongly on policy instruments, and cost-competitiveness is unlikely to be achieved if the policy is not in place.

4.2.2 Waste directive supporting technology innovation

The Waste Directive supports a shift from energy recovery toward recycling, which aligns with the deployment of thermochemical recycling technologies such as FBSC. Although carbon capture and utilization (CCU) can be applied to recover carbon from flue gases after combustion, as illustrated by the WtE+CCU pathway in Figure 15. Its installation risks creating a feedstock lock-in to energy-recovery infrastructure and delaying more energy-efficient recycling routes. In contrast, FBSC enables direct conversion of heterogeneous waste streams into plastic building blocks, with further carbon recovery possible through hydrogen-assisted conversion of CO_x.

However, increasing carbon recovery beyond by-product recovery raises electricity demand and production cost. Therefore, higher carbon recovery can reduce carbon losses and CO_2 emissions, but may also increase the levelized cost of essential carbon products. The economic attractiveness of full carbon recovery is therefore strongly influenced by the balance between electricity prices and the cost of emitting CO_2.

These results indicate that regulatory frameworks can support technology innovation by redirecting waste streams from energy recovery toward recycling. However, policy instruments should distinguish between carbon recovery routes that preserve the chemical value of waste and routes that recover carbon only after combustion. This will steer waste streams away from energy recovery (WtE plants), which is in line with resource efficiency and circular economy objectives in the Waste Directive and energy efficiency objectives in the EED. Flexible technologies capable of handling heterogeneous waste feedstocks can shift the role of waste from a low-value energy source to an adaptable carbon resource for material production.

5. Conclusions

This thesis contributes to the development of a framework to perform early-stage process screening of defossilization technologies using thermodynamic analysis for refinery and petrochemical industries under technical, feedstock and regulatory constraints. **Paper I** analyzes pathways for the production of sustainable aviation fuel (SAF) in a stand-alone process and integrated into a refinery setting. The analysis includes the implications of the EU's Renewable Energy Directive (RED III) regulatory framework, especially the definition of renewable fuel of non-biological origin (RFNBO). **Paper II** evaluates the potential of carbon recovery in a steam cracker plant for solid waste using fluidized bed technology (FBSC). Carbon recovery is performed using Fischer-Tropsch (FT) technology to produce long-chain hydrocarbons that can be cracked in the FBSC. The work analyzes the effects on energy efficiency when increasing carbon recovery in the process with different levels of waste contamination.

The methodological framework described in the introductory essay includes the analysis of performance indicators such as carbon recovery (**Paper II**), energy efficiency (**Paper I–II**), exergy efficiency and exergy losses (**Paper I**), and technoeconomic analysis (**Paper I**). The focus of this work was to synthesize the insights gained from the two papers and understand the research gaps to be addressed to further develop a framework for early-stage screening of defossilization pathways for refineries and petrochemical industries. The conclusions are structured around the research questions, linking the main findings of the specific cases analyzed in the appended papers to the broader aim of screening defossilization pathways.

RQ1: How can alternative production pathways be evaluated and compared to support defossilization of refinery systems?

The framework supports early-stage comparison by thermodynamically grouping production pathways and evaluating them using consistent performance indicators. Exergy analysis is a powerful but underused performance indicator for evaluating processes that combine heat, chemical energy, and electricity, particularly in regulatory frameworks, and it is expected to be more relevant as industrial electrification and hydrogen integration increase. The approach enables the identification of key limitations related to carbon utilization, energy demand, exergy losses, cost, and regulatory eligibility before detailed process design

RQ2: How does the integration of carbon recovery processes affect carbon utilization and energy efficiency in refinery and petrochemical systems?

Integrating carbon recovery to reform byproducts and CO_2 emitted from processes can increase carbon-to-products yield. In FBSC-based thermochemical recycling of mixed plastic waste, carbon conversion to plastic building blocks can increase from 52% (without carbon recovery) to 70% (with byproduct recovery) and up to 90% (with full carbon recovery), while maintaining the overall energy efficiency between 60–70%, although a decrease in energy efficiency is observed when going beyond byproduct recovery to full carbon recovery. The drop in energy efficiency is due to reforming carbon from an energy-depleted source, i.e., CO_2, which requires large amounts of energy for hydrogen production. This trade-off is also observed in pathways based primarily on CO_2 conversion. Waste-to-energy with carbon capture and utilization (WtE-CCU)

leads to an energy efficiency of 30–45%, ~30 percentage points less than FBSC technology with carbon recovery. Similarly, RFNBO-SAF production results in an exergy efficiency of 58%, which is 10–26 percentage points lower than pathways based on energy-containing carbon sources.

Additionally, evaluating technologies operating as stand-alone greenfield plants can overlook key opportunities, such as the re-use of existing equipment or by-products. For example, exergetic efficiency can be increased from 58 to 68% when producing RFNBO-SAF integrated into a refinery, including the co-production of hydrogen for hydrogenation demands in the plant.

RQ3: How do regulatory frameworks influence the selection and performance of defossilization pathways, particularly in terms of trade-offs between energy efficiency, carbon recovery and economics?

Regulatory frameworks can drive or hinder technological development depending on their design and alignment with other regulatory frameworks. RED III regulatory frameworks set up in the EU, requiring RFNBO production, do not support the overarching EU goals of maximizing carbon recovery (carbon-to-products) and energy-efficient production. The production of RFNBO-SAF requires CO_2 and renewable hydrogen, requiring 52% of the total exergy input as electricity to produce SAF, which leads to a cost of two to three times that of the pathways considered in this work. In contrast, the Waste Directive is more closely aligned with the EU's resource efficiency and circular economy goals by supporting recycling routes that preserve the chemical value of waste. These results show that regulatory eligibility should be evaluated together with thermodynamic and economic performance during early-stage screening.

6. Future work

Further work is necessary to formulate a complete framework for early screening technologies for the defossilization of refineries and petrochemical industries. Due to the feedstock scarcity and the low-quality waste feedstock, technologies with only once-through conversion may not be sufficient to produce the required products and minimize carbon emissions. Thus, recycling hydrocarbon byproducts within the refinery to produce more essential products is needed. How much more energy is required in order to increase carbon recovery to essential products? What is the impact on the volume of the stream to the refinery? These issues have not been addressed in detail in this thesis and will be further researched.

Additionally, more methods and topics can be included in the early-stage assessment framework, such as:

- Technology design: (i) flexibility as a key aspect and retrofitting potential to repurpose available units, (ii) retrofit recently invested technologies for long-term use, and (iii) analyzing how high internal recycling rates affect unit capacity, operability, separation demand, and downstream processing.
- Including the perspective of refinery and petrochemical systems as interconnected carbon-processing networks, where feedstocks, intermediates, byproducts, and utilities can be shared across industrial boundaries, finding potential connections.
- Investigate how carbon-containing feedstocks currently used in energy-production industries, e.g., waste-to-energy plants and industrial fuel-gas systems, could be redirected toward material or fuel production. Thus, comparing pathways that preserve carbon before combustion with pathways that recover carbon after combustion through CCU.
- Analyze how a change in the fuel market would affect the optimization of refinery products by valorizing the fuel gas or other byproducts to obtain carbon products for maritime transportation or the petrochemical industry.

Together, these developments would make the framework more suitable for identifying robust transition pathways across refinery, petrochemical industries, waste management, and energy production systems.

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