Electrification of steam cracking as a pathway to reduce the impact of the petrochemical industry on climate change

Abstract

In the Chemical Process Industry (CPI) only hydrogen production from steam methane reforming produces more greenhouse gas emissions than light olefins production. Various solutions have been proposed to reduce the CO2 emissions from olefins production. It is not clear which solution is best, particularly when you consider other sustainability factors. In this paper we report the results of our cradle-to-gate life cycle assessment (LCA) of three highly promising solutions: a so-called low-emission steam cracking furnace, the electrically driven RotoDynamic reactor (RDR), and a blue hydrogen-fired furnace. Life cycle inventory data is obtained from a first principles model for steam cracking, validated with industrial data, and combined with data obtained from process simulation. Our study shows that if the electrical supply is grey then the RDR-based cracking process has a 41% higher impact on climate change than the reference base case, a conventional plant with state-of-the-art furnaces. The low-emission furnace is only 7.2% higher. When using a mixed electrical grid, like Belgium's, the climate change impact for the RDR is 1.5% higher and the low emission furnace is 6.6% lower. With a grid that uses fully renewable power generation the RDR solution is an impressive 27% lower than the base case and the low-emission furnace 17% lower. We also analysed the impact of firing the furnaces with blue hydrogen from (a) a conventional steam-methane reformer and (b) an innovative gas-heated reformer: both (a) and (b) using carbon capture and storage (CCS). The climate change impact is reduced by 8% and 18% respectively compared to the base case. Since the blue hydrogen solution uses very little electricity, the climate change impact is insensitive to the method of electrical power generation.

Keywords: steam cracking, olefins, life cycle assessment, sustainability, carbon intensity, climate change

Abbreviations:

APH – air preheater

ASU – air separation unit

ATR – autothermal reactor

BFW – boiler feedwater

CCS – carbon capture and storage (system)

CPI – chemical process industry

EOR – enhanced oil recovery

GHG – greenhouse gases

GHR – gas-heated reformer

HPS – high pressure steam (105 atm.)

HPSS – high-pressure superheated steam (105 atm.)

HVC – high value chemicals

LCA – life cycle assessment

LCI - life cycle inventory

LCiA – life cycle impact assessment

LPS – low-pressure steam (5 atm.)

MPS – medium pressure steam (40 atm.)

NG – natural gas

RDR – rotodynamic reactor

SM – supplementary material

SMR – steam methane reformer

STOR – steam to oil (or another hydrocarbon feed) ratio

TLE – transfer line (heat) exchanger

1. Introduction

The Paris Agreement sets a target of reducing human-induced greenhouse gas (GHG) emissions to net zero by the second half of the 21st century (Masson-Delmotte et al., 2019). All economic sectors, including the Chemical Process Industry (CPI), need to defossilize. Studies suggest that achieving decarbonization within the CPI cannot be accomplished solely through energy system transformation (Luderer et al., 2018). We need to dig deeper. One approach is electrification: burning fossil fuels is replaced by using energy from green electricity (Layritz et al., 2021).

Ethylene and propylene, vital building blocks for the CPI are mostly produced from steam cracking ("Petrochemistry in Europe - Petrochemicals Europe - Petrochemicals Europe", 2022). Due to its extensive production volume (Zhao et al., 2021) and high specific energy (Zimmermann and Walzl, 2009), steam cracking constitutes the most energy-intensive process within the CPI. Steam cracking uses 8% of the CPI's primary energy (Amghizar et al., 2020) and emits 7% of the CPI's GHGs (Isella and Manca, 2022).

The European Union (EU) has pledged to decrease overall emissions in the petrochemical industry by 55% by 2030, and to be carbon neutral by 2050 (Szpilko and Ejdys, 2022). The petrochemical industry is urged to adopt energy-efficient technologies and invest in innovation. However, since current steam crackers already have thermal efficiency exceeding 90% (Zimmermann and Walzl, 2009), the EU's decarbonization strategy for the olefin industry relies heavily on transitioning to low-carbon and renewable energy sources, which is the focus of this study. The United States plans to achieve net-zero GHG emissions by 2050 (House, 2021). China is the country with the highest CO_2 emissions in the world. China's plan is reach peak emissions before 2030 and thereafter decline, reaching carbon neutrality by 2060 (Jinping, 2020). Japan also plans to become carbon-neutral by 2050 by promoting green innovations and adopting renewable energy sources (Ohta, 2021). Although this study is focused on Europe, its findings may have broader implications.

Very few LCA studies have been published on electrified steam cracking (Layritz et al., 2021) or using hydrogen as fuel (Weydahl et al., 2013). In order to close the gap, we have considered three promising technologies: a low-emission cracking furnace with an electrified separation section, an RDR, and a furnace fired with blue hydrogen. A previous study of combustion-based sustainable technologies for steam cracking is used as a benchmark (Mynko et al., 2022). A typical propane steam cracker that uses a methane-rich fraction of the process gas as fuel is the base case scenario (BASE).

Electrification of olefin production is being extensively studied by both industry and academia (Bonheure et al., 2021; Eryazici et al., 2022). Most innovations focus on electrifying the furnace or developing alternative, electrified processes to replace the furnaces such as the plasma reactor (Delikonstantis et al., 2019). There are other sections of the plant where electrification will be beneficial. Most modern light olefin plants use steam turbines (Kler et al., 2019) to drive the process gas compressor (a.k.a. the charge gas compressor) and the refrigerant compressors. The turbines rarely have an efficiency better than 45% (Durantay et al., 2021). Most propane and gasoil steam crackers are steam deficient and need boilers to make up the shortfall (Zimmermann and Walzl, 2009). One of the obvious options to reduce CO_2 emissions of a steam cracker would be to shift heat balance towards the reactor coil, or in other terms, to improve the firebox efficiency. This would allow to reduce firing rate and therefore, fuel consumption and CO_2 emissions, while the amount of heat provided to the reactor coil remains unchanged. However, in such conditions the conventional heat recovery scheme is unable to preheat the feed to optimum temperature. In order to solve this conundrum, a novel arrangement is being investigated by Technip Energies

(TechnipEnergies, 2021). In summary, the patented furnace (Oud, 2018) consists of a novel firebox design, TLE, and a heat recovery arrangement aimed to circumvent the limitations of a typical convection section and maximize the firebox efficiency. An unavoidable side effect is the reduced amount of steam and process heat produced by the furnace. This would have made such a concept rather inefficient if typical steam turbine driven compressors are used. The electrified separation section, on the other hand, will leverage the advantage of such a furnace and, according to the patent, may potentially reduce carbon dioxide emissions by up to 30%.

Several electrified cracking reactor concepts have been proposed (Gross, 2021; Tijani et al., 2022; Tullo, 2021). Coolbrook’s patented RDR cracker (Seppala et al., 2018) is one of the most advanced technologies promising up to fivefold reduction in carbon dioxide emissions compared to a conventional steam cracker (Flores-Granobles and Saeys, 2023). While both RDRs and conventional serpentine coils use the same reactants that undergo the same process, they differ in heat transfer pathway, energy source, and residence time. The RDR's short residence time is advantageous for light olefin yields (Gholami et al., 2021), making it potentially the most selective cracking technology. Unlike the low–emission furnace, in the RDR-based cracking complex the energy source for both furnace and separation areas is electricity. Several RDR cases are considered in this study to quantify the environmental impact and the effect of the electricity mix.

According to preliminary calculations and literature data (Eryazici et al., 2022), electrification of steam cracking would be rather counterproductive in case an ordinary, fossil–dominated electricity grid is used. And this is the case for the average electricity mix of the European Union (Eurostat, 2022). Therefore, during the transition period in conditions that do not allow using low carbon electricity, utilization of fossil energy coupled with carbon capture and storage system (CCS) could be preferable. Although post-combustion CCS has been covered in our previous work (Mynko et al., 2022), a pre-combustion option is worth considering as well. So-called blue hydrogen (Yu et al., 2021) combustion has been selected due to backward compatibility and the high industrial interest: in the best-case scenario, only burners must be replaced to adapt a conventional furnace for hydrogen combustion. Blue hydrogen may be produced from natural or the fuel gas via steam reforming and consecutive water-gas shift reaction. Several reformer arrangements exist nowadays (Carapellucci and Giordano, 2020). In this work, two options have been selected: a conventional steam methane reforming (SMR) process with CCS and a more novel gas heated reformer (GHR).

The high share of greenhouse gas emissions from steam cracking makes clear that reducing the CO_2 emissions of this process is essential to reduce the CO_2 footprint of the entire CPI. Unfortunately, a clear comparison of these options is currently unavailable in the literature. Moreover, there is more than just the impact of a potential solution on climate change, such as resource consumption and health-related effects. Among others, these aspects will also be discussed in this work. This study presents innovative findings by examining and contrasting three promising, yet insufficiently investigated, approaches for diminishing CO_2 emissions in steam cracking, thus addressing a void in the existing literature. The investigation of energy-efficient cracking furnace configurations, RDR technology, and the implementation of blue hydrogen as a fuel source offers valuable insights and viable strategies for industry adoption in olefin production.

2. Methodology

This study was carried out in accordance with the ISO 14040 (ISO, 2006a) and 14044 (ISO, 2006b) standards. The objective of this study is to conduct a comparative techno-environmental analysis of three electrified ethylene production plant concepts and two hydrogen-fired steam cracking furnace concepts. The findings will be juxtaposed with previously published research on the effects of furnace improvements on environmental impact (Mynko et al., 2022).

The necessity for this study stems from the significant emissions produced by steam crackers, which pose a challenge in meeting the decarbonization targets set forth by the European Union, United States, and other regions. It is crucial to understand how to efficiently mitigate the impact of steam cracking on climate change and to identify the stages of the process that contribute significantly. The outcomes of this research will be valuable for informing future research and development of environmentally friendly ethylene plant concepts, as well as potentially influencing policy decisions and industry development. The results may be used for comparative assertions in public discussions about the viability and potential benefits of transitioning to studied technical solutions. The reason for focusing on the selected concepts is that they have been identified as some of the most promising options for ethylene production with reduced environmental impacts, driven by technical feasibility and potential emission reduction capabilities.

Due to the lack of industrial data, as some of the technologies are in an early stage of development (low-emission furnace and RDR), this work is based on first principles-based models. Operating parameters, obtained by kinetic modelling and process simulation, are used as the basis to perform LCA. Hydrogen production by steam methane reforming, on the other hand, is a mature technology. However, integrated hydrogen-fired steam cracking furnaces are not operated at this moment to the best of our knowledge. Moreover, due to the conceptual status of considered solutions, capital investments and construction-related environmental impacts cannot be assessed with a sufficient level of certainty, so this paper presents a second order LCA study. In such studies capital goods are omitted while other life cycle stages are accounted for. The geographical scope of our study is Europe.

2.1. Overview of studied technical solutions

In this research, three novel steam cracking concepts were investigated as potential solutions to reducing climate change impact of light olefin production. These technologies are: a low-emission furnace with an electrified separation section, a fully electrified RDR, and the use of blue hydrogen as a furnace fuel. Each promises substantial improvements in environmental impact, energy usage, and process efficiency, and the research provides an extensive comparison of their performance to inform future industry adoption.

2.1.1. Low-emission furnace

High temperature is needed for steam cracking, typically about 850ºC. The heat comes from the flue gases (products of combustion). The temperature of the flue gases at the radiant arch of a steam cracking furnace (bridgewall temperature) is about 1100°C. The firebox fuel efficiency of a steam cracker is usually in the range 38-47% (Zimmermann and Walzl, 2009). Note that the firebox fuel efficiency is defined as the ratio of the heat absorbed by the reactor coils to the heat released by the combustion of the fuel. A typical state-of-the-art steam cracker arrangement is shown in Figure 1A. The convection section is designed to reduce the flue gas temperature to around 100ºC. A typical convection section consists of several tubular heat exchangers, called banks. In a conventional steam cracker, the heat recovered in the convection section is used to evaporate (if applicable) and preheat the feed; superheat the dilution (process) steam; and preheat the boiler feedwater (BFW) that is fed to the steam drum. The steam drum is connected to a transfer line exchanger (TLE) where process gas is quickly cooled by exchanging heat with boiling water circulating from the steam drum. TLEs are typically operated at around 100 bar and therefore the produced steam is at about 300°C. HPS leaving the steam drum is then superheated, also within the convection section, to a controlled temperature (typically 500°C) suitable for supplying the steam to the steam turbines in the separation train.

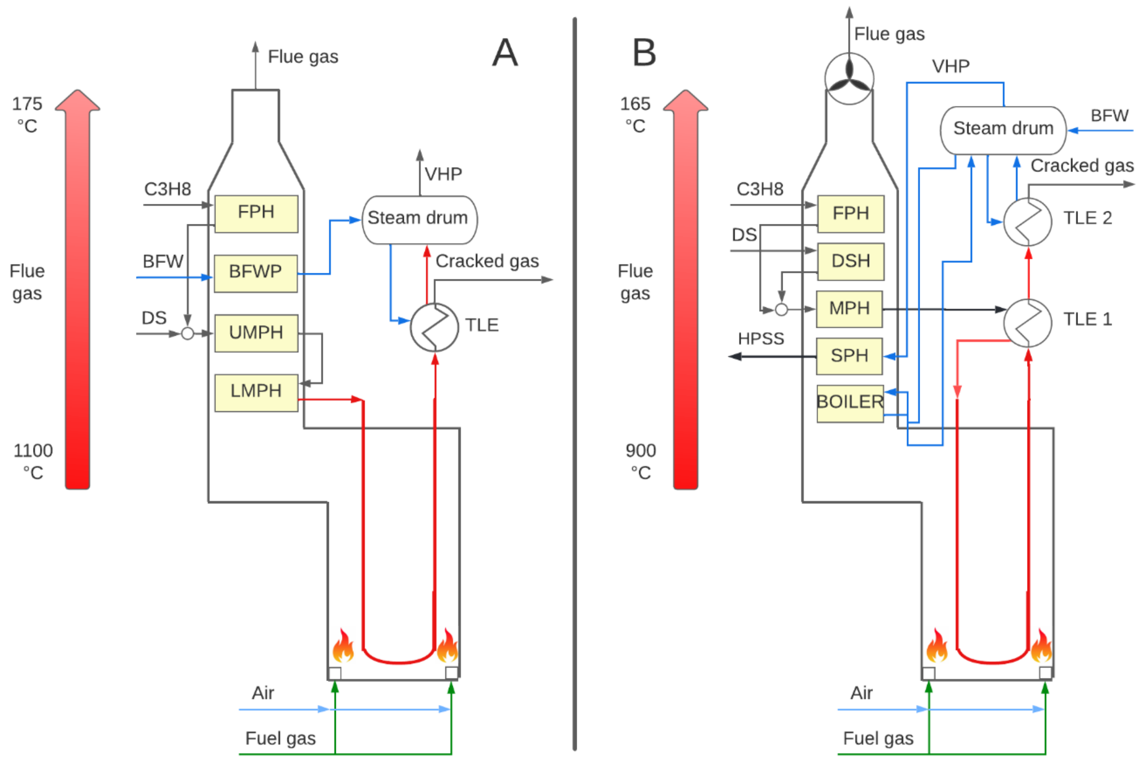


Figure 1. Arrangements of a typical steam cracking furnace (A) and a low-emission furnace (B). Modified from *(Oud, 2018).With BFW – boiler feed water, DS – dilution steam, VHP – very high pressure steam, HPSS – high pressure superheated steam, TLE – transfer-line exchanger, FPH – feed preheater, BFWP – boiler feedwater preheater, UMPH – upper mix preheater, LMPH – lower mix preheater, DSH – dilution steam heater, MPH – mix preheater and BOILER – boiler coil.*

The main idea of the low-emission furnace concept is to reduce the overall firing duty by increasing the share of heat absorbed by serpentine reactor coils that are positioned in the radiant box to provide enough heat to the cracking process with less fuel (and hence carbon dioxide emissions). To increase firebox fuel efficiency, combustion occurs in the lower part of the firebox, in a firebox designed with floor burners only or wall burners mounted close to the bottom of the furnace. Such burner arrangement maximizes the heat flux at the bottom of the furnace allowing for greater utilisation of combustion heat within radiantbox. This redesign allows an increase in firebox efficiency by 20% compared to a traditional design (Oud, 2018) and therefore, reduces fuel consumption by the same value. However, an increased share of the radiant duty in the overall furnace heat balance implies that the convection section duty is reduced and therefore it is insufficient to preheat the feed and the dilution steam mixture. To solve this challenge, Technip Energies has proposed a novel heat recovery scheme (Oud, 2018). This arrangement (shown in Figure 1B) uses two TLEs in series. In the primary TLE, the cracked gas leaving the reactor coils is quenched by exchanging heat with the preheated feed and the DS mix flow which is later fed to the reactor coil. This TLE allows for a larger share of the recovered heat to be utilized for feed preheating, and thus avoids using additional boilers even when the firebox efficiency is increased by 30%. The secondary TLE further cools the cracked gas. The convection section is altered as well.

As explained by the inventor of the low-emission furnace concept (Oud, 2018) '…A drawback of the known systems is that a lot of fuel needs to be supplied for the pyrolysis reaction. In order to reduce this fuel consumption, the firebox efficiency, the percentage of the released heat in the firebox that is absorbed by the radiant coil, can be significantly increased. However, the heat recovery scheme in the convection section of a conventional cracking furnace system with increased firebox efficiency has only limited capabilities to heat up the hydrocarbon feedstock to reach the optimum temperature to enter the radiant section... It is an aim of the present invention to solve or alleviate the above-mentioned problem. Particularly, the invention aims at providing a more efficient system with a reduced need for energy supply, and consequently, a reduced emission of CO_2 …' To increase firebox fuel efficiency, combustion occurs in the lower part of the firebox, in a firebox designed with floor burners only or wall burners mounted close to the bottom of the furnace. This redesign allows an increase in firebox efficiency by 20% (relative) compared to a traditional design (Oud, 2018) and therefore, reduces fuel consumption by the same value. To solve this challenge of reduced heat or the convection section, Technip Energies has proposed a novel heat recovery scheme (Oud, 2018). This arrangement (shown in Figure 1B) uses two TLEs in series. In the primary (first) TLE, the cracked gas leaving the reactor coils is quenched by exchanging heat with the preheated feed and the DS mix flow which is then fed to the reactor coil. Feed + steam preheat uses energy from flue gas in a conventional furnace (Figure 1A). The secondary TLE further cools the cracked gas and generates HPS, a conventional, proven concept. The convection section is altered as well. Since the mass flow and temperature of the flue gases are reduced, preheating the boiler feedwater is no longer possible. Thus, it is fed directly into the steam drum. As can be seen in Figure 1B, the convection section is used primarily to preheat the feed and dilution steam mixture. Due to the increased hydraulic resistance and lower stack temperature (and therefore weaker draft), an induced draft fan is required to ensure optimal flue gas evacuation (Oud, 2018) as in any modern furnace arrangement.

A further increase in furnace efficiency is achieved by the application of an air preheater. The air preheater bank is located at the top of the convection section and uses the remaining heat from the flue gases to preheat the combustion air (Figure 2). This arrangement will give the same process (absorbed) duty with a 30% (relative) lower fired duty. Since the flow of flue gas is lower than in the basic L-E case (Figure 1B) there is less heat for utilities and so the furnace produces HPS, not HPSS.

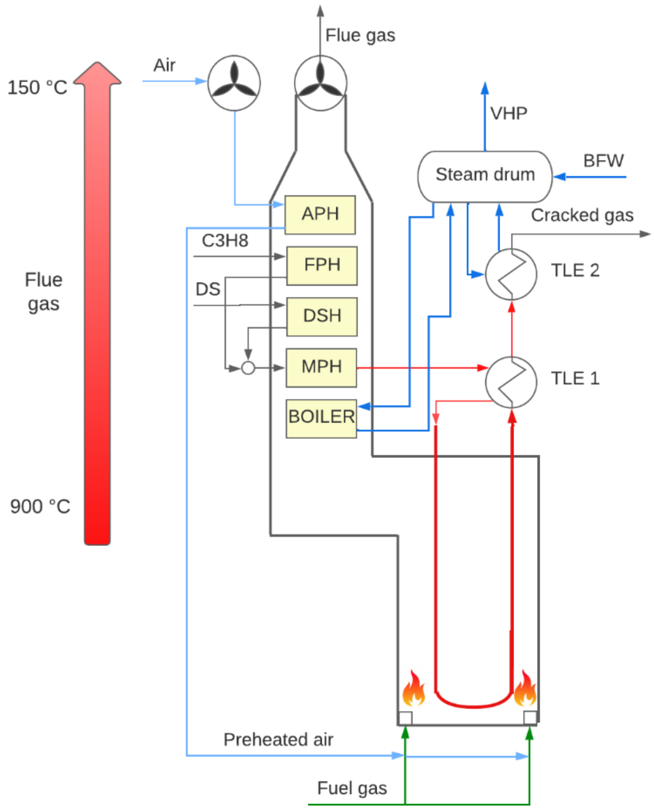


Figure 2. *Arrangement of a low-emission furnace with air preheater ('L-E APH'). Modified from* (Oud, 2018).

With BFW – boiler feed water, DS – dilution steam, VHP – very high-pressure steam, TLE – transfer-line exchanger, APH – air preheater, FPH – feed preheater, BFWP – boiler feedwater preheater, UMPH – upper mix preheater, LMPH – lower mix preheater, DSH – dilution steam heater, MPH – mix preheater and BOILER – boiler coil.

We have studied both the 'L-E' and 'L-E APH' cases. We have also evaluated the effect of the electricity mix on the results (a sensitivity study).

2.1.2. RotoDynamic Reactor

The RDR is based on Bushuev's patents (Bushuev, 1999, 2007, 2016), further developed by Coolbrook Oy. Selectivity towards light olefins (ethylene and propylene) is better than from a conventional furnace because both residence time and hydrocarbon partial pressure are lower. Rates of formation of coke are lower because the gas-solid interface in the RDR is isothermal whereas the solid surface (tube) is much hotter than the reacting gas in a furnace. The RDR is shown in Figure 3. The apparatus consists of a spinning rotor and two series of stationary blades: stator and diffuser (Figure 3a). The transonic blades of the ultra-high load rotor accelerate the preheated process gas to a supersonic velocity using mechanical energy (pure exergy) provided by an electric motor that drives the rotor.

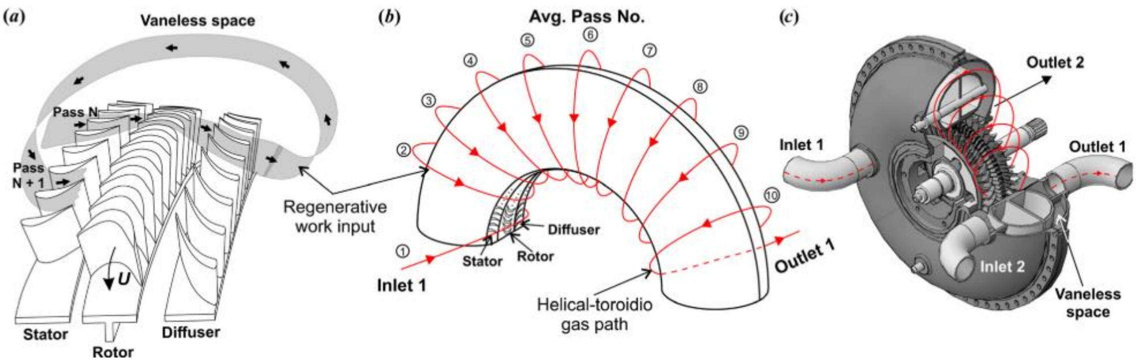


Figure 3. Scheme of the RDR reactor: a – regenerative pass, b – hemi-annulus view, c – full-annulus view (Rubini et al., 2022b)

Then the process gas is immediately decelerated in a diffuser using a shockwave system which converts the mechanical energy of the flow into heat. Blades are designed to maximize flow separation in the blade passage which leads to better mixing required to maintain constant static pressure. As the gas flow passes through the diffuser, it is turned back to the axial direction before entering the vaneless space. In other words, this apparatus can be seen as a turbomachine designed to maximize the amount of dissipated heat instead of increasing gas pressure: in fact the device is isobaric.

The RDR is multistage to get to the target conversion. Gas is guided through a toroid-shaped vaneless space into the subsequent stator passages (Figure 3b). There is a rapid stepwise increase in the process gas temperature, with no pressure increase Since the heat is introduced to the process gas via mechanical work instead of indirect heat transfer, the solid surfaces containing the gas have roughly the same temperature as the process gas. Low gas–metal surface temperature and high viscous shear stress caused by supersonic gas flow help to inhibit coke deposition (Rubini et al., 2022b). Moreover, this heating process avoids heat transfer limitations leading to the low inner volume of the multistage turbo reactor (500 times less than the volume of a tubular coil of the same capacity). This way the residence time is significantly reduced, preventing unwanted reactions.

2.1.3. Blue hydrogen combustion

When pure hydrogen is burnt in fired equipment, the direct stack CO2 emission is zero. Based on cradle-to-gate LCA (and, also the Scope 1 + Scope 2 + Scope 3 GHG Protocol), CO2 emissions from the facility producing the imported hydrogen has to be accounted for. One potential solution is to use green hydrogen generated from the electrolysis of water and powered by renewable electricity. Green hydrogen is expensive (Yu et al., 2021). An alternative is blue hydrogen, manufactured from hydrocarbon reforming and using CCS. In our study we look at two routes to blue hydrogen. It is technically rather straightforward to replace the fuel burnt in a conventional steam cracking furnace with pure hydrogen. Green hydrogen is three times more expensive than blue hydrogen (Van Geem and Weckhuysen, 2022). The price of green hydrogen is predicted to fall from 6 EUR per kg H2 (Nikolaidis and Poullikkas, 2017) to 3.7 EUR per kg H2 by 2030 (Global, 2022). That is still more than the 2023 price of blue hydrogen, 2 EUR per kg (Lagioia et al., 2023). Moreover, even at a cost of 3.7 EUR per kg H2, hydrogen continues to be a more expensive energy carrier (based on lower heating value) in comparison to renewable wind electricity at the moment of publication (2023), making electrification a preferable alternative.

In 2021, around 62% of the worldwide hydrogen demand was met through steam methane reforming (SMR) of natural gas, without carbon capture and storage (CCS) (Agency, 2022). This process is typically carried out on-site to circumvent the need for hydrogen transportation. Conventional SMR processes emit between 8.9 and 15.1 kg CO_2 eq. per kg of produced hydrogen. In contrast, blue hydrogen production results in a significantly lower emission of just 3.4 kg of CO_2

equivalent (Mehmeti et al., 2018) in terms of greenhouse gas emissions across the cradle-to-gate cycle. Due to the specific role of hydrogen in the studied system, however, a slightly unconventional arrangement has been considered in this work. Unlike most chemical processes, combustion does not require high purity of the H_2 stream. Therefore, the studied arrangement is focused on purifying the CO_2 stream for underground carbon storage or enhanced oil recovery (EOR) while producing fuel gas that contains a significant fraction of CO_2. This decision is made to reduce the capital and operating costs of such an installation. The overall scheme of the SMR process considered in this work is presented in Figure 4A. Fuel gas and steam are injected into a tubular reformer reactor at pressures of 25 bar. Heat is provided by a furnace equipped with top burners. Since the radiant efficiency of a radiant box does not exceed 60%, the remaining heat of the flue gases is used to preheat the feed and produce additional steam. The process gas leaving the reactor at a temperature of 900°C is cooled to 350°C in heat exchanger C1 and is fed into two consecutive water gas shift (WGS) reactors (HTWGS and LTWGS), where CO reacts with water to produce CO_2 and H_2. Since the reaction is exothermic, an intermediate cooler C2 is required. The process gas leaving the LTWGS is cooled to 35°C in C3 and is flashed in F1 to remove water. (Simpson and Lutz, 2007) Dry gas then enters the pressure swing adsorption module (PSA), which separates it into two streams: a hydrogen rich gas and a CO_2 rich stream. The latter does not satisfy the purity requirements for underground carbon storage or EOR. Therefore, it undergoes an additional low temperature purification step. The gas is cooled in C4, through indirect contact with a refrigerant, to a temperature that is close to the triple point of CO_2 and then flashed in two vessels, F2 and F3, operated at 23 and 8 bar, respectively. Besides CO_2 purification, the low temperature separation recovers the remaining hydrogen and unconverted methane which is then mixed and fed to the burners of the cracking furnace. According to the simulations, this arrangement produces fuel gas with ~92 vol% of H_2 and ~5 vol% of CO_2 as well as a purified CO_2 stream with ~99 vol% purity. The overall CO_2 recovery rate amounts to 87% while CH_4 conversion rate is 54%.

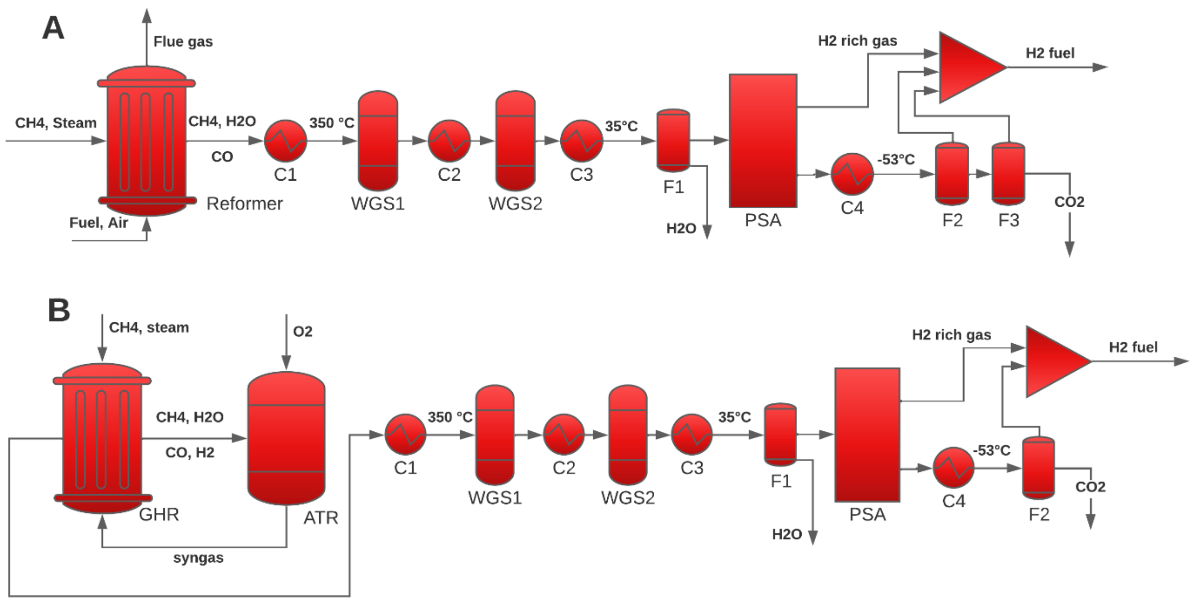


Figure 4. Simplified flow scheme of the considered blue hydrogen production processes: A – Steam Methane Reforming; B – Gas Heated Reforming.

Classical methane reforming has a disadvantage: process heat is provided by a natural gas-fired furnace that emits unrecovered CO_2 into the atmosphere. Hence, a more efficient configuration is proposed to enhance carbon dioxide recovery (Figure 4B). It consists of two connected catalytic reactors: an ATR and a GHR. First, the feed is injected into the GHR, then it is fed into the ATR. Oxidation provides sufficient heat to complete the reforming reactions. The syngas leaving the reactor at ~950 °C is then sent back to the shell side of GHR to recover its enthalpy by heat transfer to the tubes containing the catalyst. In this way, heat is provided for the endothermal reforming reaction. According to the simulations, this arrangement has a high methane conversion rate of 87% due to better heat integration and hence a reduced fraction of methane used to provide process heat. Water gas shift reactors and the separation section are effectively the same as for the SMR configuration discussed earlier with one difference: due to the lower unconverted methane stream, only one flash vessel is required for the CO_2 stream purification. Within the ATR reactor, fuel oxidation takes place, subsequently capturing the produced CO_2. Consequently, the overall carbon dioxide emissions associated with one mass unit of the resulting fuel are significantly reduced (from 5 to 1 kg CO_2). Scenarios involving a hydrogen-fired steam cracking furnace integrated with SMR and GHR reactors are denoted as 'H2-SMR' and 'H2-GHR', respectively.

2.2. Carbon intensity of the electricity mix

As a result of the high energy demand needed for the endothermic cracking reactions and the fact that modern state-of-the-art ethylene plants already use well-established and optimized technical solutions for both the reactor and separation train, there is no viable opportunity to significantly reduce the overall specific energy consumption. Shifting the fraction of heat available in the radiant box to the process (feed preheating and cracking) itself can lead to a significant reduction of the firing rate; however, lack of steam output will require an alternative way to drive the compressors of the separation section. A typical light olefin plant requires around 621 MW of fired duty and 90 MW of total compressor power for a production capacity of 125 tons of ethylene per hour and starting from propane as feed (Zimmermann and Walzl, 2009). The idea behind the low–emission furnace concept is to substitute turbine compressor drivers with electric motors. To minimize CO_2 emissions, these drivetrains must be powered by renewable or low–carbon electricity; however, the utilization of fossil electricity produced with highly efficient power generators such as combined cycle gas turbines (CCGT) (Godoy et al., 2010) will also lead to higher overall system efficiency (Durantay et al., 2021). The RDR concept, on the other hand, is aimed at replacing both process and compressor duty with electricity. Although offering a certain increase in thermal efficiency, it is highly unlikely to achieve a lower impact of cracking products on climate change than a conventional furnace if fossil electricity is used. Therefore, the carbon intensity of the electricity mix becomes a determining factor that defines the efficacy of the technologies that we have considered. An overview of the electricity mixes in our study is presented in Table 1. The selection of regions is Europe and Norway has been included as an example of a well-established renewable-based power grids. Since the impact heavily depends on the power source, an overview of the electricity sources given in Figure 5 is crucial to understand the LCA results.

Table 1. Carbon intensity of various national electricity grids for the year 2016 (Wernet, Gregor et al., 2016).

Region	**Carbon intensity [kg CO_2 eq. per kWh]**
European Union (average)	0.353
Belgium	0.203
Netherlands	0.498
Norway	0.0233

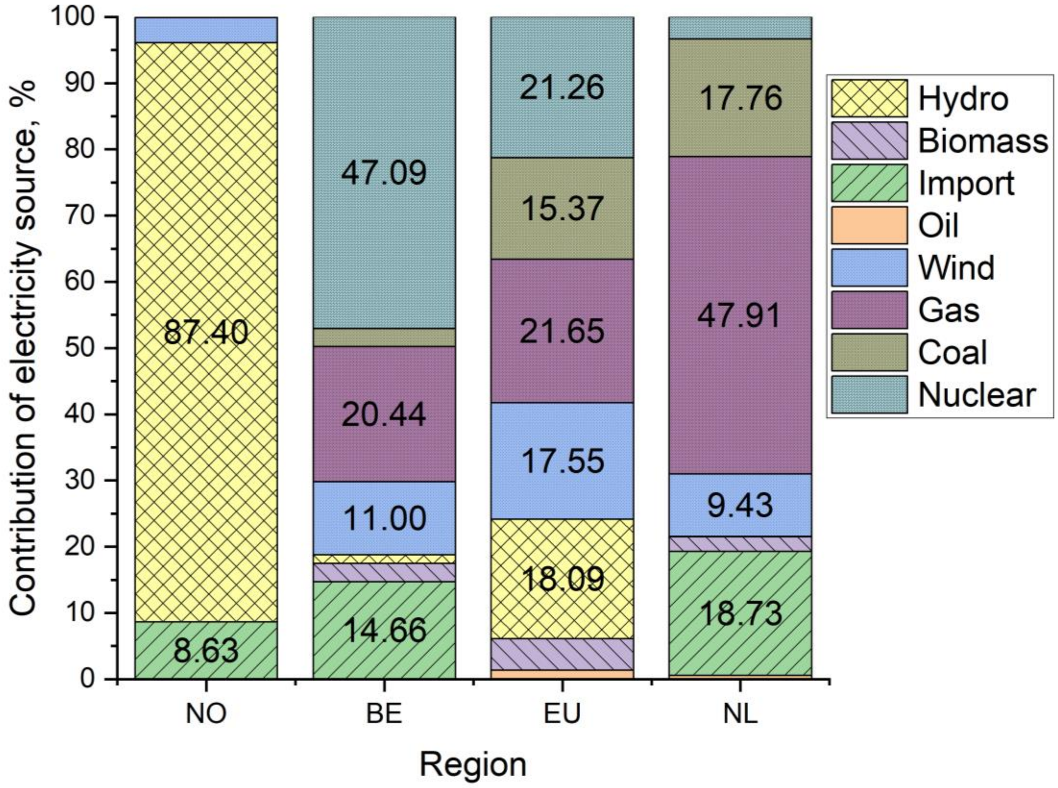


Figure 5. Overview of regional grids used in this study according to the Ecoinvent v3.7 LCI database (Wernet, Gregor et al., 2016)

2.3. System boundaries

In order to benchmark previously published data, the system boundaries and the allocation method has been defined in accordance with (Mynko et al., 2022). Since steam cracking plants produce base chemicals that have various applications, the use stage depends on factors such as the location and ownership of the plant, which are not applicable to a simulation-based study. Therefore, the scope has been restricted at the gate of the light olefin plant itself. This approach is in line with industrial guidelines (WBCSD, 2014) and is commonly referred to as a cradle–to-gate LCA. The system boundaries (Figure 6) provide an overview of all the processes and mass flows that have been simulated and used to quantify the environmental burden of the corresponding technical solutions.

2.4. Functional unit

In the steam cracking effluent, a variety of chemical species exist. The concept of high-value chemicals (HVCs) has been used to separate primary products from by-products. The HVC category encompasses the most economically significant cracking products and is defined based on the World Business Council for Sustainable Development guidelines (WBCSD, 2014) and the recommendations of PlasticsEurope (PlasticsEurope, 2017). HVCs include Ethylene, Propylene, a Benzene section of pyrolysis gasoline, Hydrogen (as a mass fraction of hydrogen-rich gas), and 1.3-Butadiene (a component of the C4 fraction). Minimum purities are selected as 99.9% for ethylene and 99.5% for propylene (European et al., 2018) whereas other products are exported as intermediate products. Every other non-energy product is treated as a by-product.

The chosen system boundaries exclude certain product treatment operations, which are typically seen as part of the downstream value chain and are not present in every ethylene plant.

Processes excluded are hydrogen valorisation through pressure swing adsorption (PSA), C4 extraction, aromatics extraction, and hydro-treatment of pyrolysis gasoline. The functional unit (FU) has been defined as the production of 1 kg of HVCs at the ethylene plant gate. Any other chemicals, such as the hydrogen fraction of methane-rich fuel gas (residue gas), are considered by-products. It should be noted that functional unit (HVCs) can consist of a mixture of these constituents in varying proportions.

Mass allocation to HVCs, accompanied by system expansion for the export of surplus methane-rich fuel gas, is implemented. It is assumed that the unburnt fuel gas is being exported. Given that ethylene plants often have connections to natural gas (NG) grids, and the composition of methane-rich gas is like NG, unused fuel gas is considered a NG substitute. The environmental impact of the process is adjusted by deducting the assumed footprint of drilling and processing natural gas with an equivalent calorific value (LHV). A similar method has been applied for steam export. All other impacts are ascribed solely to the functional unit, which is production of 1 kg of HVCs. This allocation approach, known as the avoided burden approach, is commonly used and can be found in works such as (Wiloso et al., 2012) and (Styles et al., 2016). It aligns with the guidelines provided by the WBCSD (WBCSD, 2014) and ensures that all principal intended co-products have been taken into account

2.5. Life cycle impact assessment (LCIA)

In order to assess the impact of a studied process on the environment, process data should be converted into impact factor values using an LCIA method that aggregates the following functions according to (ISO, 2006b): classification, characterization, normalization, and weighting (the latter two are optional). This stage of the study has been carried out using the Environmental Footprint v3.1 method, available in SimaPro® 9.5 that includes various midpoint LCIA models as described in the framework of Environmental Footprint (Fazio et al., 2018). LCI database ecoinvent 3.7 (Wernet, Gregor et al., 2016) is used in this work. A comprehensive description of each LCIA model utilized for each factor is provided in SM1. The GWP100 model, which is a part of the EF method, employs a 100-year time horizon and is recommended by industrial guidelines (WBCSD, 2014). Therefore, the use of GWP100a in this work is considered appropriate and the conclusions drawn are likely to scale well to other timeframes as the major contributor to climate change impact is CO_2 itself. Due to the inherent subjectivity of weighting factors, endpoint indicators are not advised by industrial guidelines (WBCSD, 2014) and were therefore not considered in this work.

While midpoint indicators may sometimes pose challenges in accurate interpretation (Bjørn and Hauschild, 2015), their usage offers a more understandable evaluation of environmental impacts and supports the early identification of key drivers for those impacts in the life cycle assessment. Moreover, midpoint indicators facilitate comparisons across LCA studies by presenting results in a consistent manner, thus improving the overall transparency and communication of the results (Jolliet et al., 2018). Although endpoint indicators offer a more aggregated understanding of overall environmental damages, midpoint indicators provide valuable information for scientists and practitioners alike in identifying priority areas and optimizing processes within the studied field.

The present study entails a comprehensive techno-environmental evaluation of a hypothetical ethylene production facility, utilizing kinetic and process flow simulations to determine direct emissions such as CO_2, CO, and NOx. In contrast, many other environmental impacts are predominantly influenced by background processes. An overview of background processes may be found in SM2.

2.6. Life cycle inventory

Due to the broad range of technical solutions, LCI had to be prepared via various tools. First, a simulation of a classically fuel-gas fired steam cracking furnace, used as a baseline scenario (or base case), has been carried out using COILSIM1D (Plehiers et al., 2019). This fundamental model has been used to simulate an industrial furnace under its typical operating conditions. The results have been verified by benchmarking against plant operating data. The capacity of one furnace is 10 tons of propane feed per hour; the same capacity is assumed for all following cases for ease of comparison. The separation of cracking effluents (commonly referred to as 'cold train' or 'separation train') has been simulated with the Petro-SIM® process simulation package to estimate recovery rates as well as compression (kinetic), heat, and cooling duties. A generic demethanizer-first separation train (Van Geem et al., 2008) with front-end hydrogenation has been selected. More details on this scenario may be found in (Mynko et al., 2022).

A hybrid approach has also been selected for the low-emission furnace. Due to intrinsic limitations, only the process side has been simulated using COILSIM1D, while the mass and heat balance of the radiant box and the heat recovery system simulated with Petro-SIM® have been estimated with the process flow scheme available in SM2. Feed rate, conversion rate, STOR, coil inlet, and outlet temperature as well as pressure drop has been assumed to be equal to the base case. As a result of the negligible difference in product yields, the kinetic and thermal duties of a cold train are assumed to be equal to those of the base case as well. However, simulation has shown that in the novel furnace arrangement, its heat recovery system is unable to produce enough steam to drive all the main compressors whilst providing sufficient heat duty. Therefore, most compressors are driven by electric motors. The process flow diagram used for simulation is available in SM3. Simulations have shown that a furnace without an APH ('L-E' case) has enough recovered heat to produce enough steam to satisfy the heat duties of the separation train, as well as 14% of the required shaft power. The remaining duty is provided with grid-powered electric motors. A conservative estimate of the efficiency of the electric drivetrain is taken as 92% (Gómez et al., 2022). Overall, such an approach represents a realistic scenario for separation train electrification.

Adding APH to the low-emission furnace embodiment ('L-E APH' case) reduces the amount of waste heat available for recovery even more. There is not even enough steam in the system to satisfy the heating requirements of the fractionation train. Therefore, an additional flow of 1200 kilograms of HPSS is provided through a standalone utility boiler, and all shaft power is provided by electric motors. All simulation results are presented in Table 2.

Table 2. Results of whole plant simulation used as life cycle inventory: 'L-E' – low-emission furnace, 'L-E APH' – low-emission furnace with combustion air preheater, RDR – RotoDynamic reactor, 'H2-SMR' – hydrogen firing furnace with steam methane reformer, 'H2-GHR' – hydrogen firing furnace with gas heated reformer.

Flow name	Flow type	Unit	BASE	'L-E'	'L-E APH'	RDR	'H2-SMR'	'H2-GHR'
Fossil C_3H_8		kg	8600	8600	8600	8590	8600	8600
Grid electricity	Inlet	kWh	-	3390	3940	1.20×10^4	314	637
Boilers duty (natural gas)[a]	Inlet	MJ	4.61×10^4	-	3750	6370	4.29×10^4	5.01×10^4
Oxygen (gaseous, from integrated ASU)	Inlet	kg	-	-	-	-	-	1050
Hydrogen	HVC product	kg	93.7	93.7	93.7	107	93.7	93.7
Ethylene	HVC product	kg	3400	3400	3400	3710	3400	3400
Propylene	HVC product	kg	1520	1520	1520	1520	1520	1520
Butadiene	HVC product	kg	235	235	235	267	235	235

Benzene	HVC product	kg	149	149	149	96.0	149	149
H_2 product, other	By-product	kg	38.7	38.7	38.7	35.4	38.7	38.7
Ethylene product, other	By-product	kg	1.03	1.03	1.03	0.910	1.03	1.03
Ethane	By-product	kg	342	342	342	302	342	342
Propylene product, other	By-product	kg	110	110	110	111	110	110
Crude BBB product, other	By-product	kg	175	175	175	184	175	175
Untreated pygas product, other	By-product	kg	363	363	363	280	363	363
Cracked gasoil product	By-product	kg	32.5	32.5	32.5	3.04	32.5	32.5
Captured CO_2	By-product	kg	-	-	-	-	1740	2740
HVC yield	Parameter	%	62.8	62.8	62.8	66.2	62.8	62.8
Fuel gas export	Energy export	kg	958	1190	1310	1470	605	981
Fuel to furnace	Inner flow	kg	1180	946	828	497[b]	1540	1160
Furnace CO_2 emissions	Emissions	kg	3180	2540	2230	1330[b]	2420	452
Furnace CO emissions	Emissions	kg	0.200	0.160	0.140	0.0800	0.150	0.0300
Furnace NO_X emissions	Emissions	kg	1.25	1.00	0.870	0.480	1.62	0.990
Heat export (steam)	Energy export	MJ	3.70×10^4	-	-	-	4.20×10^4	3.75×10^4

[a] – does not account for decoking steam generation

[b] – in case of RDR, furnace is replaced by gas-fired feed mix preheater

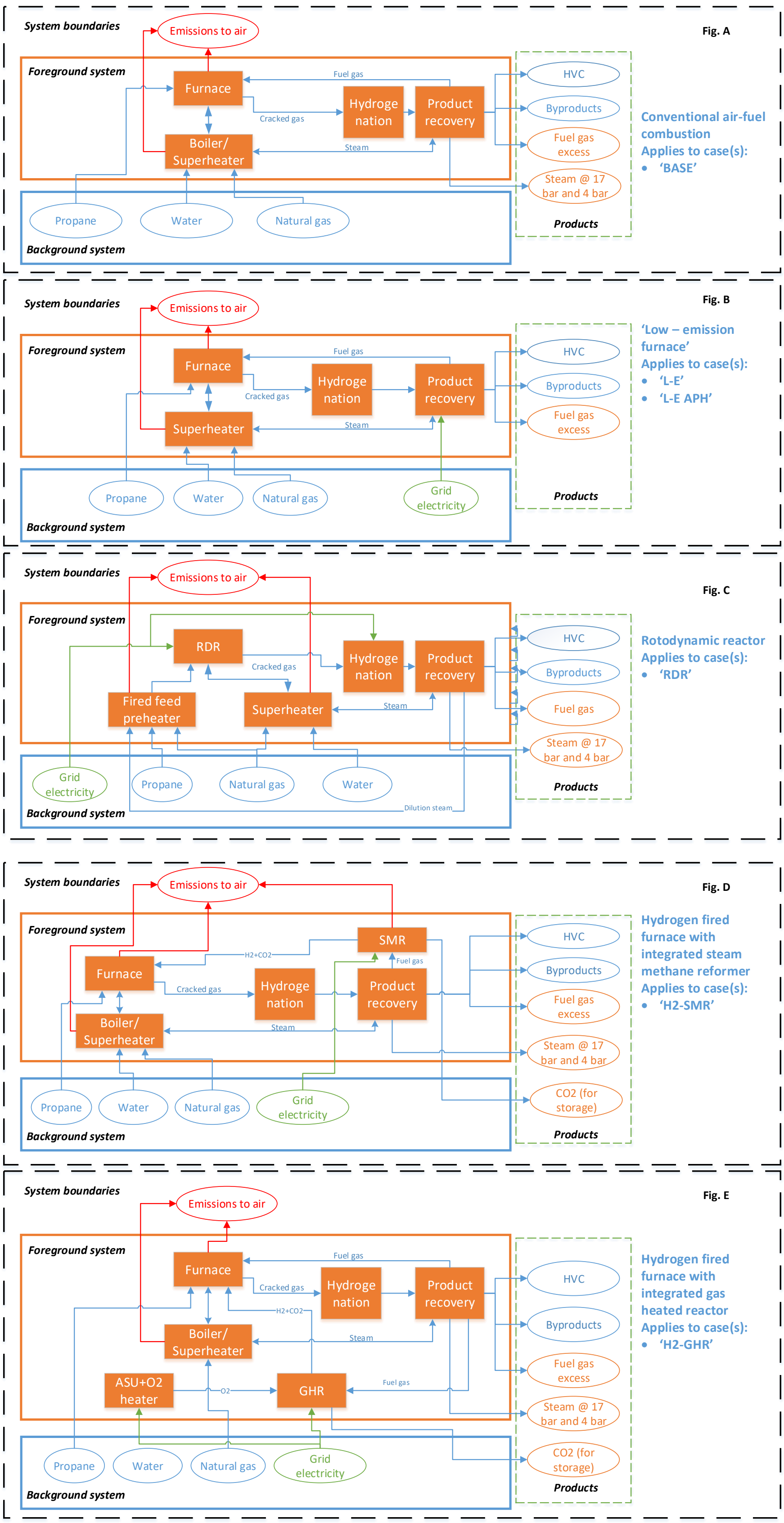


Figure 6. System boundaries of scenarios considered in this work. A – conventional ethylene plant, baseline scenario (BASE); B – low-emission furnace with electrified separation train ('L-E', 'L-E APH'); C – RotoDynamic reactor with electrified separation train (RDR); D – hydrogen-fired SC

furnace with integrated SMR and CCS (‘H2-SMR’); E - hydrogen-fired SC furnace with integrated GHR and CCS (‘H2-GHR’).

Further details regarding the simulation setup and selected assumptions can be found in SM5, which also contains the energy balances for all analysed scenarios. In compliance with the guidelines set forth by the WBCSD (WBCSD, 2014), the inherent uncertainties present in the research have been analysed using a semi-quantitative approach through the implementation of a standard Pedigree matrix (Tyson, 1976), which can be found in SM6. Each material and energy flow is assigned a score ranging from 1 (highest score) to 5 (lowest score). Specific mass and energy flows, such as cooling water and low-voltage electricity, have been omitted in line with the cut-off criteria, which state that processes with a total potential contribution to global warming impact below 5% are not considered (Joint Research et al., 2010)

3. Results and discussion

Five potential low-carbon steam cracking solutions have been benchmarked using a LCA methodology with 16 impact indicators, as suggested by industrial guidelines (WBCSD, 2014). The environmental impacts of producing one functional unit via the considered pathways are presented in SM7.

The overall impact on climate change of one functional unit at gate and contributions of each essential part of the mass and energy balance is visualized in

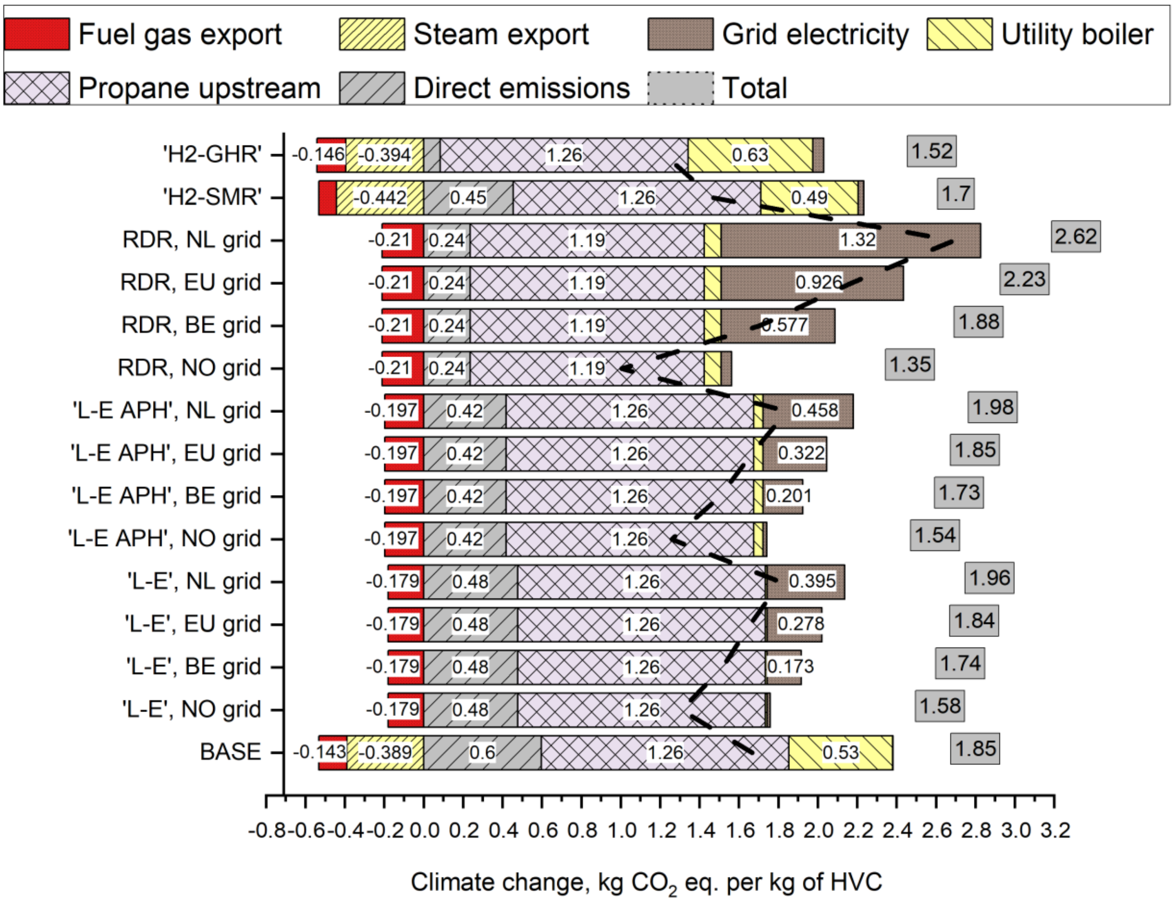
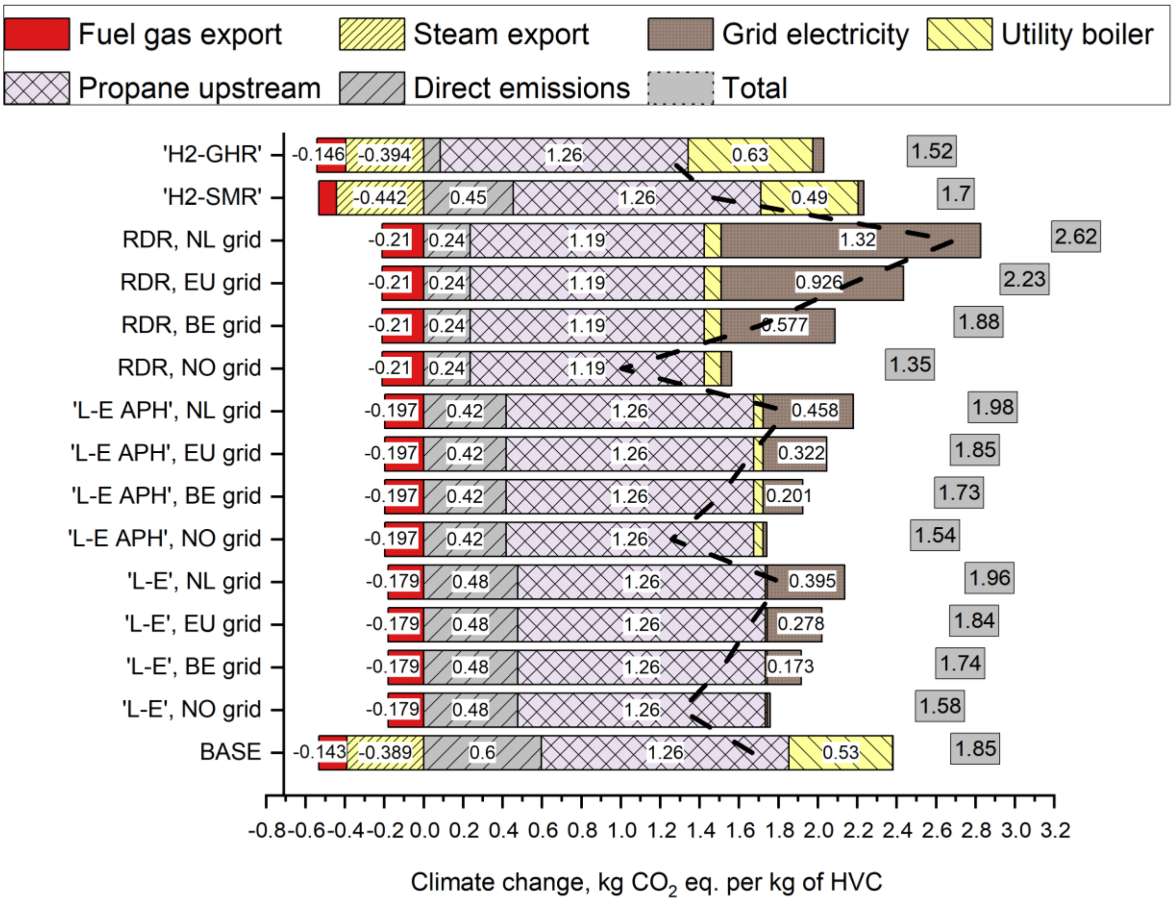


Figure 7.



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503 Figure 7. Contribution of each production stage to the impact on climate change of a
504 functional unit at gate (GWP 100a).

As can be seen in

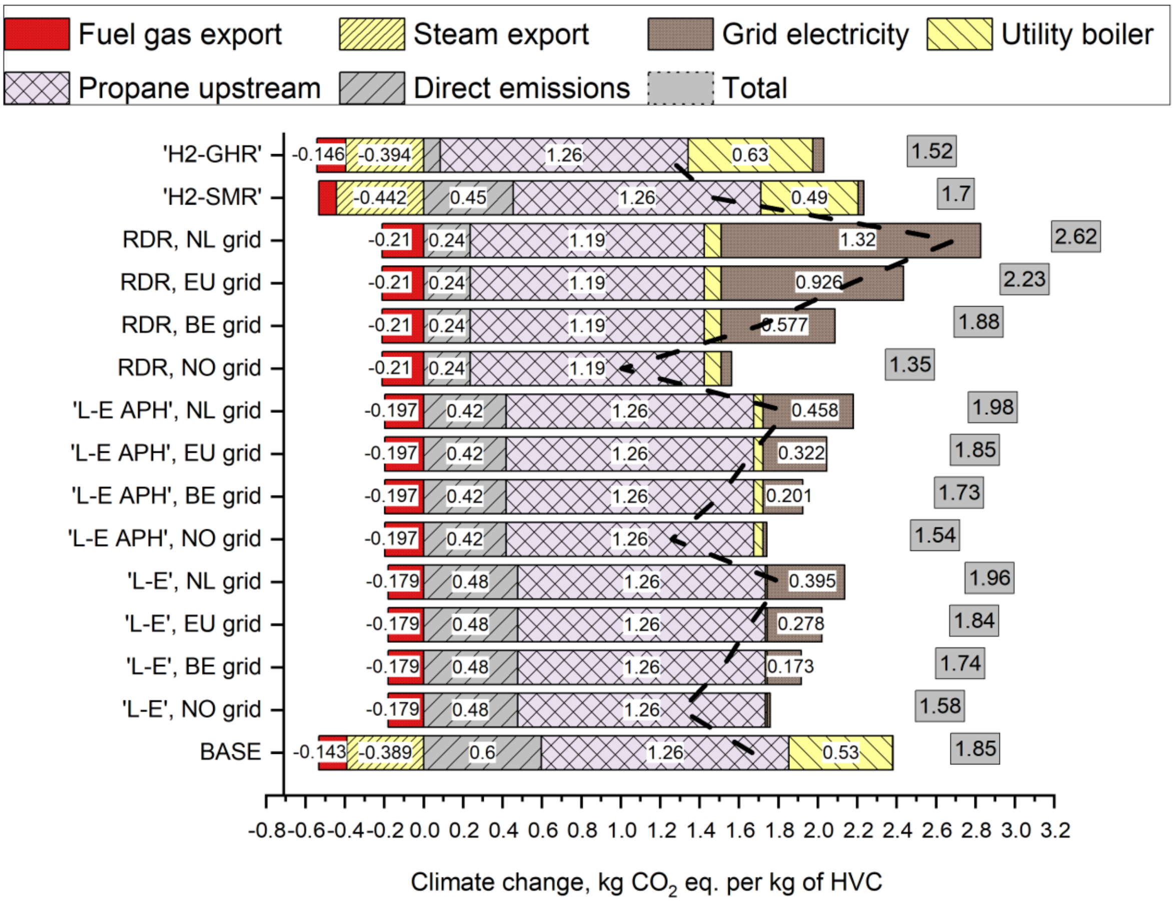

Figure 7, the impact of the electricity grid on climate change defines the applicability of the electrified solutions and, in the RDR case, becomes a major contributor if fossil energy is used to generate electricity. Such an outcome is expected since the amount of enthalpy that must be provided to the propane feed to convert it into high-value chemicals is roughly the same for all scenarios but the efficiency of fossil fuel powerplants is much lower than the thermal efficiency of a typical steam cracking furnace. The efficiency of a CCGT can range from 50% to 62% based on the fuel's LHV (Talah and Bentarzi, 2022); however, a conservative estimate of 55.5% efficiency is assumed for a modern natural gas-fired combined cycle power plant (Babaei Jamnani and Kardgar, 2020). Meanwhile, according to our BASE case furnace simulations, the overall (radiant box and convection section) efficiency of a cracking furnace exceeds 90%. This unavoidable destruction of exergy suggests a preliminary conclusion: electrification of steam cracking makes sense (from an environmental perspective) only if low–carbon electricity is available. This relation between the carbon intensity of grid electricity and high-value cracking products is illustrated in Figure 8. It can be noticed that in the case of mostly fossil grids, such as the grid in the Netherlands, all electrified options are worse than the baseline. If the electricity mix is mostly renewable, as the Norwegian grid, electrified scenarios show a tremendous advantage over traditional combustion options. The RDR reduces the impact of cracking products on climate change (at the gate) by 26.8%; the best result in this study. Low-emission furnace has shown moderate results; the impact of electricity is higher for the solution with an APH due to the higher fraction of duty that is replaced by grid electricity. If mostly renewable electricity is used, the ‘L-E’ embodiment can lead to an overall reduction of 14.5% and the ‘L-E APH’ - up to 16.5%. Figure 8 also illustrates ‘breakeven points’ – values of the electricity carbon intensity that reduce the climate change impact of HVCs production compared to the three non-electrified solutions: the conventional furnace and two blue hydrogen solutions. In fact, these two blue hydrogen solutions use electricity as well. However, because of the low contribution of electricity to the overall impact on climate change (up to 4% for ‘H2-GHG’) no sensitivity study has been carried out. Therefore, EU electricity mix has been used for LCA. The primary contributor, propane upstream, relies exclusively on process selectivity and, as a result, remains constant across all scenarios except for RDR, which provides slightly enhanced light olefin yields. The impact of this factor has been assessed by utilizing the ecoinvent unit process for the global propane market, under the assumption that it serves as a by-product of natural gas refining. While the focus of this study does not include upstream processes, it is worth noting that research on low-carbon propane production is underway. The technical solutions under investigation range from the optimisation of flaring losses to the use of fully renewable feedstock (Kallio et al., 2014; Ma et al., 2016; Payne et al., 2023).

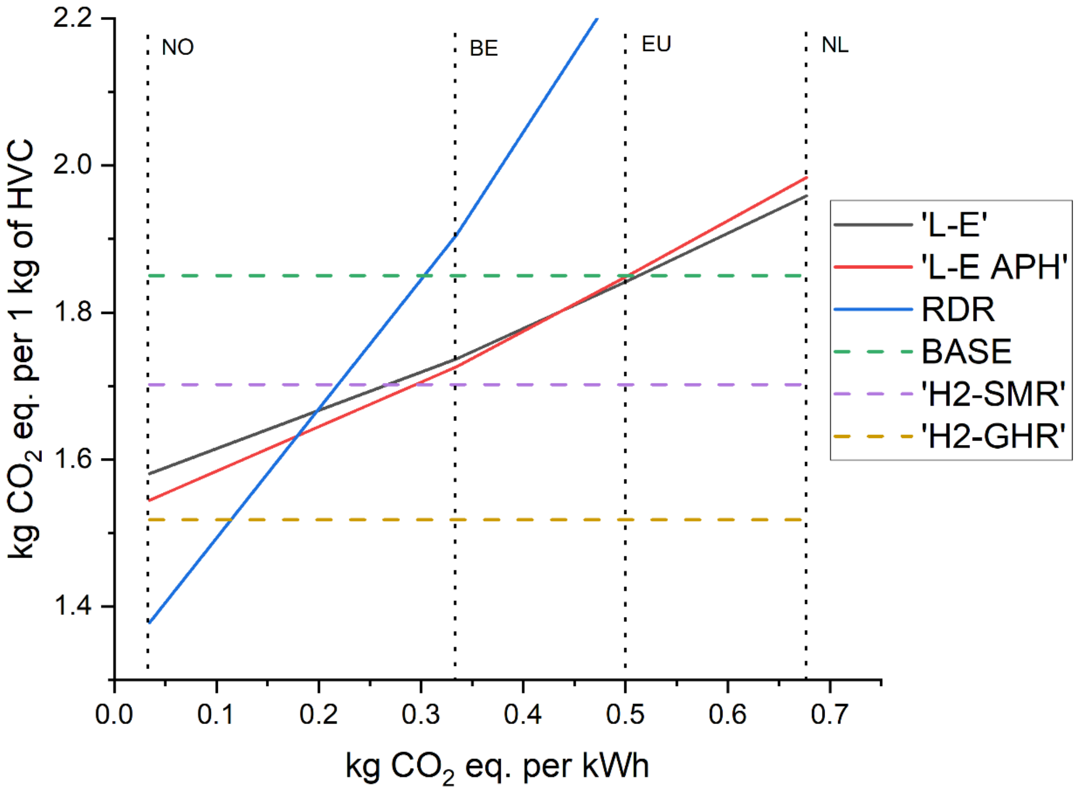

Figure 8. Impact of the carbon intensity of electricity on the sustainability of cracking products. ‘H2-SMR’ and ‘H2-GHG’ scenarios are benchmarked with EU electricity mix only.

Our results show that both ‘L-E’ and ‘L-E APH’ furnaces offer a better performance based on the carbon intensity if the grid carbon intensity is below 500 g of CO_2 per kWh while the RDR needs electricity with carbon intensity below 300 g of CO_2 per kWh to outperform the baseline. Nonetheless, due to the inherent uncertainties present in this study and the environmental impacts associated with capital expenditures (CAPEX), further decarbonization of electricity is necessary to effect significant change.

3.1. Electrified olefin plants

In this LCA study, a total of 12 electrified steam cracker scenarios have been examined: two gas-fired furnaces with electrified separation sections, and an RDR reactor with an electrified separation train, all powered by four distinct electricity mixes. The core concept of an electrified ethylene plant is to provide the requisite process heat or shaft duty using electricity rather than fossil fuel combustion. Therefore, it is fitting to initiate the discussion with a comprehensive overview of the environmental impact associated with the various electricity markets being considered. These impacts are illustrated in Figure 9 using a normalized format, where 100% signifies the highest impact value within each respective category.

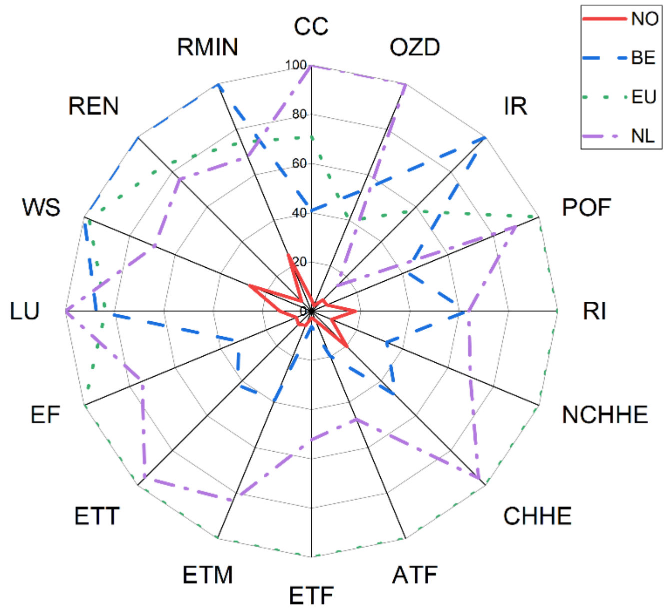

Figure 9. Normalized environmental footprints of one unit of electricity at given country market. (Wernet, G. et al., 2016)

Letters on each axis stand for impact category: CC – Climate change [kg CO_2 eq.], OZD – Ozone depletion [kg CFC11 eq],IR – Ionizing radiation, HH [kBq U-235 eq.], POF – Photochemical ozone formation (human health impact) [kg NMVOC eq.], RI – Respiratory inorganics [disease inc.], NCHHE – Non-cancer human health effects [CTUh], CHHE – Cancer human health effects [CTUh], ATF – Acidification terrestrial and freshwater [mol H+ eq.], ETF – Eutrophication freshwater [kg P eq.], ETM – Eutrophication marine [kg N eq], ETT – Eutrophication terrestrial [mol N eq], EF – Ecotoxicity freshwater [CTUe], LU – Land use [Pt], WS – Water scarcity [m^3 depriv.], REN – Resource use, energy carriers [MJ], RMIN – Resource use, mineral and metals [kg Sb eq].

In a comparable manner, the environmental impact associated with producing 1 kg of high-value chemicals using the investigated processes is illustrated in Figure 10. For the sake of clarity, the Belgian and European grids have been excluded, with only the extreme cases (Norwegian and Dutch markets) being depicted. Additionally, specific impact categories have been omitted for the same reason. A comprehensive breakdown can be accessed in Supplementary Materials SM7 and SM8.

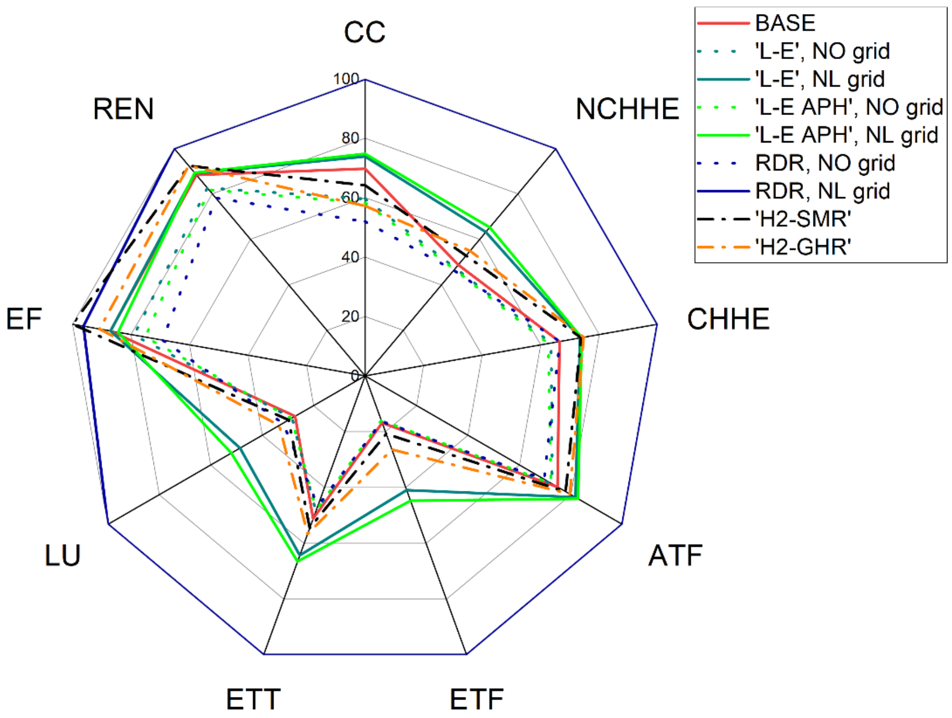

Figure 10. Normalized environmental footprints of one functional unit at gate for studied steam crackers.

Abbreviations: please see Figure 9.

The influence of various electrical grids is significantly more noticeable for the 'L-E APH' approach and the RDR, as a greater proportion of the overall energy demand is supplanted by electricity. With the highly renewable Norwegian electricity mix, both electrified crackers outperform a basic furnace in all categories except for land use and water scarcity. Both can be attributed to the use of hydropower in Norway which unavoidably leads to land use for hydropower reservoirs (Dorber et al., 2018), and water loss due to evaporation. However, some authors disagree with the second statement (Bakken et al., 2015) and suggest that such reservoirs may rather increase water availability in certain regions. In the case of the Belgian national grid, ionizing radiation stands out mostly due to a significant share of nuclear power. The indicators of eutrophication, land use, and water scarcity are partially driven by biofuels (Van Wijnen et al., 2015), which are fairly widespread in both the electricity grid in Belgium and the Netherlands, as well as on average in the EU. In the case of the modern-day, power grid in the Netherlands, all electrified crackers perform worse than the base case due to a significant share of fossil fuels that cause emissions of CO_2, $NO_{X,}$ and PM 2.5 during combustion, leading to terrestrial, marine, and freshwater eutrophication (Jonson et al., 2017). It is important to note that despite the significant variation, absolute values of the impact on eutrophication are low. For example, the highest contributor to terrestrial eutrophication, RDR powered with the EU grid, is responsible for only 2.7×10^{-2} mols of equivalent nitrogen emissions to the soil. All issues are amplified for the EU average grid due to its larger share of solid fuels – mostly brown coal and anthracite – that produce higher flows of NO_X, $SO_{X,}$ and particulate matter when burnt. It should be recognised that even the most efficient electrified solution, the RDR powered by a fully renewable grid (climate change impact reduction of 27% compared to the base case), still falls short of the biogas fired furnace's potential for reduction (which stands at 43%) (Mynko et al., 2022). However, the feasibility of biogas production at sufficient scale remains uncertain. As for human health impacts, both cancerogenic and other pollutants are mostly emitted by two background processes: propane extraction and (for electrified cases) grid electricity generation. Due to its higher selectivity toward light HVCs and minimal flowrate of fuel gases, if connected to a renewable grid, the RDR gives a reduction of 6% in non-cancer health effects, while 'L-E' and 'L-E APH' result in a reduction of 3% and 4%, respectively. In any other grid, the impact of electricity production completely offsets the benefits. The effect is more pronounced in the case of 'RDR, EU grid', which has an impact factor value that is 120% higher. The situation is slightly different for cancerogenic emissions. As we can see in Figure 9, even with the Norwegian power grid, RDR offers a minimal (1.3%) advantage over a basic steam cracker. On the other hand, 'L-E' and 'L-E APH' reduce the impact by 4% and 6%, respectively, due to the significantly reduced flue gas flow, the higher avoided burden due to fuel gas export, and the low electricity consumption. A low-emission furnace successfully valorises a 'low-hanging fruit' situation by replacing inefficient steam turbines with electric motors.

Another significant impact factor is resource depletion. Since this is a second order LCA study, construction (CAPEX) related impacts are only accounted for background processes. In this way the electrified scenarios do not lead to improvements in the 'resource use, mineral and metals' impact factor; moreover, in the case of RDR, NO grid we can observe a 5% increase that can be attributed to the impact of electricity supply that offsets the reduction in propane feed consumption. However, according to the literature, the RDR is expected to be at least one order of magnitude smaller and lighter than a conventional steam cracker of the same capacity (Rubini et al., 2022a). Currently, first–order LCA is not feasible due to the prototype stage of this reactor and the absence of industrial data; however, it might be a topic of further studies. Regarding energy resources, all three electrified concepts are beneficial if the renewable grid is considered, leading to a reduction of 6.5%, 7.5%, and 13%, respectively. In the case of any other electric grid, in which a significant fraction of fossil fuels is used, electrification becomes unfavourable and leads to an increase in impact factor value by up to 17% for the RDR, Belgian grid scenario. This can be explained by the low thermal efficiency of nuclear power plants due to their inability to use the combined cycle and reliance on steam turbines alone.

3.2. Blue hydrogen fired furnaces

Blue hydrogen combustion is an alternative pathway to defossilize steam cracking which becomes especially relevant if low-carbon electricity is not readily available. An integrated methane reformer is effectively a pre-combustion CCS with the potential to reduce all Scope 1 emissions (GHG Protocol) to zero. However, due to techno-economic constraints and high purity requirements for the captured CO_2 stream, a perfect carbon capture rate is not achievable, and hence, hydrogen-rich fuel flow still contains a significant fraction of CO_2 and CH_4. This and the emissions from the reforming furnace itself are the reasons why the 'H2-SMR' scenario can only reduce the on-site emissions by 24% and the overall impact of a functional unit on climate change by 8%. Furthermore, the low thermal efficiency of SMR leads to substantial additional firing duties. For the entire plant, it is 30% higher than the plant based on a basic furnace and causes a 4.5% increase in the impact indicator of 'resource use, energy carriers. The novel 'gas heated reformer' solves most of the problems. Since the design integrates the heat supply and reforming process into a single ATR, all the CO_2 that is generated during the process is fed to the CCS, resulting in the absence of stack emissions and a better carbon capture rate. Moreover, due to a better heat integration scheme that uses process heat for reforming reactions that occur in the 'H2-GHR' case, the overall heat duty is reduced. When integrated with a cracking furnace, this reformer leads to a reduction in Scope 1 greenhouse gas emissions by 86% and 18%, respectively, for an overall impact of 1 kg of HVC on climate change. This is the second-best result after the RDR, NO grid scenario. The captured CO_2 can be further stored in underground caverns such as depleted oil or gas wells, used in the chemical industry, or for EOR. A detailed comparison between blue hydrogen-fired furnaces and the baseline is presented in Figure 11.

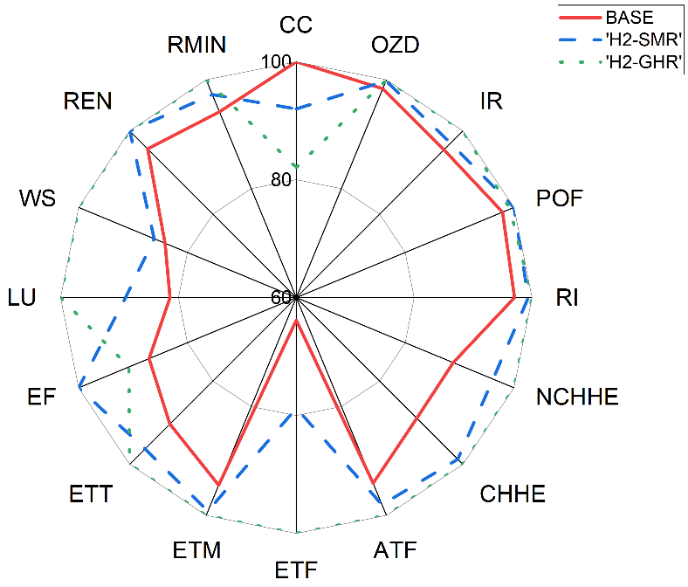

Figure 11. Normalized environmental footprints of one functional unit at gate for blue hydrogen fired steam crackers.

Abbreviations: please see Figure 9.

Impact factors such as ozone depletion, photochemical ozone formation, respiratory inorganics, both cancer and non–cancer human health effects, acidification terrestrial and freshwater, eutrophication terrestrial, and marine as well as resource use factors are dictated by the avoided burden from fuel gas exports and electricity consumption. Specifically, in the case of ‘H2, SMR’ the reformer furnace is a significant contributor to NO_X emissions. On the other hand, the ‘H2, GHR’ technology does not have an additional fired heater and is more fuel efficient than a basic furnace, but requires more electricity to produce and preheat the oxygen needed to operate the ATR. In this case, these two effects cancel each other out so similar results as for the ‘H2-SMR’ and ‘H2-GHR’ scenarios are obtained. In the case of freshwater eutrophication, the outcome is strongly influenced by the footprint of the electricity which is used. This can lead to a 67% increase in the GHG scenario. The same trend is observed for land use and water scarcity. The situation is reversed for the freshwater ecotoxicity indicator which is mostly driven by the footprint of the hydrocarbon extraction so the plant with SMR has a 15% higher impact than the baseline due to the higher fuel consumption. In comparison with our previous work (Mynko et al., 2022), simulations indicated that oxyfuel furnace with carbon capture proved more effective, offering a 24% reduction in overall climate change impact, compared to an 18% reduction achieved by 'H2-GHR'. This advantage of oxyfuel largely stems from its ability to capture all stack CO_2, whereas the methane reformers, used in blue hydrogen production, persist in contributing to the overall climate change impact.

4. Conclusions

Three major concepts of low–carbon cracking technology have been compared. The LCA with the midpoint Environmental Footprint method shows that when low-carbon renewable electricity is available, the RDR, coupled with an electrified separation train, is the optimal solution that reduces the impact of HVCs on climate change by 27% and reduces the consumption of non–renewable energy resource by 13% without any significant side effect. Only the indicators of water scarcity and inorganic resource consumption have increased by 6% and 5%, respectively, due to the massive share of hydropower in our reference renewable grid scenario (Norwegian grid) and the corresponding infrastructure and heavy machinery needed for hydroelectricity production. If the available grid is grey, however, the RDR-based cracker becomes less sustainable because of unavoidable exergy destruction which happens in any heat engine of a powerplant. If mixed networks are used, such as the Belgian grid, a low-emission furnace becomes a viable option capable of reducing the impact on climate change by 6% and 6.6% for basic and APH embodiments, respectively. This is achieved by replacing inefficient steam turbines in the separation train with electric motors while providing process duty with a hydrocarbon gas furnace, the efficiency of which exceeds 92%. Our results also prove that hydrogen-fired furnaces with integrated methane reforming units equipped with CCS systems are a promising modern-day alternative to electrification. An advantage is that the steam crackers can run on indigenous fuel from the methane-rich fraction of the cracked gas. The integration of a more efficient gas-heated methane reformer reduces the global warming potential by 18%. It is important to note that these solutions also have disadvantages, such as an increased impact in most of the NO_X, SO_X, and PM-related impact categories, as well as a 4.5% higher consumption of energy resources. In addition to the reduction of greenhouse gas emissions, also a relatively pure CO_2 stream is produced that can be used in the chemical or petroleum industry. An additional advantage of this concept is that there is an easy shift towards green hydrogen produced via water electrolysis. Therefore, on short notice, we believe that this technology will be implemented soon in case provided that prevailing economic conditions facilitate decarbonization by means of carbon taxes, incentives or consumer preferences.

Our findings offer significant insights and tactics for the petrochemical industry to progress towards accomplishing decarbonization objectives established by regions such as the European Union, United States, China, and Japan. However, no single solution is adequate to attain climate neutrality, given the immense volume of greenhouse gas emissions generated during fossil feedstock production. Consequently, future research should focus on promoting a circular economy and transitioning the life cycle of plastics toward a closed loop with minimal carbon loss.

5. Acknowledgments

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6. References

Agency, I.E., 2022. Global Hydrogen Review 2022.

Babaei Jamnani, M., Kardgar, A., 2020. Energy-exergy performance assessment with optimization guidance for the components of the 396-MW combined-cycle power plant. Energy Science and Engineering 8(10), 3561-3574.

Bakken, T.H., Kjosavik, F., Killingtveit, Alfredsen, K., 2015. Are reservoirs water consumers or water collectors? Reflections on the water footprint concept applied on reservoirs. Water Resources Management 29(14), 4919-4926.

Bjørn, A., Hauschild, M.Z., 2015. Introducing carrying capacity-based normalisation in LCA: framework and development of references at midpoint level. The International Journal of Life Cycle Assessment 20(7), 1005-1018.

Bonheure, M., Vandewalle, L., Marin, G., Van Geem, K., 2021. Dream or reality? Electrification of the chemical process industries. CHEMICAL ENGINEERING PROGRESS 117(3), 37-42.

Bushuev, V.A., 1999. Method for producing lower olefins, reactor for the pyrolysis of hydrocarbons and device for quenching pyrolysis gases. PCT/RU1999/000038.

Bushuev, V.A., 2007. Process for producing low-molecular olefins by pyrolysis of hydrocarbons.

Bushuev, V.A., 2016. Bladed reactor for the pyrolysis of hydrocarbons. Google Patents.

Carapellucci, R., Giordano, L., 2020. Steam, dry and autothermal methane reforming for hydrogen production: A thermodynamic equilibrium analysis. Journal of Power Sources 469, 228391.

Delikonstantis, E., Scapinello, M., Stefanidis, G.D., 2019. Process Modeling and Evaluation of Plasma-Assisted Ethylene Production from Methane. Processes 2019, Vol. 7, Page 68 7(2), 68-68.

Dorber, M., May, R., Verones, F., 2018. Modeling Net Land Occupation of Hydropower Reservoirs in Norway for Use in Life Cycle Assessment. Environmental Science and Technology 52(4), 2375-2384.

Durantay, L., Van Gemert, D., Lava, J., Spannagel, D., 2021. A success story of steam turbine replacement by high speed electric system driven compressor. PCIC Europe.

Eryazici, I., Ramesh, N., Villa, C., 2022. Electrification of the chemical industry—materials innovations for a lower carbon future. MRS Bulletin, 1-8.

European, C., Joint Research, C., Brinkmann, T., Falcke, H., Holbrook, S., Sanalan, T., Roth, J., Delgado Sancho, L., López Carretero, A., Clenahan, I., Roudier, S., Zerger, B., 2018. Best Available Techniques (BAT) reference document for the production of large volume organic chemicals. Publications Office.

Eurostat, 2022. "Electricity production, consumption and market overview - Statistics Explained". https://ec.europa.eu/eurostat/statistics-explained/index.php?title=Electricity_production,_consumption_and_market_overview#Electricity_generation. (Accessed 07.27 2022).

Fazio, S., Castellani, V., Sala, S., Schau, E.M., Secchi, M., Zampori, L., Diaconu, E., 2018. Supporting information to the characterisation factors of recommended EF Life Cycle Impact Assessment methods. pp. 42-42.

Flores-Granobles, M., Saeys, M., 2023. Quantitative analysis of CO2 emissions reduction potential of alternative light olefins production processes. Green Chem 25(16), 6459-6471.

Gholami, Z., Gholami, F., Tišler, Z., Vakili, M., 2021. A Review on the Production of Light Olefins Using Steam Cracking of Hydrocarbons. Energies 2021, Vol. 14, Page 8190 14(23), 8190-8190.

Global, D., 2022. The European hydrogen economy – taking stock and looking ahead. An outlook until 2030.

Godoy, E., Scenna, N.J., Benz, S.J., 2010. Families of optimal thermodynamic solutions for combined cycle gas turbine (CCGT) power plants. Applied Thermal Engineering 30(6-7), 569-576.

Gómez, J.R., Sousa, V., Cabello Eras, J.J., Sagastume Gutiérrez, A., Viego, P.R., Quispe, E.C., de León, G., 2022. Assessment criteria of the feasibility of replacement standard efficiency electric motors with high-efficiency motors. Energy 239, 121877-121877.

Gross, S., 2021. The Challenge of Decarbonizing Heavy Industry.

House, W., 2021. FACT SHEET: President Biden Takes Executive Actions to Tackle the Climate Crisis at Home and Abroad, Create Jobs, and Restore Scientific Integrity Across Federal Government. WhiteHouse. Gov. Original edition.

Isella, A., Manca, D., 2022. GHG Emissions by (Petro)Chemical Processes and Decarbonization Priorities - A Review. Energies 2022, Vol. 15, Page 7560 15(20), 7560-7560.

ISO, 2006a. 14040: Environmental management–life cycle assessment—Principles and framework, International organization for standardization.

ISO, 2006b. International Standard 14044:2006, Environmental management - Life cycle assessement - Requirements and guidelines, ISO 14044, International Organization for Standardization. pp. 652-668.

Jinping, X., 2020. Statement by HE Xi Jinping President of the People's Republic of China at the General Debate of the 75th Session of the United Nations General Assembly. Peace 3, 5-7.

Joint Research, C., Institute for, E., Sustainability, 2010. General guide for Life Cycle Assessment : provisions and action steps. Publications Office.

Jolliet, O., Antón, A., Boulay, A.-M., Cherubini, F., Fantke, P., Levasseur, A., McKone, T.E., Michelsen, O., Milà i Canals, L., Motoshita, M., Pfister, S., Verones, F., Vigon, B., Frischknecht, R., 2018. Global guidance on environmental life cycle impact assessment indicators: impacts of climate change, fine particulate matter formation, water consumption and land use. The International Journal of Life Cycle Assessment 23(11), 2189-2207.

Jonson, J.E., Borken-Kleefeld, J., Simpson, D., Nyíri, A., Posch, M., Heyes, C., 2017. Impact of excess NOx emissions from diesel cars on air quality, public health and eutrophication in Europe. Environmental Research Letters 12(9), 094017-094017.

Kallio, P., Pasztor, A., Thiel, K., Akhtar, M.K., Jones, P.R., 2014. An engineered pathway for the biosynthesis of renewable propane. Nat Commun 5.

Kler, A.M., Stepanova, E.L., Maksimov, A.S., 2019. Investigating the efficiency of a steam-turbine heating plant with a back-pressure steam turbine and waste-heat recovery. Thermophysics and Aeromechanics 2018 25:6 25(6), 929-938.

Lagioia, G., Spinelli, M.P., Amicarelli, V., 2023. Blue and green hydrogen energy to meet European Union decarbonisation objectives. An overview of perspectives and the current state of affairs. International Journal of Hydrogen Energy 48(4), 1304-1322.

Layritz, L.S., Dolganova, I., Finkbeiner, M., Luderer, G., Penteado, A.T., Ueckerdt, F., Repke, J.U., 2021. The potential of direct steam cracker electrification and carbon capture & utilization via oxidative coupling of methane as decarbonization strategies for ethylene production. Applied Energy 296.

Luderer, G., Vrontisi, Z., Bertram, C., Edelenbosch, O.Y., Pietzcker, R.C., Rogelj, J., De Boer, H.S., Drouet, L., Emmerling, J., Fricko, O., Fujimori, S., Havlík, P., Iyer, G., Keramidas, K., Kitous, A., Pehl, M., Krey, V., Riahi, K., Saveyn, B., Tavoni, M., Van Vuuren, D.P., Kriegler, E., 2018.

Residual fossil CO2 emissions in 1.5–2 °C pathways. Nature Climate Change 2018 8:7 8(7), 626-633.

Ma, Z., Trevisanut, C., Neagoe, C., Boffito, D.C., Jazayeri, S.M., Jagpal, C., Patience, G.S., 2016. A micro-refinery to reduce associated natural gas flaring. Sustain Cities Soc 27, 116-121.

Masson-Delmotte, V., Zhai, P., Pörtner, H.-O., Roberts, D., Skea, J., Shukla, P.R., Pirani, A., Moufouma-Okia, W., Péan, C., Pidcock, R., Connors, S., Matthews, J.B.R., Chen, Y., Zhou, X., Gomis, M.I., Lonnoy, E., Maycock, T., Tignor, M., Waterfield, T., 2019. Global warming of 1.5°C An IPCC Special Report on the impacts of global warming of 1.5°C above pre-industrial levels and related global greenhouse gas emission pathways, in the context of strengthening the global response to the threat of climate change, sustainable development, and efforts to eradicate poverty Edited by Science Officer Science Assistant Graphics Officer Working Group I Technical Support Unit.

Mehmeti, A., Angelis-Dimakis, A., Arampatzis, G., McPhail, S.J., Ulgiati, S., 2018. Life Cycle Assessment and Water Footprint of Hydrogen Production Methods: From Conventional to Emerging Technologies. Environments 5(2), 24.

Mynko, O., Amghizar, I., Brown, D.J., Chen, L., Marin, G.B., de Alvarenga, R.F., Uslu, D.C., Dewulf, J., Van Geem, K.M., 2022. Reducing CO2 emissions of existing ethylene plants: Evaluation of different revamp strategies to reduce global CO2 emission by 100 million tonnes. J Clean Prod 362, 132127-132127.

Nikolaidis, P., Poullikkas, A., 2017. A comparative overview of hydrogen production processes. Renewable and sustainable energy reviews 67, 597-611.

Ohta, H., 2021. Japan's Policy on Net Carbon Neutrality by 2050. East Asian Policy 13(01), 19-32.

Oud, P., 2018. Cracking Furnace System And Method For Cracking Hydrocarbon Feedstock Therein. TECHNIP FRANCE OP - EP 17176502 A 20170616.

Payne, A., Garcia-Garcia, G., Styring, P., 2023. Production of propane and propene via carbon capture utilisation: comparison of its environmental and economic performance against conventional production methods. Green Chem 25(10), 4029-4057.

"Petrochemistry in Europe - Petrochemicals Europe - Petrochemicals Europe". 2022. https://www.petrochemistry.eu/. (Accessed 1.12 2022).

PlasticsEurope, 2017. PlasticsEurope recommendation on Steam Cracker allocation. (September).

Plehiers, P.P., Symoens, S.H., Amghizar, I., Marin, G.B., Stevens, C.V., Van Geem, K.M., 2019. Artificial Intelligence in Steam Cracking Modeling: A Deep Learning Algorithm for Detailed Effluent Prediction. Engineering 5(6), 1027-1040.

Rubini, D., Karefyllidis, N., Xu, L., Rosic, B., Johannesdahl, H., 2022a. A new robust regenerative turbo-reactor concept for clean hydrocarbon cracking. Journal of the Global Power and Propulsion Society 6, 135-150.

Rubini, D., Xu, L., Rosic, B., Johannesdahl, H., 2022b. A New Turbomachine for Clean and Sustainable Hydrocarbon Cracking. Journal of Engineering for Gas Turbines and Power 144(2).

Seppala, J., Hiltunen, J., Purola, V.-M., 2018. Process and rotary machine type reactor.

Simpson, A.P., Lutz, A.E., 2007. Exergy analysis of hydrogen production via steam methane reforming. International Journal of Hydrogen Energy 32(18), 4811-4820.

Styles, D., Dominguez, E.M., Chadwick, D., 2016. Environmental balance of the UK biogas sector: An evaluation by consequential life cycle assessment. Science of The Total Environment 560-561, 241-253.

Szpilko, D., Ejdys, J., 2022. European Green Deal - Research Directions. A Systematic Literature Review. Ekon Srod 81(2), 8-38.

Talah, D., Bentarzi, H., 2022. A General Overview of Combined Cycle Gas Turbine Plants. Algerian Journal of Signals and Systems 7(4), 135-155.

TechnipEnergies, 2021. "An energy transition leader". https://www.technipenergies.com/en. (Accessed 07.27 2022).

Tijani, M.E.H., Zondag, H., Van Delft, Y., 2022. Review of Electric Cracking of Hydrocarbons. ACS Sustainable Chemistry & Engineering 10(49), 16070-16089.

Tullo, A., 2021. PETROCHEMICALS Sabic, BASF, Linde join for electric cracking. AMER CHEMICAL SOC 1155 16TH ST, NW, WASHINGTON, DC 20036 USA.

Tyson, T.R., 1976. Hospitals must develop creative marketing programs or lose business. Modern healthcare. [Short-term care ed.] 6(5), 56-56.

Van Geem, K.M., Marin, G., Grootjans, J., 2008. Energy efficient design of the cold train of a steam cracker.

Van Geem, K.M., Weckhuysen, B.M., 2022. Toward an e-chemistree: Materials for electrification of the chemical industry. MRS Bulletin.

Van Wijnen, J., Ivens, W.P.M.F., Kroeze, C., Löhr, A.J., 2015. Coastal eutrophication in Europe caused by production of energy crops. Science of the Total Environment 511, 101-111.

WBCSD, 2014. Life Cycle Metrics for Chemical Products. pp. 120-120.

Wernet, G., Bauer, C., Steubing, B., Reinhard, J., Moreno-Ruiz, E., Weidema, B., 2016. The ecoinvent database version 3 (part I): overview and methodology. International Journal of Life Cycle Assessment 21(9), 1218-1230.

Wernet, G., C, B., Steubing, B., Reinhard, J., Moreno-Ruiz, E., Weidema, B., 2016. Ecoinvent Version 3. The International Journal of Life Cycle Assessment 21(9), 1218-1230.

Weydahl, T., Jamaluddin, J., Seljeskog, M., Anantharaman, R., 2013. Pursuing the pre-combustion CCS route in oil refineries – The impact on fired heaters. Applied Energy 102, 833-839.

Wiloso, E.I., Heijungs, R., de Snoo, G.R., 2012. LCA of second generation bioethanol: A review and some issues to be resolved for good LCA practice. Renewable and Sustainable Energy Reviews 16(7), 5295-5308.

Yu, M., Wang, K., Vredenburg, H., 2021. Insights into low-carbon hydrogen production methods: Green, blue and aqua hydrogen. International Journal of Hydrogen Energy 46(41), 21261-21273.

Zhao, Z., Jiang, J., Wang, F., 2021. An economic analysis of twenty light olefin production pathways. Journal of Energy Chemistry 56, 193-202.

Zimmermann, H., Walzl, R., 2009. Ethylene, Ullmann's Encyclopedia of Industrial Chemistry. Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim, Germany, pp. 465-526.