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Published in: SN Applied Sciences

DOI: 10.1007/s42452-022-04991-4

Published: 01/04/2022

Document Version Publisher's final version

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Link to publication

Please cite the original version: Reznichenko, A., & Harlin, A. (2022). Next generation of polyolefin plastics: improving sustainability with existing and novel feedstock base. *SN Applied Sciences*, *4*(4), [108]. https://doi.org/10.1007/s42452-022-04991-4



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Next generation of polyolefin plastics: improving sustainability with existing and novel feedstock base

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Received: 21 June 2021 / Accepted: 8 February 2022 Published online: 17 March 2022 © The Author(s) 2022 OPEN

Abstract

In this account, we present an overview of existing and emerging olefin production technologies, comparing them from the standpoint of carbon intensity, efficiency, feedstock type and availability. Olefins are indispensable feedstock for manufacture of polyolefin plastics and other base chemicals. Current methods of olefin production are associated with significant CO₂ emissions and almost entirely rely of fossil feedstock. In order to assess potential alternatives, technical and economic maturity of six principal olefin production routes are compared in this paper. Coal (brown), oil and gas (grey), biomass (green), recycled plastic (pink) as well as carbon capture and storage (purple) and carbon capture and utilization (blue) technologies are considered. We conclude that broader adoption of biomass based "green" feedstock and introduction of recycled plastic based olefins may lead to reduced carbon footprint, however adoption of best available technologies and introduction of electrocracking to existing fossil-based "grey" olefin manufacture process can be the way to achieve highest impact most rapidly. Adoption of Power-to-X approaches to olefins in the long term, however substantial development and additional regulatory incentives are necessary to make the solution economically viable.

Article highlights

- In this account, we introduce a color coding scheme to differentiate and compare carbon intensity and feedstock types for some of the main commercial and emerging olefin production routes.
- Most viable short term improvements in CO₂ emissions of olefin production will be achieved by discouraging

"brown" coal based production and improving efficiency of "grey" oil and gas based processes.

 Gradual incorporation of green and recycled feedstock to existing olefin production assets will allow to achieve substantial improvements in carbon efficiency in longer term.

Key words Olefins · Ethylene · Propylene · Production · Alternative feedstocks · Carbon intensity

1 Introduction

Ethylene and propylene are the two largest volume petrochemicals in the world, with particular relevance to industrial manufacture of polyolefins. Conventionally, the major fraction of ethylene and propylene is produced via the thermal cracking of petroleum and natural gas feedstock [1]. Well-known environmental considerations, new directives and tighter regulations drive the interest in development of new industrial processes with lower carbon

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SN Applied Sciences (2022) 4:108

https://doi.org/10.1007/s42452-022-04991-4

emissions and reduced fossil feeds consumption. In this context, possibility to produce ethylene and propylene from renewable feedstocks and waste including recycled plastics and further with processes applying low-carbon energy is of interest. These developments may lead to a totally new generation of sustainable polyolefin plastics. In this account, we will review and compare the impact of potential scenarios introducing alternatives to the "brown" and "grey" fossil-based ethylene and propylene.

Despite growing attention to the development of biobased polymer solutions [2] as potential replacement to at least a fraction of 70 Mt PP and 110 Mt PE produced annually [3], such replacement is unlikely occur to a significant extent within next several decades. Primary reasons are unmatched cost and performance of polyolefin based resins and slow commercialization of biobased alternatives such as polylactic acid (PLA), thermoplastic starch and polyhydroxyalkanoates (PHA) [4]. It is therefore of paramount importance to consider alternative sustainable feedstocks for the manufacture of ethylene and propylene as primary raw materials for the polyolefin production. We are convinced that the near-to-mid-term "replacements" for the existing polyolefins shall still be polyolefins prepared from non-fossil feedstocks or recycled monomers.

In order to distinguish between different types of industrial hydrogen, a use of different colors to describe the production method and carbon footprint have been introduced in the literature. The most common terms are green, blue and grey hydrogen [5]. Similar nomenclature has been used to differentiate different types of ammonia [6]. To our knowledge, no such systematic "color coding" scheme has been adopted e.g. for ethylene and propylene, with an exception of the term "green ethylene" which has been in use for over 10 years primarily referring to ethylene produced via catalytic dehydration of bioethanol [7]. Herein, we would like to propose the following system to distinguish different types of olefin feedstock, namely



Fig. 1 Six feedstock types for olefin manufacture considered in this review. NG stands for natural gas, CCS for carbon capture and storage

brown, grey, green, pink, blue and purple olefins (Fig. 1). This simple classification can contribute to streamlined product branding, increased consumer awareness and will eventually serve as a structure for future governmental and regulatory practices.

In the next section of this account, we describe basic principles and analyze the state of technical and commercial readiness of six principal olefin production routes, including both commercial and emerging technologies. Overall comparative assessment of the possible impact on carbon intensity of olefin and polyolefin production via various routes and feedstock combinations is provided in concluding section.

2 Commercial technologies

2.1 Grey olefins

Most common, oil or natural gas (NG) based "grey" ethylene is produced mainly by thermal cracking of hydrocarbons in the presence of steam [8]. Ethylene production by the thermal cracking of naphtha is an energy-intensive process (up to 40 GJ heat per ton ethylene), leading to significant formation of coke and nitrogen oxide (NO_x), along with 1.6–2 kg of carbon dioxide (CO₂) process emissions per kilogram of ethylene produced [9].

The olefin industry is one of the largest consumers of primary energy, resulting in a total of 30% of direct CO_2 emitted from chemical plants [10]. The indirect CO_2 emissions attributed to electricity consumption by olefin production processes is roughly 12% of the total CO_2 emissions of chemical and petrochemical plants [11].

Ethylene is the main product of steam cracking, and the process also yields significant amounts of propylene and butylenes, depending on the feedstock [12]. Propylene is primarily produced as a co-product of naphtha cracking. In addition, on-purpose propylene production technologies such as propane dehydrogenation (PDH) and metathesis are used industrially. As cracking plants worldwide are being increasingly converted to ethane as a main feedstock, a demand for on-purpose propylene production is growing since ethane cracking is highly selective towards ethylene [13]. Figure 2 depicts major relevant routes to both grey an brown fossil based olefins.

Energy intensive steps of olefin production via steam cracking are pyrolysis, quenching, compression and separation [9]. Over several decades, olefin production industry has been gradually adopting incremental improvements in overall process efficiency by optimizing feedstock, improving heat recovery and separation efficiency, reducing coke formation, etc. It is estimated that adoption of best available technologies (BAT) may lower energy costs to 12 GJ/t

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ethylene [14]. Current European average energy consumption level is 12.2 GJ/t, whereas the world average is at 13.8 GJ/t, hence it can be estimated that global adoption of BAT across the industry may lead to 585 kt/a CO₂ emission savings [15].

While adoption of BAT can bring a significant shortterm sustainability improvement to the production of grey olefins, introduction of disruptive technologies such as replacing gas fired furnaces with inductive or resistive electric heating can, depending on electricity applied, offer CO₂ emission reduction of up to 90% as reported by BASF corporation [16]. In addition to electrocracking which is currently explored by several industrial players [17], an innovative path to electrifying the cracking process via Rotor Dynamic Reactor as a turbomechanical approach has been recently proposed [18].

Although using electricity produced from renewable sources reduces cracker emissions, a major challenge in developing electrical cracker technology is to ensure that the solution is technologically and economically viable against the existing process. As pointed out in an excellent recent review by Van Geem and co-authors, introduction of advanced manufacturing techniques such as high emissivity coatings, 3D coil set ups and novel furnace designs can bring substantial fuel savings (up to 30%) to the conventional cracker operations without relying on technologies still under basic development (electrocracking) [19].

Growing availability of shale gas has revolutionized the petrochemical production industry in the 2010s and has led to significant added capacity for the olefin crackers, especially in US [20]. It can therefore be expected that expanded cracker fleet will remain in the operation during at least several decades to come. In addition to implementation of BAT to reduce carbon footprint of "grey" olefins, gradual transition towards non-fossil or recycled feedstock such as renewable bio-naphtha, renewable propane or liquefied plastic waste as a feedstock to the crackers can all be a practical path to reduce olefin carbon footprint while utilizing existing asset base. These alternatives will be discussed in the subsequent subchapters.

2.2 Brown olefins

Coal gasification (brown) route remains industrially significant for the production of synthesis gas and olefins. Two primary conversion technologies are most relevant and mature at present:

- "Brown" syngas conversion to methanol, followed by methanol-to-olefins (MTO) process.
- Coal gasification with High Temperature Fischer– Tropsch (HTFT) process.

Methanol-to-olefins (MTO) reaction is among most important C1 conversion routes enabling the production of basic olefins and petrochemicals from coal and natural gas [21]. Many institutions and companies have put great effort to the research and development of MTO reaction since it was pioneered by Mobil Corporation in 1977 [22]. Brown olefins via MTO route represent substantial and growing share of olefin manufacture in China, in particular in coal-rich regions of the country [23, 24]. It must be pointed out that coal-to-olefins (CTO) process is by far more carbon intensive compared to e.g. naphtha cracking, and should ideally be discouraged by implementation of carbon tax or mandatory carbon capture and storage (CCS) technologies [24]. Without such governmental restrictions, brown olefins remain economically feasible and represent up to 40% of olefin capacity in China.

Fischer–Tropsch (FT) process can be used to convert synthesis gas (syngas) directly into olefins, or to produce different products that can be used for the production of olefins (e.g. liquefied petroleum gas and intermediate oxygenated liquids) [25]. The product mixture of the FT process depends on the catalyst, process conditions (pressure and temperature), type of reactor, and the synthesis gas composition [26, 27]. HTFT process using iron catalysts results in highest (\geq 50%) olefin content in FT crude. MTO route holds some advantage over HTFT as the latter results in a complex product mixture containing light and heavy olefins, paraffins, oxygenates and FT-water. Sasol is currently using the HTFT process at their Secunda complex, with an iron-based catalyst, allowing for integrated production of fuels, olefins and chemicals from coal [28].

Despite high carbon intensity, "brown" olefin processes have good potential to serve as a basis for low carbon transformation of olefin value chain. Major syngas conversion technologies of the brown route such as methanol synthesis, MTO and FT process are technically mature and can readily be used for upgrading of syngas produced from non-coal (or non-fossil) feedstock, including gasified biomass (green), CO₂ (blue) and plastic waste (pink). Such possibilities will be discussed in detail in the subsequent subchapters.

2.3 Green olefins

Several routes have been proposed for the production of ethylene from renewable feedstock, but the most promising are: the dehydration of ethanol obtained by fermentation of sugar, starch or cellulose [29];upgrading and cracking of hydrotreated bio oils and the dehydration of methanol obtained from any biomass via gasification to synthesis gas (MTO process) [30]. Principal routes are depicted in Fig. 3.

Bioethanol is mostly used as a biofuel for transport (in pure state or blended in different proportions with conventional fuel), or as an octane enhancing additive in motor vehicles to improve performance [31]. Technoeconomic analysis of ethanol-to-ethylene reveals that this route cannot be presently competitive with grey ethylene produced at world scale steam crackers [32]. However, ethanol dehydration could be of interest for relatively small-scale production in an area where public policy encouraged the development of a chemical industry based on local agricultural sources. As of 2020, 200 kt/a of green PE is produced by Braskem from sugarcane-derived ethanol and is marketed in a variety of low and high density grades [33, 34].

Renewable methanol can be produced from the gasification of biomass to synthesis gas. In theory, any feedstock containing carbon and hydrogen can be used, but because the production of methanol is a cost intensive process, different low-value waste streams are preferred, e.g. black liquor, old wood or biowaste [35]. Technical aspects of biomass gasification, syngas cleaning and integration with heat production has been researched in detail for over several decades. In particular, cleaning and conditioning of syngas to make it suitable for catalytic conversions is among most challenging and cost intensive process steps [36, 37]

Downstream technology to convert methanol to olefins is mature and available to be applied on a commercial scale, as discussed in the previous subchapter [38, 39]. Alternatively, FT route to olefins can be used in combination with biomass gasification [40]. While biomass gasification followed by MTO or FT is a scalable solution, major challenge is reaching economic profitability. Techno economical calculations performed at VTT suggest a need for an extra "green" premium price for the bio-olefin of up to 100 €/t to make the gasification/MTO route economically viable [30]. Biomass gasification for synthesis applications is still under active development aiming to improve process economics by using small to medium scale decentralized gasification/biocrude production with improved thermal efficiency [41]. Simultaneous heat, power, and water integration is essential for the gasification plants to become more favorable economically [42].

Bio oils originating from edible and nonedible biomass can serve as sustainable non-fossil based raw material to produce fuel and chemicals utilizing existing asset base. Ideal, drop-in solution would be a direct feed of bio oils to either steam cracking [43, 44] or a catalytic cracking [45]



Fig. 3 Main conversion pathways in biomass-to-olefins (green) route

SN Applied Sciences A SPRINGER NATURE journal process to produce olefins. However, reports on direct cracking of bio oils are scarce and mostly limited to laboratory scale studies. Major challenge is vastly different composition of bio oils, in particular the presence of varied amount of oxygen. Co-feeding with a petroleum based feedstock can provide a partial solution, however only low amount of high-oxygen content bio-oil is miscible to the hydrocarbon-containing feed, and therefore feeding to fixed-bed systems such as hydrotreater in large volumes is problematic [46].

It is now universally accepted that oxygen containing bio oils need to undergo catalytic hydrodeoxygenation (HDO) in order to become processable at existing refineries and crackers [47, 48]. Liquefaction of lignocellulosic biomass via pyrolysis, catalytic pyrolysis or hydrothermal processes has become a technically mature process and respective bio oils are available at commercial scale. Unfortunately, bio-oils from liquefied lignocellulosic biomass typically feature high oxygen content (20-50 wt.%, depending on the biomass liquefaction process), thus relatively intensive HDO upgrading step is necessary to use existing refinery processes for the further processing [49]. Substantial amount of hydrogen and harsh reaction conditions are needed for the upgrading step, making overall production route more challenging [49]. To date, conversion of lignocellulosic bio oils to olefins and chemicals remains a challenge at industrial scale, mainly due to difficult upgrading step [50].

Vegetable oils, animal fats or triglyceride based biomass residues have relatively low oxygen content (\leq 10 wt. %) compared to lignocellulosic bio oils and thus can be easier processed in a milder hydrotreatment process. Particular examples of bio oils suitable for mild HDO upgrading are palm oil and crude tall oil from coniferous wood pulping [51]. Hydrotreatment of vegetable oils has been performed in commercial scale for production of renewable diesel and jet fuels over the last two decades [52]. Hydrotreatment of triglyceride vegetable oils results in renewable naphtha and propane, which can be subjected to steam cracking and PDH process, respectively. Resulting green ethylene and propylene can then be used for PE and PP production, as has been demonstrated in commercial scale [53, 54].

Advantage of hydrotreated bio oils as a feedstock is the possibility to use existing infrastructure such as cracker and PDH plants for the green olefin production. Vegetable oil production volume is 200 Mt as of 2021 [55], and a hypothetical conversion of an entire olefin industry to such feedstock would cause enormous supply–demand imbalance, likely leading to increased competition with the food value chain and deforestation [56].

Lignocellulosic bio oils from nonedible biomass (fast pyrolysis and catalytic fast pyrolysis oils, hydrothermal

liquefaction oil) are becoming increasingly available but still represent a major challenge from the catalytic upgrading (HDO) standpoint. Additional important consideration is availability of low carbon hydrogen, necessary for the hydroprocessing of bio oils. Thermal decomposition of methane is a suitable technology to provide clean "turquoise" hydrogen when available renewable power is not sufficient to fulfill the "green" hydrogen demand [57]. Methane pyrolysis splits CH₄ directly into its components, i.e., hydrogen and amorphous carbon thereby avoiding direct CO₂ emissions associated with other hydrogen types, such as coal gasification (brown) or steam methane reforming (grey) [58, 59].

A combination of technical challenges, feedstock availability and limited economic profitability suggest that "green" olefins will likely remain a minor part of the olefin portfolio in the forthcoming decades, albeit some growth can be anticipated. Technical feasibility of lignocellulosic oils conversion to olefins and chemicals remains to be demonstrated at industrial scale, while gasification and ethanol routes cannot compete with grey olefins economically. Vegetable oils and fats represent a growing segment with high technical maturity, however existing feedstock base and availability of low carbon hydrogen will need to undergo massive transformation to allow for substantial growth. To date, no "renewable content directive" exists for polyolefins to allow for additional stimulus for the growth of green PO segment.

3 Emerging technologies

3.1 Plastic recycling (pink route)

Plastic pollution and the carbon footprint of plastic value chain are two major societal drivers for increasing the circularity of polyolefins [60]. The full extent of societal and environmental aspects of plastic pollution is outside the scope of this review and the reader can be addressed to some recent publications [61]. Herein, we consider chemical recycling of polyolefins and overall plastic waste in the context of providing alternative feedstock for the olefin manufacture.

The recycling rate for plastics in US was as low as 9% in 2018, according to EPA, while certain products had higher recycling rates, such as PET bottles and jars at 29%, and HDPE natural bottles at 29% [62]. As part of its plastics strategy adopted in 2018, the European Commission has set the goal of at least 50% of all plastic packaging waste to be recycled by 2025, and then 55% by 2030. This is a combination of mechanical and chemical recycling, meaning that later is up to one third of total. It must be

pointed out that mechanical recycling, while not in the focus of this account, is estimated to contribute positively to the carbon footprint reduction of the plastic value chain [63].

Chemical recycling of plastics allows to transform them into low molecular weight products which can then be further upgraded to monomers to produce virgin quality polymer resins. Thermochemical processes based on pyrolysis and gasification are the most mature technologies in the field which are still under active development [64].

At present, advanced thermo-chemical recycling of polyolefins still lacks the optimal process design and control over complex depolymerization kinetics to effectively target tailored products and/or selected chemicals. An onpurpose production of pyrolysis oils, light gases and waxes can, to a certain extent be optimized in cases of pyrolysis technologies [65]. Multiple petrochemical producers are currently establishing alliances with technology providers and investing in pyrolysis technology, to build pilot and demonstration scale (below 100 kt/a) plants [66-68]. Some (presumably, limited volume) commercial product launches have already been announced with mass balance approach used to calculate the recycled plastic content in the final product [69]. In all cases, production of pyrolysis crude which needs to be further processed to yield the monomers is targeted.

Direct, thermochemical conversion of polyolefins to respective monomers could be an attractive solution eliminating the need of upgrading or cracking. However monomer yields reported for pyrolysis and catalytic pyrolysis of polyethylene or polypropylene are typically quite modest [70] with some notable exceptions reported for small-scale low residence time fluidized bed pyrolysis of HDPE [71] and LDPE [72]. Some simulation results indicate that it could be possible to produce up to 49% and 34% wt of ethylene and propylene respectively at 850 °C [73], however such selectivities are yet to be achieved experimentally.

Hydrothermal depolymerisation can be considered a special case of pyrolysis approach. The process is conducted in the presence of water, which acts as a reactant, catalyst and solvent and has been reported for supercritical [74] as well as low pressure [75] configurations. Low char formation and high liquid yields were reported. First commercial-sized plant using hydrothermal technology has been announced to come on stream in 2025 [76]. Available results indicate that production of pink olefins from hydrothermal crude from plastic waste would still require steam cracking to obtain olefins.

As discussed in the previous section, gasification combined with MTO or FT process can be applied for production of green olefins from biomass. The same approach can

SN Applied Sciences A Springer Nature journat be applied to the mixed plastic waste. Gasification converts mixed plastic waste into gases such as CH₄, H₂, CO and CO_2 via high temperature reaction with steam, CO_2 , and/or limited amount of oxygen. Gasification is an energy intensive process as it requires a temperature above 700 °C [77]. The resulting syngas needs to be cleaned and conditioned to remove acid gases and impurities, before being compressed and sent to either methanol of FT synthesis reactor. In some cases the syngas needs to undergo a water-gas shift (WGS) reaction to optimize the H₂/CO ratio for synthesis [78] When plastic waste is co-processed with biomass, the resulting syngas has optimal composition, thus simplifying the system configuration and reducing the projected costs [79]. To our knowledge, no demonstration scale project following gasification-synthesis approach to polyolefin or plastic recycling have been launched yet.

In summary, plastic recycling (pink) path to olefins is under dynamic and rapid development at the moment, with pyrolysis technologies in the most advanced stage. Strong interest and commitment from the industrial players along with growing regulatory pressure will ultimately result in broader adoption of technology. At the same time, further development of thermochemical technologies towards improved yields and selectivities and lower process costs is still necessary. Available estimations indicate that pink olefins have generally higher production costs compared to grey olefins, however the difference is not prohibitively high [80]. Pre-treatment of complex post-consumer plastic waste streams containing various polymers types, halogens and metal residues still represents a number of technical challenges to be solved before deploying at full industrial scale [64]. Pyrolysis and gasification of complex mixed feeds may also result in formation of polyaromatic hydrocarbons, nitrogen and sulfur oxides, HCN and dioxins, among many other toxic compounds of high concern [64].

Impact of plastic recycling on carbon intensity of olefin manufacture is discussed in the final subchapter.

3.2 Role of CCS (Carbon capture and storage) (purple) and P2X (blue) technologies.

CCS means carbon capture and storage, the process by which carbon dioxide is captured from a point source and stored on site or often off site – for example, at a depleted gas or oil field, or other geological formation. Sequestrated carbon dioxide is expected to stay underground without significant release to the atmosphere. CCS is considered a viable option in many scenarios regarding the petrochemical industry [81]. It is therefore of interest to consider the impact of CCS on manufacture of either "brown" or "grey" olefins. Analysis of Chinese coal-to olefins processes showed that emission reduction up 30% can be expected with implementation of CCS [24]. However there is presently no centralized plan for implementation of CCS in Chinese olefin industry, and the current CCS capacity available in the country is about 2 Mt CO₂ per annum [82]. Global capacity of CCS as of 2021 is estimated to be 40 Mt by the IEA [83]. CCS deployment has been relatively slow to gain traction as multiple projects around the world have been hit by technical issues and cost overruns. Accelerated deployment and scale-up of CCS technologies would likely require creation of investment incentives, CO₂ transportation and storage infrastructure and clarification of regulatory and policing issues [84]. Before such policies and

incentives are created, no substantial impact of CCS on olefin production can be expected.

In contrast to CCS, Carbon Capture and Utilization (CCU) technologies create possibilities to re-use carbon emitted as carbon dioxide and re-introduce it into carbon value chains. The concept of carbon reuse economy, utilizing CCU integrated with direct air capture (DAC) and bioenergy (BE) technologies is the approach leading to carbon-neutral or carbon-negative fuels and chemicals, including olefins [85].

Renewable electricity used for generation of low carbon hydrogen is a key ingredient in transforming CO₂



Fig. 4 Main conversion paths for waste plastic based pink olefins



Fig. 5 P2X chemical paths to chemicals, and energy carriers including "blue" olefins. Scheme from ref 85

SN Applied Sciences A Springer Nature journal to chemicals via Power-to-X approach (Fig. 5). Most of the processes depicted in Fig. 4 are developed to a high Technology Readiness Level (TRL), however commercialisation has been very slow due to the lack of attractive business cases. Growing availability in renewable electric power as well as advances in electrolysis technology (including high temperature co-electrolysis of CO₂ and water) may speed up the development. Carbon Recycling International has been operating demonstration Power-to-X methanol plant in Iceland (4kt/a) since 2011, transforming CO₂ emissions from geothermal power plant to renewable methanol [86]. Larger scale renewable methanol plant is planned for construction in China [87] while several European projects on commercial scale methanol production are pending [88, 89]. None of the renewable methanol projects explicitly mentions olefins as a planned downstream product, however blue olefins can certainly be produced from renewable methanol as MTO technology is readily available.

Alternatively, HTFT route from CO_2 and renewable H_2 can be taken to produce light olefins, as proposed and tested in development project BECCU, currently run by VTT and partners [90].

Several biotechnical conversion routes from CO₂/CO and renewable H₂ were developed and brought to commercial scale to complement chemical paths depicted in Fig. 5. Fermentation processes are advantageous over FT/methanol conversion routes due to low sensitivity to impurities and syngas composition, thus deep syngas cleaning and H₂/CO ratio adjustment is not necessary [91]. Enzymatic conversions are run at ambient or close to ambient conditions, thus high pressure/high temperature reactors are not needed. However the disadvantage of fermentation processes is a low volumetric efficiency, thus extra attention needs to paid to reactor design [92]. Typical products of syngas fermentation are ethanol/ acetic acid or longer chain alcohols such as butanol and hexanol, depending on the type of enzymes used [91]. Recent project by LanzaTech, Total and I'Oreal was a first commercial scale PE packaging solution with carbon originating from industrial CO₂. CO₂ was converted to ethanol with Lanza's proprietary fermentation platform, followed by ethanol dehydration to yield polymerisation grade ethylene [93].

Economic considerations are crucial when considering potential scalability of the technology. Cost analysis calculations performed by VTT in 2017 showed that power-to-olefins approach via MTO is characterized by higher production costs of up to 2000 Eur/t for blue olefins which is roughly twice the price of fossil-based light olefins. To satisfy the global demand (ca 200 Mt/a) of light olefins through Power-to-X route, 644 GW of renewable electric power and 924 Mt/a CO₂ (3% of annual

SN Applied Sciences A SPRINGER NATURE journal global emissions) would be required [94, 95]. As of 2021, in most cases the production of green hydrogen, e-fuels and e-chemicals (such as methanol) still cannot directly compete with fossil based production routes due to high capital and operational costs [96].

In summary, despite the exciting emission-saving potential of Power-to-X concepts, broader adoption would require additional economic incentives (additional carbon tax, government grants for capital expenditures) as well as further deployments and technical developments in renewable energy and hydrogen production and process integration.

4 Comparison of alternative solutions

Carbon emissions are directly related to the conversion process efficiency and secondary to means of carbon capture storage or utilization. Fossil carbon content can be reduced by means of utilization of bio feedstoc and applying recycling. The latter does not influence fossil carbon content in the product as such, but does reduce the need of fossil feedstock to supply the needed amount of PO product. General concepts to minimize carbon emissions and fossil carbon contents in olefins are presented in Fig. 6. Bio replacement path (green) is based on all-renewable carbon. Carbon emission free energy and hydrogen (blue) is result of electrification of chemical process. Carbon loss minimization (grey) reflects to recycling of plastic waste and more efficient operation of cracker such applying BAT. Finally carbon emission capture (purple) processes can be applied to either of routes for olefin manufacture, allowing to lower final carbon footprint.



Fig. 6 Conceptual paths to minimum carbon emissions and fossil carbon contents in olefins: bio replacement (green), carbon emission free energy (blue), carbon loss minimization (grey) and carbon emission capture (purple)

Carbon dioxide emissions for different routes of olefin production have been reported earlier in several studies and compilations [1, 97, 98]. We have selected several important cases of olefin production, representing all six types of feedstock, are compared carbon dioxide emissions associated with each case (Table 1). Single illustrative values for the CO₂ emissions as well as energy demand are given to provide a comparative overview. The reader is addressed to the referenced studies for the details and methodology of LCA analyses which may differ from one study to another. For table entries, first column is the fossil carbon intensity of raw material only, energy describes the energy intensity of olefin conversion process, and "total" is sum of all fossil-based CO₂ emissions at gate as the authors have performed own assessment calculations.

Traditional naphtha steam cracker in based on highenergy consumption, especially in pyrolysis process. The process has been improved continuously over the decades and best available technology (BAT) is 7 to 12% more efficient than that of contemporary applied technology in average. Main contributor in conversion is feedstock, where gas condensates are some 10% units more efficient. "Brown" coal-to olefins route is clearly most emissionintensive, independent of the primary conversion technology. The use of this route should ideally be discouraged by additional carbon taxes or mandatory CCS implementation. Implementation of CCS can substantially improve the situation, based on simulation data [24].

Several comparisons show that bio based feed stock can be markedly high in carbon dioxide emission, even if the emissions are dominantly biogenic [100]. Bio-based materials emissions are related to fertilizers, transportation, and various energy demanding process steps. When considering total carbon emission profile of sugarcane derived ethylene via ethanol dehydrogenation, cradle-to gate values reported in the literature vary significantly. Provided net emission values range from highly carbon negative [99] to slightly carbon negative [100] to net positive [101] and highly net positive [101], as different authors took e.g. fertilizer production and energy supply for alcohol dehydrogenation into account very differently. When most realistic scenario of bioethanol-to-ethylene is considered, some 35% of biogenic carbon is present in the overall emissions, and total carbon emissions are 45% lower than traditional grey naphtha-based steam cracking process.

Hydrodeoxygenated (HDO) bio-oils are possible feedstock for steam cracker, where the main additional factor affecting carbon emissions is the source of hydrogen used for oxygen removal. The hydrogen can come from the cracker itself, and could thus be biogenic. More efficient alternatives from the energy balance standpoint are methane pyrolysis with carbon free energy, but similarly also carbon-free electric cracker provides low carbon emission hydrogen. In general, one can conclude that commercially viable "green" polyolefins are 35–45% less carbon intensive compared to "grey" benchmarks as evident from the examples in Table 1. MTO from cellulose pulping side streams black liquor is somewhat more attractive that ETE, with carbon emissions 55% lower than traditional SC naphtha process.

Detailed LCA for the scenario of plastic pyrolysis applied to sorted recycled plastic followed by steam cracking has been reported by BASF corporation [102]. Pyrolysis and steam cracking have major contribution to the process emissions, resulting in 3000 kg CO₂/t ethylene which is higher compared to the production of fossil-based ethylene. A "differential credit" approach is taken to consider also incineration of plastic waste as a base scenario, which would account for an additional 6000 kg CO₂/t ethylene emission as a part of plastic lifecycle. In this regard, chemical recycling is producing ca 50% lower emission Vs virgin "grey" olefins production via naphtha steam cracking. Presently available technologies for both pyrolysis and steam cracking were considered in the study. One can envision that further emission reduction would be possible with e.g. electrification of steam cracking and pyrolysis, as discussed in the previous sections.

In gasification/ Fischer-Tropsch (FT) process there is a possibility to apply mixed hydrocarbon raw material, including plastic waste, to produce either naphtha or olefins directly. Later alternative is essentially more efficient, than route where first paraffins are formed from synthetic gas via LTFT and are subjected to steam cracking to produce olefins. Most efficient is a direct HTFT to olefins, when feedstock is paraffin-like polyolefin plastics. Bio feedstock, rich with oxygen, is less efficient and leads to pronounced carbon dioxide emissions. Later can be controlled by means of carbon capture followed by storage or utilization. This seems to reduce carbon emission by some 15%. Utilization of plastic waste in FT provides similar benefits as ethanol to olefin process and with carbon capture 25% better than ETE, which is 60% better than traditional naphtha cracking.

Power-to-X approaches have highest potential to reach carbon neutrality. Dechema report estimates emissions of CO_2 -based methanol via direct air capture to be of ca -0.67 t CO_2 / t Methanol [14]. Process-related CO_2 emissions of a standard MTO process are estimated at 0.5 t CO_2 /t ole-fin. Since about 2.8 t of methanol would be required to produce 1 t of olefins, total net cradle-to-gate emissions of ca -1.5 t CO_2 /t olefin would be associated to production of blue olefins. More comprehensive cradle-to-grave LCA analysis by Rosental et al. [103] considers end of life of the olefins and indicates slightly positive net carbon emissions, which is still 88–95% lower than that of a fossil-based feedstock [103]. Such improvement comes

Table 1 Comparison of carbon intensity of alternative paths for olefin production

Route	Feedstock	Feedstock O/C	Primary conversion process	Comments	Fossil CO ₂ , kg/t	Energy con- sumed CO ₂ / kg/t	Processing total CO ₂ / Kg/t	Total CO ₂ /t	Ref
Brown	Coal	0.001	HTFT		6100	1760	2200	8300	[104]
Brown	Coal	0.001	MTO		6800	1500	1875	8650	[105]
Grey	Naphtha	0.0001	Steam crack- ing		2900	1063	1635	4535	[<mark>9</mark>]
Grey	Naphtha	0.0001	Steam crack- ing, BAT		2900	840	1063	3863	[<mark>9</mark>]
Grey	Condensate	0.0005	Steam crack- ing, BAT		2800	818	1062	3392	-
Grey	Naphtha	0.0001	Electro crack- ing	Renewable electricity	2900	200	328	3228	-
Green	Ethanol-Corn	0.57	Dehydration	Fermentation	158	1625	1816	2680	[106]
Green	Ethanol-Sug- arcane	0.57	Dehydration	Fermentation	6	1472	1508	2528	[101]
Green	Palm oil	0.08	Stream crack- ing	Grey H ₂ for HDO	2120	1027	1533	3653	[107]
Green	Tall oil	0.06	Steam crack- ing	Turquoise H ₂	740	991	1436	2142	[108]
Green	Tall oil	0.06	E-cracking	Turquoise H ₂	740	200	328	1068	[103]
Green	Black liquor	0.16	MTO	Gasification/ methanol	540	1300	1625	2165	
Purple	Coal CCS	0.001	MTO		800	1700	2125	2925	[<mark>24</mark>]
Pink	Plastic waste		HTFT	Gasification	360	1760	2200	2560	-
Pink	Plastic waste		steam crack- ing	Pyrolysis	-	_	-	3013ª	[108]
Blue- P2X	CO ₂ , DAC	2.67	MTO	DAC, HTCOE	- 2000		500	- 1500	[14]
Brown	Coal	0.001	HTFT		6100	1760	2200	8300	[104]
Brown	Coal	0.001	MTO		6800	1500	1875	8650	[105]
Grey	Naphtha	0.0001	Steam crack- ing		2900	1063	1635	4535	[<mark>9</mark>]
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Pink	Plastic waste		HTFT	Gasification	360	1760	2200	2560	-
Pink	Plastic waste		steam crack- ing	Pyrolysis	-	_	-	3013 ^a	[108]
Blue- P2X	CO ₂ , DAC	2.67	MTO	DAC, HTCOE	- 2000		500	– 1500	[14]

a Total processing and energy impacts, derived from value 3348 kg/t PE reported in ref 108 minus 10% attributed to polymerization process

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Table 2								
Color	Raw material	Primary conversion	TRL ^a	Production costs ^b	CO ₂ emissions	Opportunities and develop- ment needs	Outlook	References
Grey	Naphtha, NG	Steam cracking, PDH	6	Benchmark	Benchmark	Mid term: improving cracker efficiency, partial incorpora- tion of green and pink feed- stock Long term: E-cracking	Major path for olefin produc- tion for next 10–20 years, with gradually growing recycled and renewable feedstock fractions	I
Brown	Coal	HFT, MTO	6	+-20% ^c	+ 100–200%	Incorporation of CCS/CCU. Most preferably discouraged with carbon tax	Highly dependent on national regulatory policies	[109]
Green	Vegetable oils, fats	HDO, cracking	8	+ 20-40%	- 30%	Growing availability of bio- naphtha	Moderate growth, limited by feedstock availability	[110]
	Bioethanol	ETE	8	+30-50%	- 40%	Availability of G2 bioethanol	Limited growth due to unfa- vorable process economics	[32]
	Lignocellulosic residues	Gasification with HTFT/MTOor liquefaction/HDO then cracking	4-6	+ 20-40%	- 30–50%	Major technical challenges still to be solved, robust HDO upgrading of lignocellulosic bio oils to be developed, Economically viable gasifica- tion/synthesis concept to be developed	First-of-a kind demo plant within next 10 years	[95]
Pink	Plastic waste	Pyrolysis and steam cracking	7-8	+10-30%	- 30%	Robust pre- and post-treat- ment options for mixed and contaminated plastic waste streams. Improved process yield and selectivity	Continuous growth in next decades, pink (poly)olefins may reach 5–10% of PO market	[80, 107]
	Plastic waste	Gasification and FTO/HTFT	5-6	+ 20-40%	- 40%	Technology maturity and economic viability need to be improved	First demo plants within next 10 years	[111]
Purple	Coal	Gasification/MTO + CCS	~	+10-40%	- 20%	Relatively mature technol- ogy, would require systemic infrastructural changes to enable capture transport and storage	Limited growth only in case of major policy changes	[24]
Blue	CO_2 point sources	(co)Electrolysis, RWGS, HTFT or MTO	4-6	+ 100%	- 90%	Technology maturity needs improvement, price of low carbon electricity and renew- able hydrogen will be major economic factor	Several industrial demo cases at TRL 6–7 in the next dec- ade, no commercial volumes	[94, 95, 105]
aTechno	ology readiness level, valu	e given for the process step(s) of	lowest	: technical maturity.	: ^b cost estimate r	anges based on available literatu	ıre references ^c highly dependen	t on oil price,

see reference 111.

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at a price of an enormous need in renewable energy, as energy needed for production of "blue" olefins is ca twice the energy needed for the "grey" benchmark [108].

Comparative overview of six principal paths for olefin production is presented in Table 2. While there are seemingly multiple options to produce olefins, the most technically mature fossil-based "grey" olefins and polyolefins will likely continue to dominate the markets in foreseeable future. Pink olefins are clearly on their way to transition from "emerging" to "commercial" status.

5 Conclusions

While multiple options to reduce carbon footprint of olefin production exist in the form of relatively mature technologies, we expect that traditional, "grey" fossil based olefins will dominate the markets for next 2-3 decades which is in line with other projections in the literature [1]. Introduction of BAT in naphtha cracking and utilization of condensates may reduce carbon emissions by 20% compared with standard olefin production. Transition to green feedstocks such as ethanol to ethylene route will bring substantial benefit in CO₂ emission reduction of up to 45%, however broader implementation will likely be hampered by economical disadvantages Vs conventional "grey" route. The share of "green" olefins will grow over next decades, however major technical breakthroughs in biomass gasification or lignocellulosic bio oils upgrading will be necessary, as availability of vegetable oil feedstock is limited. Plastic waste recycling does not markedly reduce carbon emissions, but could reduce fossil feedstock utilization for 25 to 55%. We expect that "pink" olefins produced from plastic waste will experience a strong growth over next decade as manifested by strong commitment from the industry. Ultimate emission reduction by 80-90% can be achieved via adoption of Power-to-X technologies which at present are not economically competitive with fossil based routes without additional carbon taxes and governmental incentives. Olefins and polyolefins are going to remain carbonpositive material in the next 3-4 decades. However PO's contribution to lowering emissions remains significant due to indispensable functions such as energy transport and storage, food chain security and low carbon mobility. This solid net negative effect fully justifies and will further strengthen polyolefins' role in future sustainable society.

Funding This work was partially funded by Academy of Finland Plastic Hero project.

Declarations

Conflict of interest There are no conflicts of interest to declare.

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