

Evaluation and Comparison of the Conventional and Renewable-based Polyolefin Production based on Greenhouse Gas Reduction

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Introduction

The Paris Agreement has the ambition to reduce greenhouse gas (GHG) emissions to achieve a maximum global temperature increase of 2 °C above pre-industrial levels [1]. To render this goal possible, the greenhouse gas emissions of the different sectors need to be greatly reduced. In Austria, the main contributors can be divided into energy and industry (37.0 % ETS (European Trading System), 7.4 % non-ETS) and transport sector (27.8 %), which accounted for about three quarters of the total Austrian greenhouse gas emissions in 2021. The remainder consists of other sectors such as buildings (11.7 %), agriculture (10.6 %), waste management (3.0 %) and fluorinated gases (2.4 wt.-%) [3]. As the electrification of transport vehicles (cars, trucks) is only one way to decarbonize the transport sector, research on renewable fuels, so-called e-fuels, is booming in Europe. Nevertheless, proposals for the other sectors, especially for the energy and industry sector, need to be identified and evaluated for decarbonization [4].

In this work, the focus is therefore on the reduction of greenhouse gases in the chemical industry, especially for the production of polyolefins. In 2020, Borealis AG produced around 1 million tons of polyolefins at its Schwechat site in Austria [5]. The production of polyolefins is based on the polymerization of olefins. There are several methods to produce the feedstock olefin. On the one hand, this is possible with fossil feedstocks such as naphtha, which is a product of fossil crude oil distillation [6]. Another approach is the use of biomass or power-to-X (PtX) processes for climate-neutral production of olefins. In this work, the assumption is made that the industry and transport sectors cannot be completely decarbonized from biogenic feedstock, as the amount of biomass required would be far greater than the amount that can be produced. The production of 1 million ton polyolefins per year requires 12.1 million tons of biomass, which accounts for about 60 wt.-% of Austria's forest biomass potential [7,8].

Therefore, Power-to-X (PtX) technologies are considered in this work. In a first step, in order to come closer to climate neutrality, carbon sources, suitable for further processing to polyolefins must be found and defined. The capture effort from gas streams decreases with increasing CO₂ concentration. Direct air capture (DAC) would therefore be 2.9 times more energy-intensive (in combination with high electricity prices economical infeasible) than concentrated sources (e.g. exhaust gas streams). Therefore, DAC is not considered in this work [9]. Point sources, especially from process-related CO₂ emissions, represent a favorable application area for such PtX plants.

In Austria, the cement industry emitted 9.0 % of ETS certificates and 3.3 % of the total national greenhouse gases in 2019. In the cement production process, two thirds of CO₂ is emitted due to the calcination of the limestone in the pre-heater and rotary kiln and is accounted as process related CO₂. The remaining third is emitted by the heat supply for calcination, especially the firing of conventional

fuels (e.g., coke) and substitute fuels (e.g., waste oil, residual waste, such as plastics, which are not mechanically or chemically able to be recycled) [10].

Carbon Capture and Utilization or Storage "CCU/S" is an indispensable option to decarbonize the cement industry sector. The "Association of Austria's Cement Industry" stated in their report [11], that CO₂ reduction can be achieved by various factors such as clinker reduction, savings in transportation and the use of green electricity. But the remaining 44 % of the CO₂ emissions must be reduced by CCU/S [11]. In Austria, the "C2PAT – Carbon to Product Austria" project is a flagship project, where CO₂ should be utilized for the production of renewable based polyolefins. A consortium of four industrial partners, Lafarge Zementwerke GmbH, Verbund AG, OMV AG and Borealis AG are working together with the scientific partners Montanuniversität Leoben and Wirtschaftsuniversität Wien to gain experience in the implementation of such a Carbon Capture and Utilization unit, which should be realized in the cement plant in Mannersdorf am Leithagebirge. In order to analyze the impact of a large-scale plant on the reduction of fossil raw materials and savings of CO₂, the 700,000 tons of CO₂ emitted annually by the cement plant is considered.

At present, fossil raw materials are required for the production of polyolefins. Additionally, CO₂ emissions are also generated during the production of polyolefins. On the other hand, in the cement industry, process related CO₂ emissions arise, as long as cement is produced. By combining these two industry sectors, the emitted CO₂ from the cement plant could be used for the production of renewable plastics. This would have two effects: 1. the reduction of the amount of fossil crude oil used for naphtha and in the broadest sense a reduction of fossil olefin production and CO₂ emission in its production chain, and 2. the use of "anyway" emitted CO₂ from the cement industry, i.e. the reuse of CO₂ and, if necessary, storage in longer-lasting plastic products. It should be mentioned that the use in plastic production, e.g. for high-voltage cable sheathing, would have a further advantage of long-term CO₂ storage (~50-100 years).

Consequently, this work aims to provide an overview of the production of polyolefins from conventional fossil raw materials and the use of process-related CO₂ from the cement industry. The PtX pathway from CO₂ and hydrogen via the intermediate Fischer-Tropsch syncrude to polyolefins (mainly polypropylene – PP, or polyethylene – PE) is described in this work. A post-processing unit such as a steam cracker will also be considered in the process comparison. A mass balance of these process routes is established in order to calculate the GHG reduction potential in kgCO₂/tpolyolefin. Furthermore, conclusions on the used feedstock are made and discussed.

Methodology

The work is based on a calculation of the mass balance of the conventional fossil production of polyolefins using literature data. For comparison, a simulation of a PtX plant producing polyolefins based on 700,000 tons of CO₂ from the cement plant off-gas will be created [12]. The yearly operation time of the cement plant amounts about 7,880 h. As a first step, the ASPEN Plus based simulation will be performed, and the mass flows will be analyzed. The contained product amount of polyolefins will then be used as a basis for back-calculation to fossil naphtha and crude oil to determine the fossil feedstock reduction potential.

Figure 1 defines the two process routes for polyolefins production. Process route 1 describes the conventional production process with the feedstock fossil crude oil. In a first step, the crude oil is separated into different fractions, with naphtha being the feedstock for the refinery's olefins production. Depending on the composition of the crude oil, a yield of 2 – 14 vol.-% is obtained from crude oil for light naphtha (5 – 90 °C) and 8 – 26 vol.-% for heavy naphtha (distillation range 90 – 180°C) [13]. Naphtha is converted into the main products ethylene and propylene and several by-products in the steam cracker. For 1 ton of ethylene and 0.53 tons of propylene, 3.25 tons of naphtha are needed [14]. The by-products are not considered further here. The conversion of the olefins into polyolefins is done almost completely in Borealis' own Borstar plant. Today, the distillation and steam cracker are still operated with fossil fuels, which means that CO₂ emissions are already released during the production of naphtha. It is assumed that around 1.91 kgCO₂/kg of product is emitted for polypropylene and 1.98 kgCO₂/kg of product for polyethylene [6,15].

In the PtX routes defined in process route 2, CO₂ is captured from the cement plant off-gas. Hydrogen is produced via an electrolysis and converted with CO₂ in a reverse water-gas-shift (rWGS) reactor to synthesis gas, which is the feed for the downstream Fischer Tropsch synthesis (FTS). The FTS product, also called syncrude, is separated in flash units to obtain the product fractions naphtha, middle distillate and waxes. These products are further converted in the steam cracker to olefins and in the polymerization unit to polyolefins. The simulation includes the carbon capture unit, electrolysis, rWGS and FT reactor including the product separation unit of the PtX route. The simulation is carried out in ASPEN Plus and described below. A detailed flow sheet is shown in Figure 3.

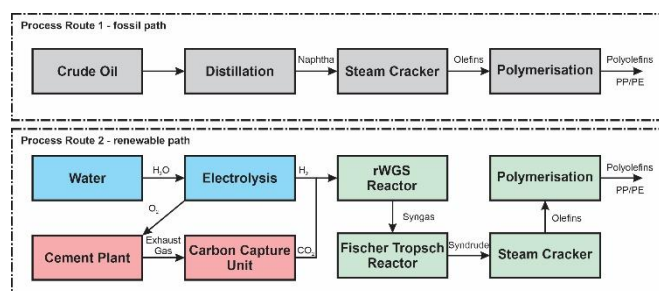


Figure 1: Process Route description (1 - fossil path; 2 - renewable path via rWGS and FTS)

Carbon capture unit

The carbon capture unit is realized with an amine scrubber, since this process has a generally high technology readiness level (TRL). Other sustainable capture technologies such as PSA and cryogenic capture will also be considered in the further project design phase. In order to get the exhaust gas free of particles and pollutants (e.g., sulfur) and at operating temperature for the absorber, it is subjected to a pre-cleaning unit [2]. The pre-cleaned gas is fed to the absorber tower in counterflow to the monoethanolamin (MEA) solution in the lower section. CO₂ dissolves in the solution at ambient pressure, while the other gases leave the absorber at the top and are released

into the environment. The loaded amine mixture is heated and fed to the desorber tower. Reboiler heat of about 3.8 MJ/kgCO₂ is supplied, releasing the CO₂ from the solution, which is stripped from the head of the stripper with water vapor, cooled, and separated in a flash to achieve concentrated CO₂ and water. The CO₂ stream has a purity of about 95 wt.-% (wet), or 99.8 wt.-% (dry) [2].

Hydrogen production

Hydrogen must be provided for the catalytic conversion of CO₂ to hydrocarbons via the Fischer Tropsch synthesis. Using water electrolysis technologies, this can be produced by splitting water with the supply of electricity according to endothermic reaction 1.



It is important to note that decarbonization only makes sense if the electricity used comes 100 % from renewable sources. Otherwise, if electricity is produced by fossil power plants, e.g. coal or natural gas, CO₂ would already be emitted by the electricity supply for hydrogen production. Although not at the production site, this emission must also be included in the overall system analysis [2]. High-temperature electrolysis is used in the simulation because excess heat from the FTS can be used to evaporate feed water from the electrolysis, which improves the overall efficiency. It is assumed that the electrolysis is operated at temperatures between 700 and 900 °C and ambient pressure with a specific system efficiency of 3.6 kWh/Nm³H₂ [16]. The oxygen flow is directly linked to the cement plant, if the electrolysis is located there. For an electrolysis comparison, the electricity demand for a low-temperature PEM electrolysis (5 kWh/Nm³H₂) is also considered [16].

Reverse water gas shift reaction

The rWGS reaction is the conversion of CO₂ and hydrogen to syngas. According to the thermodynamic equilibrium (Figure 2), high temperatures (950 °C) and low pressures (10 barg) favor the endothermic reaction (reaction 2) and conversion.



Furthermore, these operating conditions reduce the production of by-products, e.g. methane and solid carbon. Moreover, it is necessary to adjust the hydrogen flow to guarantee a H₂:CO ratio at the inlet of the FT reactor (2.08:1). In addition to the high operating temperature, the over stoichiometric operation suppresses the formation of carbon [2]. It is important to mention that short-chain hydrocarbons (C₁ – C₄) are recycled back to the rWGS reactor through the recycling stream from the FTS, which is explained in the FTS section. Steam and dry reforming take place, whereby methane in particular is converted with H₂O to synthesis gas.

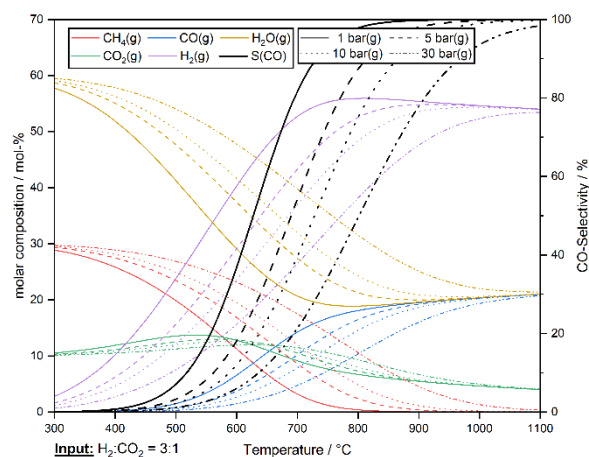


Figure 2: Thermodynamic equilibrium of the rWGS reaction for an operation temperature range from 300 to 1,100 °C and pressures of 1, 5, 10 and 30 barg

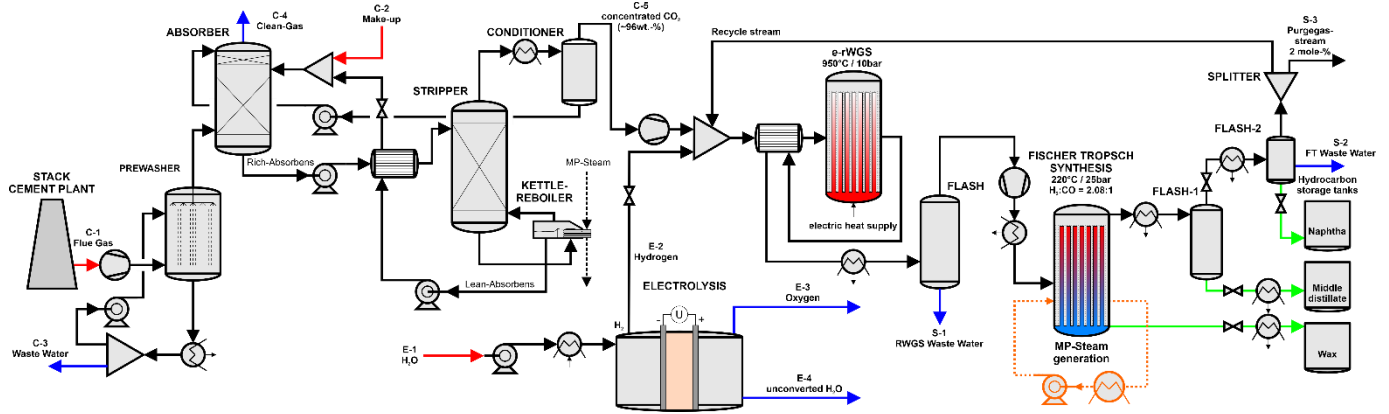


Figure 3: Flow sheet of the simulated PtX process including carbon capture, electrolysis, rWGS and FT reactor with product separation unit [2]

Fischer Tropsch synthesis

It can be differentiated between low and high temperature FTS, whereas the main distinction is the product formation. The product of the low temperature FTS consists mainly of normal paraffinic hydrocarbons, compared to low chain olefins in the high temperature FTS [17]. Therefore, low temperature FTS (220 °C and 25 barg) with a cobalt catalyst is used, which has the advantage that the product is similar to fossil crude oil. In the simulation a stoichiometric reactor is implemented, whereas reaction 3 describes the conversion of CO₂ and hydrogen to hydrocarbons, with a single conversion rate of 40 % [2].



The production of the hydrocarbons is described by the Anderson-Schulz-Flory (ASF) distribution, which assumes a chain growth probability of 0.92 [18]. Accordingly, the required input ratio of H₂:CO is calculated to be 2.08:1 [19]. It must be added here that the distribution has been modified because in reality there is a higher methane content in the product than the ASF distribution specifies [20]. Accordingly, in addition to the main reaction (reaction 3), the methanation (reaction 4) with a methane selectivity in the FT product of 16 % is also considered in the simulation [2].



Both reactions are strongly exothermic, which requires an appropriate cooling system for the FT reactor to avoid "runaway". This dissipated heat is taken into account in the heat integration to obtain a holistically high efficiency of the entire process setup.

The wax produced (>C₂₂) is liquid and is drawn off directly from the reactor. The gas phase is separated in a two-stage flash separation

into the fractions middle distillate (C₁₁ – C₂₂) and naphtha (C₅ – C₁₀), and the by-product water. These fractions are then transported to the Schwechat refinery for further use. The remaining gaseous stream, consisting of predominantly C₁ – C₄ hydrocarbons, is recycled upstream the rWGS reactor to achieve the highest possible conversion rate to liquid products. In order to avoid accumulation of inert gases (e.g. nitrogen), 2 vol.-% of the recycle gas is purged.

Steam cracker and polymerization unit

In contrast to the fossil operation of a steam cracker, studies have been conducted for FTS syncrude from CO₂ and H₂, with the result that the complete syncrude (naphtha, middle distillate and wax) can be processed. The conversion parameters for the different fractions are taken from Karaba et al. [21]. The downstream polymerization efficiency is assumed with 99 %. The prerequisite for a green process would be again, that the energy required in the steam cracker and polymerization unit must be provided from renewable sources (i.e. green electricity).

Results

Using the previously defined process routes, the mass balance of the fossil and renewable pathways can be established. Table 1 shows the mass balance based on the 700,000 tons of CO₂ emitted annually by the cement plant. It is clearly evident that approx. 90 wt.-% of the CO₂ fed from the cement plant is separated in the amine wash and converted into syncrude in the synthesis process. It is important to mention that CO₂ is contained in the clean gas of the amine scrubbing, which is released to the environment. This CO₂ is emitted, but possible improvements and developments of the separation technology could further increase the reduction of CO₂ released to atmosphere.

Table 1: Mass balance of the simulated PtX process route

Mass balance	Inlet			Outlet								
	Flue gas	Makeup amine	Water electrolysis	Prewasher	Clean gas	Water treatment plant	Unconverted water	Oxygen	rWGS water	Purgegas	Syncrude	
Total	[t/h]	427.1	50.1	222.8	15.5	378.6	111.4	11.1	89.0	24.0	3.1	24.5
CO	[t/h]			0.0							2.0	
H ₂	[t/h]			0.0							0.3	
H ₂ O	[t/h]	32.2	50.1	222.8	15.5	64.2	111.4	11.1	0.0	24.0		0.1
CO ₂	[t/h]	88.8		0.0		8.3					0.4	
N ₂	[t/h]	258.7		0.0		258.6					0.1	
O ₂	[t/h]	47.4		222.8		47.4			89.0			
MEA	[t/h]		0.002									
C ₁₊	[t/h]										0.4	24.4

Considering the water consumption of the electrolysis, which is a result of the simulation, this plant requires about 11.4 t/h or 127,440 Nm³/h of hydrogen, which corresponds to an electrolysis power of about 460 MW (SOEC, 3.6 kWh/Nm³H₂) or 637 MW (PEM, 5 kWh/Nm³H₂). The implementation of this plant size at the cement plant site in Mannersdorf am Leithagebirge is currently impossible due to the lack of grid capacities.

Table 2 shows the mass balance of the steam cracker and the polymerization unit. Thus, from the 700,000 tons of CO₂ per year, 120,560 tons of polyolefins (sum of polyethylene and polypropylene) can be produced annually. By-products (e.g., Butene, Butadiene, ...) generated in the steam cracker and in the polymerization unit are listed as "rest" [21].

Initially, it can be stated that in the PtX process (process route 2) CO₂ is only released in the amine wash. The energies required for electrolysis, synthesis, steam cracker and polymerization unit are provided with green electricity, which means that no CO₂ is produced by electricity supply. [22].

Table 3: Mass balance of the steam cracker and the polymerization unit

Inlet – Syncrude			Outlet – Olefins			Polymerization		
Total	[t/h]	24.3	Total	[t/h]	24.3	Total	[t/h]	15.3
C ₂ -C ₁₁	[t/h]	3.5	Ethylene	[t/h]	10.8	PE	[t/h]	10.7
C ₁₂ -C ₂₄	[t/h]	9.9	Propylene	[t/h]	4.5	PP	[t/h]	4.5
>C ₂₄	[t/h]	10.9	Rest	[t/h]	9.0	Rest	[t/h]	0.2

With the obtained amount of polypropylene (PP) and polyethylene (PE) from the PtX process, the mass balances of the fossil route is recalculated and given in Table 3. Here it is clearly evident that a higher amount of naphtha is required for the same amount of polyolefins, i.e. about one third more. The steam cracker for fossil naphtha has correspondingly different conversion yields. The distribution between ethylene and propylene is the same as for the PtX process route. Furthermore, naphtha is provided by the distillation of crude oil. For the production of 15.1 t/h polyolefins, about 114 t/h (898,320 t/year) crude oil are required, assuming that light and heavy naphtha can be converted in the steam cracker. It should also be taken into account that only naphtha is included in olefin production and the rest is used for other applications.

Table 4: Calculation of the naphtha and crude oil demand in regard to the overall polyolefin production

Polymerization			Steam Cracker			Distillation – Crude Oil		
Total Olefins	[t/h]	15.3	Total Naphtha	[t/h]	32.6	Total Crude	[t/h]	113.8
PE	[t/h]	9.9	Ethylene	[t/h]	10.0	Naphtha	[t/h]	32.5
PP	[t/h]	5.2	Propylene	[t/h]	5.3	Rest	[t/h]	81.3
Rest	[t/h]	0.2	Rest	[t/h]	17.3			

Finally, Table 4 compares the CO₂ emissions generated by the conventional (fossil) and PtX process routes. In any case, the emissions from the fossil process route are significantly higher compared to the PtX process route. This is mainly due to the fact that the CO₂ in PtX process route 2 is provided as a resource for polyolefin production. In a broader sense, this means that 1.96 kgCO₂/kg_{polyolefin} is emitted during production in process route 1, while process route 2 is a CO₂ sink with 3.37 kgCO₂/kg_{polyolefin}. If the difference is considered, the so-called CO₂ saving potential, around 110 t of CO₂ can be saved per hour by switching to the PtX process route. This corresponds to 866,800 tons of CO₂ per year. It should be noted that the entire electrical supply must be provided by green electricity.

Table 2: Comparison of the emitted CO₂ amount for the conventional, fossil and PtX process route

CO ₂ emissions reduction				
Process route 1 Crude oil	PP	PE	Total	Saving potential
	[t/h]	[t/h]	[t/h]	[t/h]
	10.0	19.6	29.6	-110.1
	Specific CO ₂ [kgCO ₂ /kg _{polyolefin}]			
1.96				
Process route 2 PtX	Exhaust gas	Clean gas	Total	
	[t/h]	[t/h]	[t/h]	
	-88.8	8.3	-80.5	
	Specific CO ₂ [kgCO ₂ /kg _{polyolefin}]			
			-3.37	

Conclusion

In this paper, the comparison of two process routes for the production of polyolefins is considered and critically analyzed. The consideration of the mass balances of both process routes gives a clear statement that the use of CO₂ from the cement plant off-gas results in a reduction of the fossil naphtha and its production from crude oil. A PtX plant with an annual capacity of 700,000 tons of CO₂ per year would save 256,100 tons of fossil naphtha, which has to be produced from 890,440 tons of crude oil by energy-intensive distillation per year. With these 700,000 tons of CO₂ emitted annually, about 120,000 tons of polyolefins can be produced. This covers the demand of about one tenth of Austrians Borealis' production demand for polyolefins. Other point sources, e.g. from other cement plants, CO₂-rich waste gases from the steel industry or refinery, can be used as feedstock for the upscaling of such a plant.

Corresponding logistic considerations regarding decentralized separation plants in the factories on site and transport of CO₂ to a central intake and turnover location (e.g. transport of concentrated CO₂ from Mannersdorf via pipeline to the Schwechat refinery), as well as the production or import of green hydrogen have to be analyzed in detail and evaluated in the industrial consortium.

Under the circumstances that the PtX process route is fully powered by green energy, it is evident that the production of polyolefins has a clear advantage in CO₂ reduction. In contrast to production with fossil raw materials, which emits 1.96 kgCO₂ per kg of product, production with renewable energies saves CO₂ emissions of 3.37 kgCO₂ per kg of product. As a prerequisite, it must be mentioned here that the lifetime of the products does not play a role in this consideration, since the area of application is undefined for both process routes. If CO₂ emissions were taken into account, as in the lifecycle analysis, this would have a negative impact on the calculated CO₂ emissions for both process routes. In conclusion, it can be said that the conversion of polyethylene and polypropylene produced via a PtX plant into long-life products is desirable, since the carbon remains bound in the product in the long term. Carbon sequestration in a product over many years (e.g., more than 50) should also be considered carbon capture and storage in the most general sense.

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