CHIMICA & INDUSTRIA

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DECARBONIZING CHEMICAL INDUSTRY

Chemistry is the largest industrial energy consumer since it uses fossil fuels as both energy sources and raw materials. Most of the sector's CO_2 emissions are due to the production of hydrogen, ammonia, methanol, and light olefins. Several strategies will be discussed which may contribute to decarbonize their production even if the goal is hindered by the increase of their demand.

Introduction

Transition to carbon neutrality requires drastic changes to happen as summarized by the International Energy Agency in "Net Zero by 2050" report, which defines a roadmap to reach the net-zero CO_2 emissions (NZE) within 2050 [1]. Decarbonization of chemical manufacturing is crucial for achieving Net Zero 2050, as this sector is responsible for around 14.5% of all industrial CO_2 emissions (1,342 Mt vs 9,316 Mt) [2]. The chemical industry is particularly challenging to decarbonize due to its fundamental reliance on the inputs of fossil feedstock (coal, crude oil and natural gas), used both as energy source and raw material. Indeed, the chemical industry ranks as the largest industrial energy consumer even if it is just the third largest industry subsector in terms of

direct CO_2 emissions because a substantial portion of the carbon (and the energy) of the feedstock remains locked into its products. Thus, the CO_2 emissions of the overall chemical industry are lower than it may be expected from its energy demand (Fig. 1) [3].

However, should any organic chemical be burnt as a fuel, the carbon locked into it would soon result in CO_2 emissions. This actually happens for final products with a short life-cycle (e.g. single-use plastic) which are incinerated instead of recycled or buried in the landfills. While these emissions do not necessarily

14

happen within the physical boundary of the plant, they are a direct consequence of manufacturing strategies deployed at the production site **[4]**. To deeply decarbonize the chemical sector, few technology areas can be considered **[5]**:

- Energy efficiency and renewable energy, particularly to decarbonize process heat;
- Substitution of fossil feedstock for biomass;
- Production and use of green hydrogen;
- Use of CO₂ as building block;
- Combination with CCUS (Carbon Capture, Utilization, and Storage) technologies;
- Circular economy, including the recovery and recycle of plastic waste.

In the following, few examples will be discussed of the application of these technologies to the chem-



Fig. 1 - Energy demand and CO_2 emissions for different industrial productions. Mtoe (Million Tonnes of Oil Equivalent) (adapted from [3])



	Use as energy source, EJ/y	Use as feed- stock, EJ/y	Total, EJ/y
Oil	2.6	18.9	21.5
Natural gas	5.7	7.7	13.4
Coal	4.5	0.1	4.6
Electricity	4.6	-	4.6
Heat	2.4	-	2.4
Biomass and waste	0.1	1.0	1.1
Total	19.9	27.7	47.6

Tab. 1 - Energy and non-energy use of raw materials, heat, and electricity for petrochemical production (data 2017; EJ: exajoule, 10^{18} joule) [5]

ical productions which are the main responsible of CO₂ emissions.

Chemical production and energy

As already stated, in the case of the chemical and petrochemical sector, a large share of the energy inputs is used as feedstock, and around two-thirds of all carbon input is stored in chemicals.

Tab. 1 reports a breakdown of the energy sources used in the chemical industry, distinguishing between their energy and non-energy (*i.e.* as feedstock) uses **[5]**. Oil represents the main feedstock, followed by natural gas and biomass. On the other hand, energy is mainly provided by natural gas, electricity or coal.

Few major chemical productions account for more than 60% of the sector's total global energy and non-energy uses and related CO_2 emissions: high value chemicals (HVC, which include ethylene, pro-

	EJ/y
High value chemicals	21.2
Ammonia	6.2
Methanol	2.7
Carbon black	1.0
Total	31.1

Tab. 2 - Estimated global energy and non-energy use of raw materials, heat, and electricity for main petrochemical productions (data 2017) [5] pylene, butadiene, butenes, and BTX: benzene, toluene, and xylenes), ammonia, methanol and carbon black (Tab. 2) [5].

Global chemicals demand is projected to growth at around 2.5 fold from 2017 to 2050, even according to the ambitious 1.5 °C scenario of the pathway designed by IRENA to accelerate the energy transformation limiting climate change to 1.5 °C above pre-industrial levels. Accordingly, the energy and non-energy uses of raw materials, heat, and electricity are expected to increase from 47.6 EJ to 88 EJ [5].

Chemical production and GHG emissions

The climate-changing emissions include other gases beside CO_2 (e.g. CH_4 , F-gas, N₂O), even having a more detrimental impact. Therefore it is compulsory to take into account the global GHG emissions. For instance the CO_2 emissions in 2019 accounted for 36.9 Gt but, while considering the global GHG emissions taking into account the different impact of each one and referring them as CO_2 equivalent, the figure for 2019 becomes 49.8 Gt [6]. Again, while the chemical sector contributes with around 14.5% of CO_2 emissions its contribution rises to 21%. As a matter of fact, the chemical and petrochemical industry is the top emitter among all the industrial sub-sectors.

Adding also the emission of oil refining (which is also the precursor of practically all the petrochemical productions), the contribution becomes 30% of the industry emissions which, in their turn, represent the 31% of the world emissions. Therefore oil refining and chemical industry contributes with over 9% on the overall GHG emissions.

A detailed analysis of the impact of chemical production on GHG emissions has been recently reported by Isella and Manca [7]. Their analysis deals with commodity chemicals which are produced in quite huge amounts and are consequently associated to high direct and indirect GHG emissions. The analysis is based on the gate-to-gate GHG emission factor: this parameter, expressed in mass of CO_{2eq} per mass of product, quantifies the carbon footprint associated with a production plant from the entrance "gate" to the exit "gate".

Tab. 3 reports annual production, average gate-togate GHG factors and annual emissions for select-

	Annual production (2020), Mt	Average gate-to-gate GHG emissions, t CO ₂ eq/t	Annual GHG emissions as CO ₂ eq, Mt
Ammonia	185.4	2.37	440.3
Ethylene and Propylene (steam cracking)	239	0.95	227.6
Methanol	102.3	1.65	168.4
Chlorine	99	1.45	143.3
Nitric acid	65.9	1.55	102.4
PET including TPA	155.8	0.64	99.9
BTEX	163.9	0.49	80.1
Soda ash	60.3	1.09	65.6
PVC (including EDC and VCM)	130.4	0.4	52.7
Polyethylene	121.9	0.37	45.4
Adipic acid	2.7	16.67	45.0
Monoethylene glycol	27.6	1.11	30.7
Propylene (Fluid Catalytic Cracking)	34.8	0.87	30.2
Polypropylene	74	0.35	25.8
Ethylene oxide	29.1	0.88	25.6
Phosphoric acid	86.6	0.28	24.4
Propylene oxide	9.3	1.91	17.8
Styrene	31.1	0.51	15.9
Acrylonitrile	6.1	2.39	14.6
Phenol	11.2	0.88	9.9
Sulphuric acid	256	0.01	2.56
Cumene	15.1	0.12	1.8
Urea	181.5	-0.66	-120.2

Looking at the gateto-gate GHG factor, the most pollutant production is that of adipic acid, because of its emissions of NO, On the opposite side, next to urea is sulfuric acid showing the lowest gate-togate factor, of course not including the contribution of sulfur. Tab. 3 lacks of the contribution of hydrogen since it has been, so far, mainly used in oil refining rather than in the chemical industry: 87 Mt of hydrogen were produced from unabated natural gas, oil and coal in 2020 [8]. According to its gate-to-gate factor (7.24 t CO₂eq/t H₂) [7], the overall emission is 630 Mt of CO eq, so becoming the main contributor to GHG emissions.

Therefore hydrogen,

Tab. 3 - Production, average GHG gate-to-gate emissions, and GHG emissions per year of main chemical commodities. BTEX (Benzene, Toluene, Ethylbenzene, Xylenes); EDC (Ethylene dichloride); PET (Polyethylene terephthalate); PVC (Polyvinyl chloride); TPA (Terephthalic acid); VCM (Vinyl chloride monomer)

ed chemical commodities according to the data reported in **[7]**. The table is ranked according to the annual GHG emissions of each chemicals, obtained multiplying the annual production with the gate-to-gate GHG factor.

Ammonia is the most pollutant chemical, due to the sources of hydrogen utilized for its synthesis (e.g. natural gas, oil, or coal). Next are ethylene, propylene and other olefins produced by steam cracking (SC) of ethane, propane, virgin naphtha or condensate. These olefins are the building blocks of several chemicals and polymers. Third in the rank is methanol for reasons similar to those of ammonia. On the opposite of the rank, urea is the only carbon-negative major chemical as it is produced from CO_2 , together with ammonia.

16

ammonia, light olefins, and methanol productions are the most polluting processes of oil refining and petrochemistry and deserve the harder efforts to decarbonization.

Decarbonizing H₂ production

In the last years, a color coding scheme has been introduced to label the production routes of several chemical commodities according to their carbon intensity. Most of the hydrogen currently produced is "grey" H_2 , produced from methane via steam reforming (SMR, Steam Methane Reforming). Grey H_2 has a high CO₂ footprint, that can be even worse if oil fraction partial oxidation or coal gasification are used to produce it. This is why alternative production methods to make "blue" and "green" H_2 have been



	Technology	Feedstock	Energy source	CO_2 footprint, kg CO_2 /kg H ₂
Green	Electrolysis	Water	Wind	0.4
Green	Electrolysis	Water	Solar	1.5
Blue	ATR wit CCS	CH4		4
Blue	SMR with CCS	CH4		4.6
Grey	SMR	CH4		9
Pink	Electrolysis	Water	Nuclear Power	0
Turquoise	Electric plasma pyrolysis	CH4	Nuclear Power	-

Tab. 4 - Proposed classification of hydrogen production from different feedstock. The carbon footprint values (kg $CO_2/kg H_2$) are estimated according to cradle-to-grave life cycle assessments, so that they are larger than the corresponding gate-to-gate values. They are shown just for comparison purposes: actual figures are matter of debate. ATR (Auto Thermal Reforming); CCS (Carbon Capture and Storage); SMR (Steam Methane Reforming)

considered. Blue H_2 is also made from methane, oil fractions or coal, but has CO_2 capture added on the outlet. Green H_2 does not use at all fossil sources as a feedstock and is made by the electrolysis of water, using renewable energy in an electrolyzer (~70% efficiency). Electrolyzers are based upon alkaline, proton exchange membrane (PEM) or solid-oxide electrolysis (SOE) cells: a PEM electrolyzer requires 50 MW of electricity to produce 1 t of green hydrogen. Beside the technologies for producing blue and

green hydrogen, other routes have been proposed recently. Tab. 4 summarizes them according to Buehler [9].

Hydrogen, with its low- or zero-carbon footprint, will play a key role not only in oil refining and petrochemistry but also as a fuel, specifically in the production of renewable fuels, biofuels and e-fuels (electrofuels). This is why hydrogen consumption could increase by 6-10 times its current demand. Green, blue and pink hydrogen will likely meet this increased demand **[9]**.

Decarbonizing ammonia production

The production of ammonia is still based on the Haber-Bosch process, first industrialized in 1913, which involves the catalytic reaction of hydrogen and nitrogen at high temperature and pressure.

As previously reported, hydrogen is today mainly produced from fossil fuels and the ammonia so obtained is labeled as brown ammonia. Low-carbon

ammonia produced from blue hydrogen is referred to as blue ammonia, while green ammonia is obtained when green hydrogen is utilized. There are, however, other recognized routes to green ammonia. Among them, it is worth to cite the direct electrochemical production. This technology produces green ammonia directly from water and nitrogen using electricity. This way avoids the separate hydrogen production process step and would be ideal for distributed, small-scale generation and more suitable to intermittent power supplies. Haldor Topsoe is developing a demonstrator that integrates a solid oxide electrolysis cell (SOEC) to produce ammonia synthesis gas (H_0 : N_0 = 3:1). This is then converted to ammonia

via the conventional process. The process operates at high temperatures and can separate oxygen from air without using an air separation unit (ASU). This results in some reduction of the energy consumption per t of ammonia: 5-10% lower with respect to a conventional SMR-based process.

The production of green ammonia will positively impact the transition towards zero-carbon through the decarbonization of its current major use in fertilizer production: it should be indeed emphasized that, according to the figures already provided, ammonia synthesis accounts for almost 1% of global GHG emissions. Moreover, green ammonia may have other potential uses **[10]**:

- as a medium to store and transport chemical energy. Then the energy may be released either by combustion with air or by the full or partial decomposition to release hydrogen;
- as a transport fuel, by direct combustion in an engine or through chemical reaction in a fuel cell to produce electricity to fuel a motor;
- to store thermal energy through phase changes between material states. In fact ammonia can also store and release significant energy on changing between its liquid and gas forms (1371.2 kJ/kg at atmospheric pressure). This property may be valorized in heat pumps, using low-grade waste heat to generate heated water.

With its quite high energy density (~3 kWh/L) and existing global transportation and storage infra-



Fig. 2 - Proposed classification of methanol production from different feedstock. Renewable CO₂: from bio-origin and through direct air capture; Non-renewable CO₂: from fossil origin (adapted from [12])

structure, ammonia could supply a new integrated renewable energy storage and distribution solution. Accordingly the ammonia demand is forecasted to grow of a CAGR (Compound Annual Growth Rate) of 8% from now to 2031, with green ammonia growing of a CAGR of 75% [11].

Decarbonizing methanol production

Methanol can be produced from different carbon sources, such as natural gas, coal, biomass, or even CO_2 . Fig. 2 summarizes the possible production routes of methanol as recently reported by IRE-NA and Methanol Insitute **[12]**.

According to Fig. 2, the production of methanol can be categorized as high or low carbon intensity, depending on the feedstock and associated carbon emissions. It will be considered as high carbon intensity if fed with coal or natural gas without CO_2 capture, nor renewable power input (brown and grey methanol). Productions based on renewable energy and feed or on fossil fuels with CO_2 capture, or a combination thereof, will be considered as low-carbon intensity (blue and green methanol). To be classified as renewable, methanol must be produced from biomass or water using renewable energy, *i.e.* solar, wind, hydro, geothermal, etc. energy.

At any rate, for economic reasons methanol is currently produced almost exclusively from fossil fuels. About 65% of methanol production is based on nat-

18

ural gas *via* SMR (grey methanol), while the rest (35%) is largely based on coal (gasification, brown methanol) **[13]**. Currently, only about 0.2% comes from renewable sources (green methanol).

The conventional production of methanol is based on the catalytic reaction of synthesis gas (shortly syngas), a mixture of CO and H_2 which is obtained from natural gas or coal and which is an intermediate also in the productions of hydrogen and ammonia, already discussed. Then syngas is converted into methanol by a catalytic reaction, usually over copper-zinc catalysts on alumina. Methanol plants from natural gas have typical production capacities of 1-1.7 Mt/y [14].

Renewable methanol can be produced by two main routes:

- bio-methanol from biomass. Sustainable biomass feedstocks include forestry and agricultural waste, biogas, sewage sludge, municipal solid waste (MSW), and black liquor from the pulping industry;
- e-methanol obtained from captured CO₂ and green hydrogen.

The technologies used in the production of methanol from biomass or MSW are similar to that used in the gasification-based industry, where the feedstock is usually coal or heavy oil residues. Also for biomass or MSW the main process steps are: feedstock pretreatment, gasification (syngas production), water gas shift (WGS) reaction to increase the hydrogen content of syngas, gas cleaning, methanol synthesis and purification. Several companies have been developing technologies for the production of methanol from MSW. Among them Enerkem in Edmonton Canada has been operating a demo plant since 2015. 60 kt/y of MSW feedstock is gasified and converted to methanol. In 2017 a methanol to ethanol conversion unit was installed, therefore the plant is since producing ethanol from MSW.

In Italy NextChem, Maire Tecnimont Group's company, is proposing a chemical recycling technology to produce Circular Methanol[™] from MSW, refuse derived fuel (RDF, a fuel produced from various types of waste) as well as non-recyclable plastic waste (PW) **[15]**.



E-methanol is obtainable from CO_2 and green hydrogen through a one-step catalytic process. E-methanol is considered an electrofuel (e-fuel) and its production is classified as a Power-to-X (P2X) process. The reaction of CO_2 with hydrogen from water electrolysis is currently the only practical method to produce e-methanol. Each molecule of CO_2 requires three molecules of hydrogen and produces one molecule of water for each molecule of methanol. The catalytic step is very similar to that of methanol synthesis from syngas but the traditional CuO/ZnO/ Al_2O_3 catalyst has been slightly modified to accommodate the formation of larger amounts of water during the production of e-methanol **[16]**.

The first commercial CO_2 -to-methanol plant has been operated in Iceland by Carbon Recycling International (CRI) since 2011. This demo-unit has a capacity of 4 kt/y of methanol. Hydrogen is produced by water electrolysis using cheap geothermal electricity. The produced methanol, called Vulcanol, is currently mixed with gasoline or used for biodiesel production **[12]**.

Renewable methanol could play a larger role in decarbonizing certain sectors, *e.g.* as a feedstock in the chemical industry or as a fuel in road or marine transport. This is why the global methanol consumption is expected to reach 500 Mt by 2050 **[5]**, practically 5 times the 2020 demand.

Decarbonizing the light olefin production

Steam cracking (SC) is the thermal processing of fossil feedstock (*e.g.* ethane, propane, virgin naphtha, condensate) affording HVC, *i.e.* ethylene, propylene, butenes, butadiene, and BTX. It is one of the most energy intensive processes of petrochemistry. As its products, mainly ethylene, serve as the major building blocks of the polymer industry, the critical issues regarding the broader environmental impacts of plastic products adds further relevance. The SC process was estimated to emit 227,6 mil-

lion tons of CO_{2eq} globally in 2020. SC is initiated by thermally breaking C-C and C-H bonds (above 800 °C) forming radicals which propagate, isomerize, and terminate to form a distribution of products (olefins and aromatics) and byproducts (hydrogen, methane and tars). The products yields depend on feedstock and furnace run conditions, such as temperature and residence time. The process is endothermic and the emissions comes from the use of byproducts methane and hydrogen as a fuel in the furnace.

To reduce these emissions, direct electrification is currently proposed by a number of chemical companies. The "Cracker of the Future" consortium, aimed to develop a technology for the electrification of the steam cracking process, includes Borealis (member of the OMV Group), BP, Total Energies SE, Repsol and Versalis (Eni). The consortium covers ~1/3 of the European Union's SC capacity with units in Austria, Belgium, Finland, France, Germany, Italy, Portugal, Spain, and Sweden **[17]**.

Shell Chemicals and Dow have started up an experimental unit to electrically heat steam cracker furnaces at the Energy Transition Campus Amsterdam, The Netherlands, in June 2022 [18].

More recently BASF, Sabic and Linde have announced to have signed a cooperation agreement for the development and demonstration of solutions for electrically heated steam cracker furnaces [19].

The extent of emission reduction by direct electrification is obviously dependent on the electricity source. However, direct electrification of SC alone, even with renewable electricity, cannot bring emissions to a level compatible with a carbon neutral economy, because of the residual emissions from oil refining and the energetic utilization of fossil-based off-gas, that make up 70% of the overall emissions **[20]**. For such a reason the olefins obtained by SC, even electrified with green electricity, are still considered grey olefins **[21]**.

Better emission reduction could be obtained by indirect electrification, i.e. combining water electrolysis with methane oxidative coupling (OCM) to ethylene **[20]**. The process scheme is summarized in Fig. 3. However OCM can supply only ethylene, without the other olefins, in particular propylene and butadiene.



	Feedstock	Technology	Notes	
Brown	Coal	Gasification/MTO		
Grey	Ethane, Propane, Virgin Naphtha, Condensate	Steam Cracking		
Grey	Ethane, Propane, Virgin Naphtha, Condensate	Electro-Steam Cracking	Renewable electricity	
Blue	CH_4	SMR/MTO	In combination with CCS	
Purple	Coal	Gasification/MTO	In combination with CCS	
Pink	Plastic waste	Gasification/MTO		
Pink	Plastic waste	Pyrolysis		
Green	Bioethanol	Dehydration		
Green	Biomass	Pyrolysis - Liquefaction/ Cracking	Bio-crude stabilized by green H ₂	
Green	Biomass	Gasification/MTO		
Green	CO ₂	P2X/MTO	P2X by green H ₂	
Tab 5 (Tab. 5 - Comparison of different olefin production routes [21]			

Tab. 5 - Comparison of different olefin production routes [21]

Another way to produce olefins is the Methanol-to-olefins (MTO) process, among the most important C1 conversion routes enabling the production of basic olefins and petrochemicals from methanol. 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. The reaction of methanol is catalyzed by acidic zeolites, properly selected to selectively drive the formation of light olefins, in particular ethylene and propylene. Depending on the origin of methanol, the obtained olefins can be considered brown, blue, purple, pink or green as summarized in Tab. 5.

Brown olefins *via* MTO represent a relevant share (40%) of olefin production in China, where coal is particularly utilized. However this coalto-olefins (CTO) process is by far more carbon intensive compared to e.g. naphtha cracking **[21]**.

Several routes have been proposed for the production of green olefins from biomass. The most promising are: the dehydration of cellulosic ethanol obtained by fermentation of sugar derived by saccharifica-

20

tion of lignocellulosic biomass; the upgrading and cracking of bio-oils produced from biomass by liquefaction or pyrolysis; the MTO process fed with methanol obtained from any biomass *via* gasification to syngas followed by methanol synthesis. These routes are depicted in Fig. 4.

Finally, olefins can be produced from recycled plastics according to a circular economy approach. The plastic waste properly collected can be recycled into low molecular weight olefins which can then be further upgraded to monomers to produce again polymer resins. Two main thermochemical processes based on pyrolysis and gasification are the most mature technologies in the field. Olefins produced from recycled plastics

are classified as pink. Pink olefins have generally higher production costs than grey olefins, however the difference is not prohibitively high [21].

According to **[5]** the production of HVC will slightly increase from 2017 to 2050, passing from 369 to 448 Mt (+21.4%). In the meantime plastics will almost double, moving from 348 to 659 Mt. This will be possible as the contribution of recycled plastics will increase significantly (6.5 times). Moreover, the contribution of biomass feedstock will increase for both HVC and plastics. The combination of these two contributions will result in ~25% reduction of fossil feedstock for HVC production.



Fig. 4 - Green olefins from biomass (adapted from [21])



Conclusion

Carbon neutrality of chemical and petrochemical industry is a feasible target for 2050. Today, fossil feedstock (coal and hydrocarbons) is at the core of the chemical and petrochemical industries, making CO₂ mitigation strategies very challenging. A set of options have been identified that can be deployed for this purpose, mainly devoted to the most carbon intensive productions, *i.e.* hydrogen, ammonia, methanol and light olefin monomers.

These options include the development of renewables-based process heating (e.g. electrification of steam cracking), substitution of fossil feedstock for biomass (e.g. in the production of green olefins), circular economy approach (e.g. by recycling plastic waste), synthetic chemicals (e.g. ammonia) from green hydrogen, and CO_2 capture and utilization (e.g. in the methanol production).

Lower-carbon routes to chemicals will likely be more expensive than today's routes: according to Bloomberg NEF, decarbonizing petrochemicals by 2050 will cost nearly \$800 billion **[22]**. However, this target may no longer be postponed.

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Decarbonizzare l'industria chimica

Tra i vari comparti industriali, quello chimico è responsabile del maggior consumo di energia e combustibili fossili. Più del 60% delle sue emissioni di CO_2 sono dovute alla produzione di idrogeno, ammoniaca, metanolo e olefine leggere. Sebbene la domanda di queste *commodities* sia in continuo aumento, si stanno delineando più strategie per la decarbonizzazione delle loro produzioni.