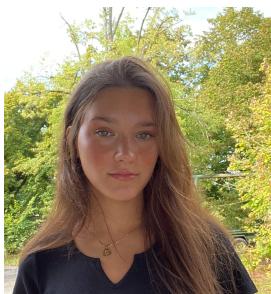




Challenges of sustainable chemistry

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How can CO₂ be efficiently converted into valuable fuels and materials while reducing greenhouse gas emissions?

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I. Introduction

Carbon dioxide (CO₂) is today one of the main contributors to climate change. Mostly released from the combustion of fossil fuels, it traps heat in the atmosphere, causing a gradual increase in global temperatures. The consequences are already visible: more frequent and intense heatwaves, droughts, wildfires, floods, melting ice, and rising sea levels, with direct impacts on biodiversity, food production, and human societies. According to the latest IPCC report, limiting global warming to 1.5 °C by the end of the century requires a massive and rapid reduction in CO₂ emissions, with immediate action on a global scale [1]. Scientists warn that every fraction of a degree matters, and the time left to act is extremely limited.

The graph below reproduces the IPCC SPM.10 diagram (1850–2019) and shows the near-linear relationship between cumulative anthropogenic CO₂ emissions and the increase in global temperature. Since 1850, human activities have emitted about 2,411 GtCO₂, leading to a warming of roughly 1.1–1.2 °C. This relationship makes it possible to estimate how much additional CO₂ emissions would push global temperatures beyond the 1.5 °C or 2 °C thresholds [2].

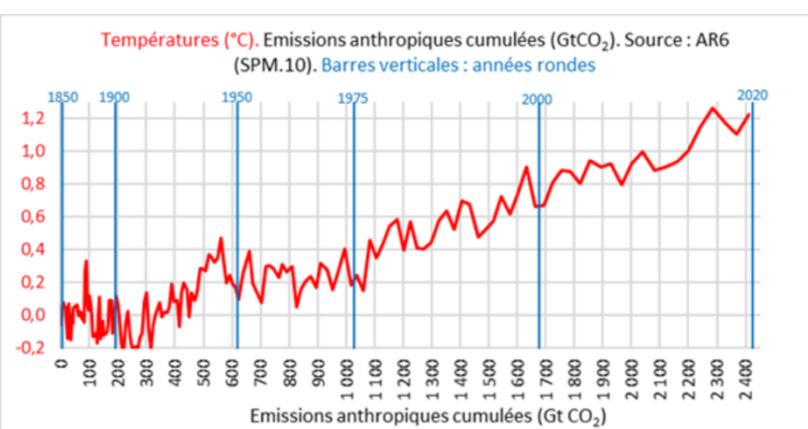


Figure 1: Relationship between global warming and cumulative CO₂ emissions (based on the IPCC, SPM.10)

Faced with this urgent situation, carbon capture, utilization and storage (CCUS) is

emerging as a key solution for the future. CO₂ capture consists of isolating the gas directly from the emissions of power plants, factories, or other industrial facilities. Among the most promising technologies are amine-based solvent absorption effective but energy-intensive and the use of zeolites, microporous solid materials capable of selectively separating CO₂. Once captured, CO₂ can be converted into high-value products such as methanol, used as a fuel or chemical intermediate, and epoxide monomers, which are essential for producing biodegradable polycarbonates and other sustainable materials.

These processes are not just technical innovations: they represent a strategy for the future, transforming a problematic gas into a valuable resource. Modern organometallic and bimetallic catalysts now make it possible to produce these polymers with high selectivity and efficiency, reducing both carbon footprint and energy consumption. This paves the way for a circular economy in which CO₂ is reused, decreasing reliance on fossil raw materials and supporting industrial sustainability.

The urgency of the situation is reinforced by the most recent data: global greenhouse gas emissions reached around 53.5 billion tonnes of CO₂ equivalent in 2023 [3], continuing to rise despite mitigation efforts. These figures highlight planetary limits and the need for innovative and rapidly deployable solutions to avoid irreversible climate impacts.

CO₂ valorization is therefore not just a scientific challenge, but a strategic necessity to ensure climate resilience, energy security, and long-term sustainability. By turning CO₂ into a raw material rather than a waste product, it becomes possible to reconcile industrial development, environmental protection, and the transition toward a low-carbon economy, offering concrete solutions for future generations

II. State of the Art and Discussion

1) Post-combustion CO₂ capture using amine-based solvents.

The reduction of carbon dioxide (CO₂) emissions from industrial activities particularly from flue gases is one of the major challenges of the 21st century. Among the technologies considered to mitigate the climate impact of highly emitting sectors, post-combustion capture using amine solvents has emerged as one of the most widely studied solutions. Initially developed for natural gas purification, this process has gradually been adapted to industrial effluents such as those from coal-fired power plants, cement factories, and steel mills. Its operation relies on a relatively simple chemical principle: the reversible absorption of CO₂ in an aqueous amine solution, followed by the thermal regeneration of the solvent.

The process operates through two main steps: absorption and regeneration. In an absorption column, the still-hot flue gases (around 40 to 60 °C) flow counter-current to an aqueous amine solution, such as monoethanolamine (MEA). The carbon dioxide binds to the amine to form an intermediate compound, carbamate, according to the reaction:



This exothermic reaction leads to the formation of a “CO₂-rich” solvent, which is then sent to a regeneration column (or stripper). In this second column, the solvent is heated to around 120 °C; the supplied heat reverses the chemical reaction, releasing pure carbon dioxide and regenerating the

solvent. The regenerated “lean” solvent is then recycled back to the absorber for a new cycle. The released CO₂ can be compressed for geological storage or used in various chemical processes [4].

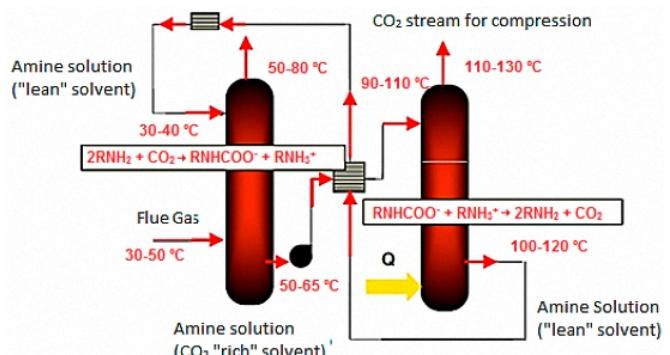


Figure 2: Process diagram for CO₂ absorption using amines, with the corresponding temperatures and reactions.

The reaction between CO₂ and amines relies on the basic nature of the latter. Primary and secondary amines, such as monoethanolamine (MEA) or diethanolamine (DEA), react rapidly with carbon dioxide to form stable carbamates.

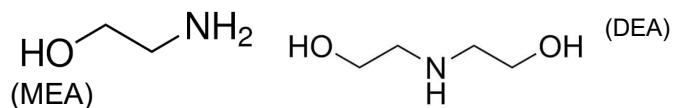


Figure 3: MEA and DEA molecules

This type of chemical bonding enables efficient CO₂ capture but requires significant energy for regeneration. Tertiary amines (R₃N) such as methyl-diethanolamine (MDEA), behave differently: they do not form carbamates but rather bicarbonates,

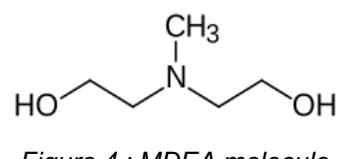


Figure 4 : MDEA molecule

which are less stable and require less energy to break down.

Since the nitrogen is fully substituted, it can no longer form a covalent bond with CO₂.

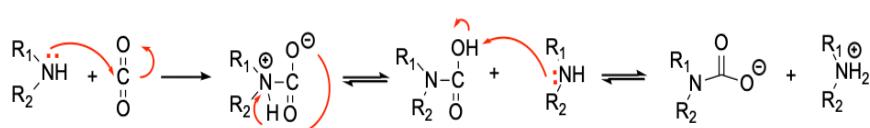
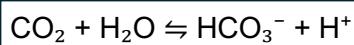


Figure 5: Mechanism of carbamate formation from amines and CO₂

Capture in this case relies on an acid-base reaction:



In this case, the fully substituted nitrogen does not react directly with CO₂ but acts as a base by capturing a proton (H⁺) from water thereby catalyzing the formation of bicarbonate.

This difference explains why tertiary solvents, although less reactive, are often preferred in processes aiming to reduce energy consumption. Numerous studies, notably those by Kim et al. (2016) [5], have shown that the reaction between CO₂ and amines occurs in two successive steps: the formation of a zwitterionic intermediate (a molecule carrying both a positive and a negative charge but remaining overall electrically neutral), followed by deprotonation leading to carbamate. In amine mixtures, such as monoethanolamine (MEA) and piperazine (PZ), CO₂ capture is enhanced through the synergistic effect of the components. One of the amines, for example PZ, acts as a catalytic base, facilitating the reaction between CO₂ and MEA.

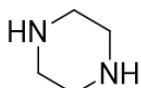


Figure 6 : Piperazine

This interaction lowers the reaction's energy barrier and accelerates the adsorption kinetics, which explains the superior efficiency of amine mixtures compared to single amines.

The mechanism occurs in two steps: first, the formation of a zwitterion RNH₂⁺-COO⁻, followed by deprotonation by a second amine molecule or water. The equilibrium constant, K_{carbamate}, is highly temperature-dependent (~10⁴ at 40 °C) and decreases at higher temperatures, favoring desorption [6].

The reaction of CO₂ with a Lewis base (blue) produces a zwitterion. If the amine has a proton, the zwitterion can rearrange into the corresponding carbamic acid. A second sufficiently basic amine (pink) can

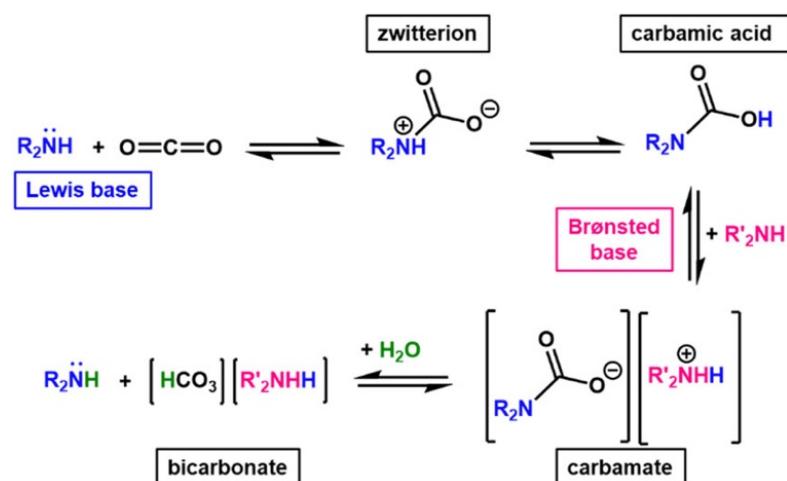


Figure 7: Mechanism of carbamate formation from CO₂ and an amine

then form the carbamate. In the presence of water (green), the carbamate is converted into bicarbonate. Colors are used to help visualize the reactions [7].

The efficiency of this process strongly depends on operating conditions: temperature, pressure, amine concentration, and solvent flow rate. In industrial systems, MEA solutions are typically used at 30–40 wt%, operating at atmospheric pressure. Too low a temperature reduces the reaction rate, while excessively high temperatures favor premature CO₂ desorption. Balancing these parameters allows optimization of capture rate and energy consumption.

CO₂ capture using amine solvents offers many advantages that explain its status as a reference technology. It is based on a proven principle, well-known to engineers, and has already been applied in several pilot and industrial installations. The Boundary Dam power plant located in Saskatchewan, Canada, commissioned in 2014 is a prominent example of post-combustion capture using amine solvents. The facility uses the Cansolv technology developed by Shell, based on a regenerable amine solvent, and can capture up to 90% of the CO₂ in the treated flue gas amounting to more than 5 million tonnes of CO₂ captured since its commissioning [8].

Moreover, the process is reversible: the solvent is regenerated and reused over multiple cycles thus limiting material

consumption. Its compatibility with existing facilities makes it a realistic transitional solution toward a low carbon industry. Finally, the purified CO₂ can be valorized: used in the production of methanol, urea, or carbonates, or injected into oil reservoirs to enhance recovery (EOR).

However, despite these advantages, several major limitations hinder the large-scale deployment of this technology. The main drawback lies in the high energy consumption required for thermal regeneration. The energy needed to heat the solvent is approximately 3.5–4 GJ per tonne of CO₂ captured, leading to an 8–12% reduction in the overall power plant efficiency. This energy penalty represents a significant economic obstacle, especially since the heat is often supplied by the steam produced by the plant itself.

Additionally, amine solvents gradually degrade under the combined effects of heat, oxygen, and impurities present in the flue gas (NO_x, SO₂).

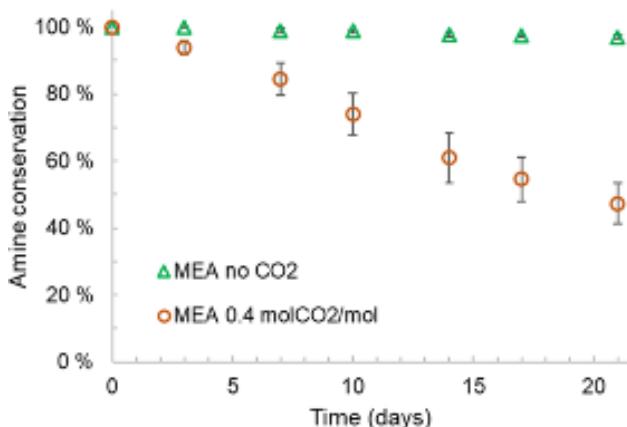


Figure 8: Oxidative degradation of MEA in the absence and presence of CO₂

This degradation leads to the formation of undesirable, sometimes corrosive, by-products and reduces the solvent's lifespan. MEA oxidation produces aldehydes (glycolaldehyde, formaldehyde) and acids (formic, oxalic). In the presence of NO_x, nitrosamines can form carcinogenic compounds that require strict emission control. Periodic solvent replacement is therefore necessary, increasing operational costs. Since the reaction environments are hot, basic, and CO₂ rich, they are also highly corrosive to metallic materials. Equipment must be constructed from

resistant alloys (stainless steel, Inconel, etc.), which increases initial investment. Finally, a fraction of the solvent can be carried over in the outlet gas as aerosols, requiring additional scrubbing and filtration systems to prevent secondary emissions [9].

Given these constraints, several improvement strategies have been proposed. Optimizing operating parameters such as increasing the stripper pressure to around 2.2 bar and using a 35–40% amine solution could reduce the process's energy consumption by approximately 20%. Additionally, better thermal integration, for example through heat exchangers or the utilization of waste heat, contributes to improving the overall system efficiency [10].

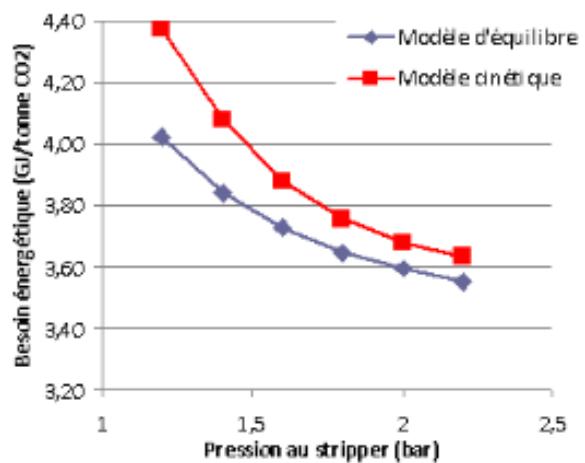


Figure 9: Influence of stripper pressure (a) and solvent concentration (b) on the process energy requirement.

Research is also focusing on the development of new solvents. Amine mixtures (MEA/PZ, MDEA/PZ) offer a good compromise between reactivity and low regeneration energy. Biphasic solvents, which spontaneously separate a CO₂ rich phase from a lean phase, help reduce the volume that needs to be heated. New compounds, such as sterically hindered amines, ionic solvents, or deep eutectic solvents, are also being studied for their stability and enhanced absorption capacity. Additives can be incorporated to limit corrosion and thermal degradation of the solvent [11].

Amine solvents degrade over repeated cycles producing impurities such as organic acids, ammonium salts, and dissolved nitrogen oxides. These by products reduce process efficiency and can pose environmental problems if not properly treated. Continuous purification systems are currently under study, including activated carbon filters, vacuum distillation, or membrane processes, which allow impurities to be removed without fully replacing the solvent.

Additionally, researchers are investigating the chemical recycling of degraded amines to extend their lifespan and reduce operating costs [12].

A particularly promising approach involves replacing thermal regeneration with electrochemical regeneration. The EMAR (Electrochemically Mediated Amine Regeneration) process, developed by Wang et al. (2020) [13], uses copper electrodes to release CO₂ under an electric current, without relying on steam. This fully electric alternative is more compact and can potentially be powered by renewable energy. Even more innovatively, some teams, such as Leverick et al. (2023), propose directly integrating the electrochemical conversion of captured CO₂: instead of regenerating the amine to release the gas, CO₂ is reduced *in situ* into valuable products (CO, methanol, hydrocarbons). This “reactive capture” combines the capture and conversion steps and could revolutionize carbon valorization by eliminating the thermal desorption phase entirely [14].

Other innovations concern the design of the equipment itself. The use of Rotating Packed Beds allows for intensified mass transfer and reduced equipment size [15]. The use of solid amine-functionalized materials, although still experimental, also opens interesting prospects for capture in drier or higher-temperature environments [16].

Finally, integrating CO₂ capture with other industrial processes, such as heat recovery in cement plants or refineries, can help improve the economic viability of the process.

Thus, CO₂ capture using amine solvents remains a reference technology, though not

without challenges. The use of amines in industrial settings raises chemical safety and occupational health concerns. Some amines, particularly monoethanolamine (MEA), are corrosive and can emit vapors that irritate the respiratory tract. Toxicological studies therefore recommend sealed installations, controlled ventilation, and fine particle filters to prevent diffuse emissions [17].

Recent research also focuses on the development of more “green” solvents, which are less volatile and biodegradable, such as amino acids or ionic solvents [18]. These compounds offer a promising alternative in terms of safety and performance. High capture efficiency, compatibility with existing infrastructure, and the potential for CO₂ valorization make amine-based capture a credible solution for transitioning to a cleaner industry. However, high energy consumption and solvent degradation remain major barriers to widespread adoption. Current research efforts aim to overcome these obstacles by developing more stable, less energy-intensive solvents, electrochemical regeneration processes, and better integration with renewable energy.

Sterically hindered amines, such as 2-amino-2-methyl-1-propanol (AMP), have been identified as promising candidates for reducing regeneration energy [19]. The presence of bulky substituents around the nitrogen limits the formation of strongly bound carbamates and favors the formation of bicarbonates that are more easily regenerated. These solvents thus allow a reduction in energy consumption of approximately 15–25% compared to monoethanolamine (MEA), while maintaining satisfactory capture kinetics in industrial gas streams.

AMP/piperazine or AMP/MDEA mixtures combine reaction rate and chemical stability, making them a viable option for large-scale industrial applications.

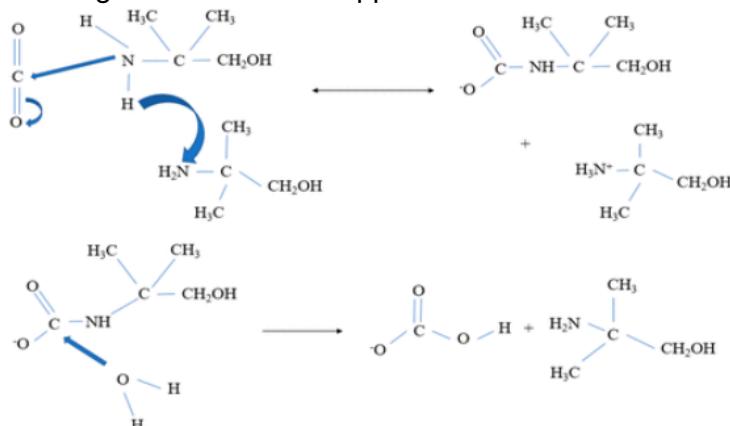


Figure 10: Reaction mechanism of AMP with CO₂

In summary, CO₂ capture using amines offers many advantages. It is based on a well-understood chemical principle that has already been proven at an industrial scale with regenerable solvents that integrate easily into existing facilities. This technology provides great flexibility and can be combined with CO₂ valorization to produce methanol, urea, or other chemical products. Recent advances, such as “chemical memory” solvents or electrochemical regeneration, open new opportunities to reduce energy consumption and make the process more sustainable.

However, there are still important limitations. Thermal regeneration remains energy-intensive and reduces the overall efficiency of the installations. Solvents degrade under the effects of heat, oxygen, and impurities in flue gases, producing corrosive or toxic by-products. Equipment corrosion and the management of amine emissions also present economic and operational challenges. Finally, the energy and material costs as well as the complexity of recycling degraded solvents, hinder large-scale deployment.

Despite these challenges, amine-based capture remains a benchmark technology with strong potential for a low carbon industrial future. By improving energy efficiency, extending solvent lifetime, and integrating renewable energy or direct CO₂ valorization processes, this transitional solution could become a truly competitive and sustainable technology.

2) CO₂ capture using zeolites

While amine solvents are widely used for CO₂ capture, other materials offer promising alternatives. Among them, zeolites play a central role due to their microporous crystalline structure and high selectivity for certain gas molecules. Zeolites are aluminosilicates composed of interconnected SiO₄ tetrahedra linked by oxygen bridges forming a three-dimensional network of nanometer-sized pores and cages.

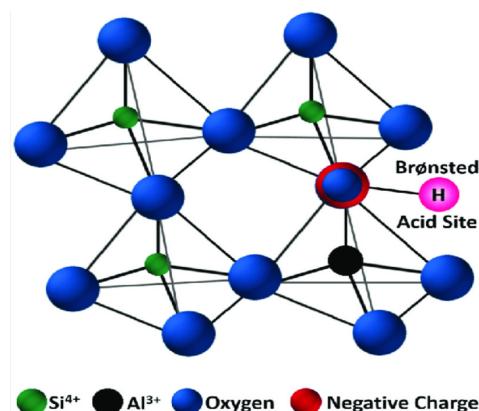


Figure 11: Crystal structure and typical microporous network of an aluminosilicate zeolite

The partial substitution of Si⁴⁺ with Al³⁺ generates a negative charge on the framework, which is balanced by extra-framework cations such as Na⁺, K⁺, Ca²⁺,

or Cs⁺. The nature and distribution of these cations play a fundamental role in CO₂ adsorption and in selectivity toward other gases, such as methane (CH₄) or nitrogen (N₂) [20].

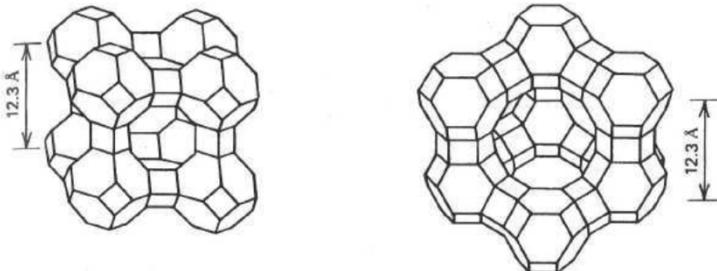


Figure 12: Crystal structure and microporous network of X and Y zeolites

X and Y zeolites (Figure 12) are representative examples of large-pore zeolites widely used for industrial CO₂ capture. Among the zeolites studied, 13X (NaX) is the most commonly used for CO₂ capture [21].

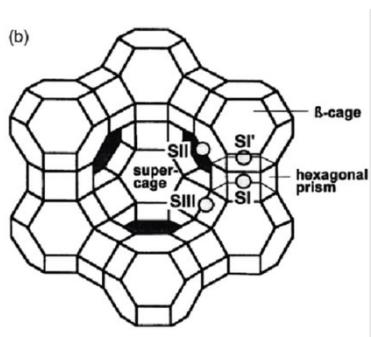


Figure 13: Schematic representation of the NaX zeolite used for CO₂ capture.

With a Si/Al ratio of approximately 1.3, 13X zeolite contains a significant proportion of aluminum in its structure. This creates numerous negatively charged sites that attract CO₂ molecules, explaining its high affinity for this gas. At the same time, this composition provides good mechanical and chemical stability in industrial adsorption processes, particularly those conducted under vacuum (VSA) or pressure swing adsorption (PSA), where CO₂ is

successively adsorbed and desorbed for recovery.

Its main drawback remains its sensitivity to water, which reduces capture efficiency in humid gas streams. Water molecules (H₂O) are highly polar and interact more strongly than CO₂ with the charged sites of the zeolite (Na⁺, Ca²⁺, etc.). When they enter the micropores, they occupy the same adsorption sites as CO₂ and form hydrogen bonds with the aluminosilicate framework. This prevents CO₂ from binding effectively and thus decreases the overall adsorption capacity [22].

Depending on the size of their pores and cages, zeolites are generally classified into two categories: small-pore zeolites, with openings between 0.30 and 0.45 nm, and medium- to large-pore zeolites, with pore sizes ranging from 0.45 to 0.80 nm [23].

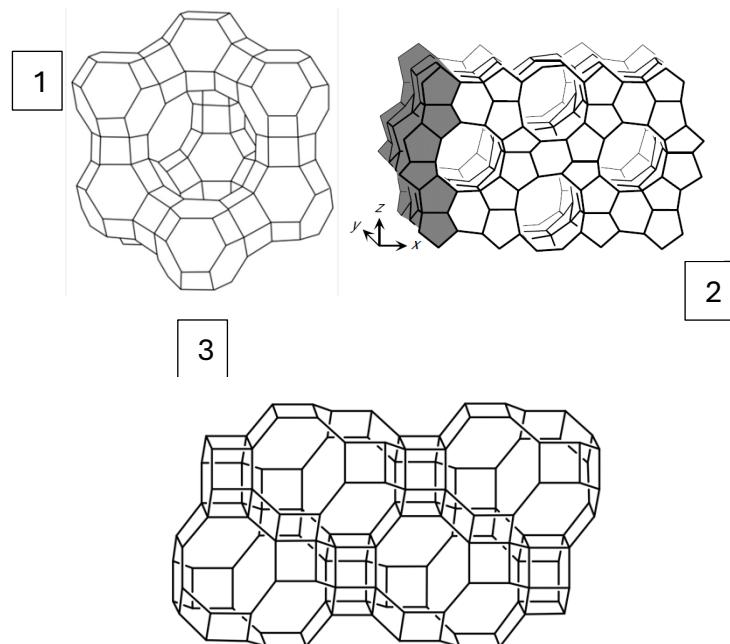


Figure 14: Representative structures of zeolites according to pore size: FAU 1 (large pores), MFI 2 (intermediate pores), and CHA 3 (small pores)

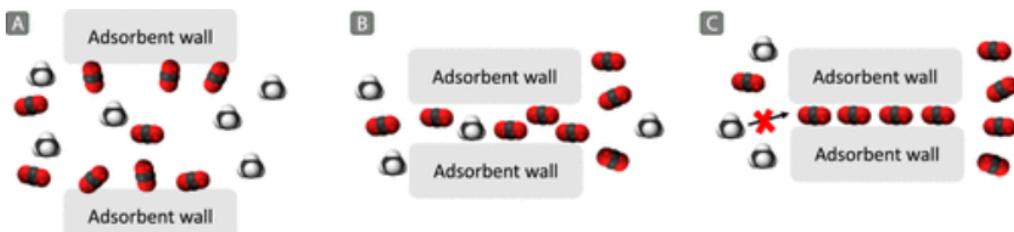


Figure 15: CO₂ separation mechanisms in zeolites:
 (A) Equilibrium separation: CO₂ is preferentially adsorbed on the material's surface.
 (B) Kinetic separation: CO₂ diffuses faster than CH₄ due to its smaller diameter.
 (C) Molecular sieving: only pores large enough allow CO₂ to pass, while CH₄ is blocked.

This distinction is important because it directly influences the CO₂ separation mechanism. In medium- and large-pore zeolites, adsorption mainly relies on electrostatic interactions between CO₂ and extra-framework cations, a process referred to as equilibrium separation.

In contrast, in small-pore zeolites the diffusion of CO₂ within the micropores plays a crucial role leading to kinetic separation, where CO₂, being smaller and more mobile, diffuses faster through the pores than other gases. In some cases, when the pore size is just sufficient to allow CO₂ to pass but blocks larger molecules such as CH₄ or N₂, this is known as molecular sieving. This mechanism thus acts as an atomic scale filter, enabling highly selective CO₂ separation. (Figure 15)

Small-micropore zeolites are often synthesized as nanocrystals to optimize their performance for CO₂ capture. As explained by Debost (2019), reducing the crystal size increases the available specific surface area, providing more adsorption sites for CO₂. Additionally, shorter diffusion paths allow CO₂ molecules to access the pores more quickly, thereby improving adsorption kinetics and selectivity compared to other gases such as N₂ or CH₄.

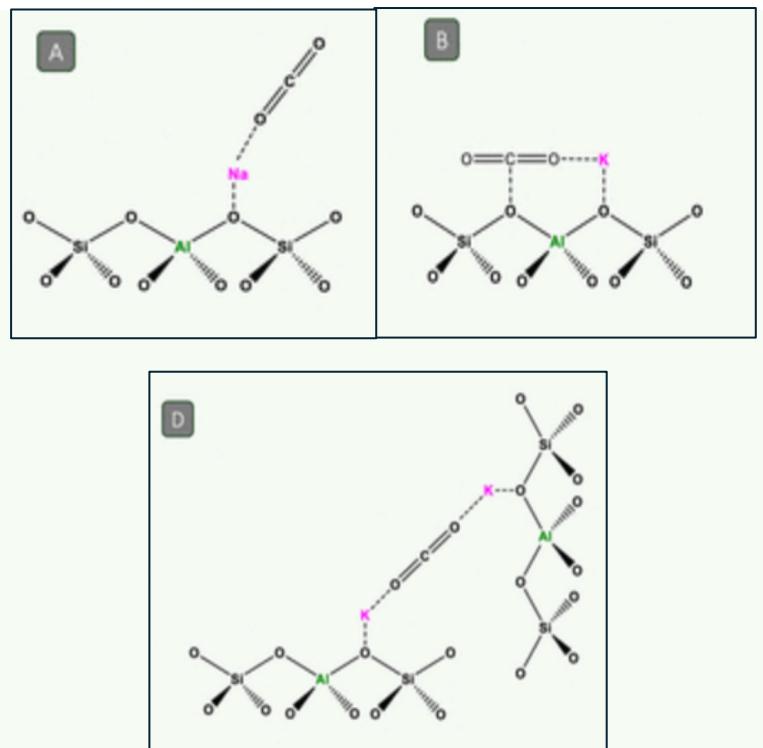


Figure 16: CO₂ interaction mechanisms with extra-framework cations in zeolites
 (A) Simple interaction of one oxygen of CO₂ with a cation.
 (B) Formation of a carbonate precursor: one oxygen of CO₂ interacts with a cation while the carbon interacts with an oxygen atom of the zeolite framework.
 (D) Bridging interaction: both oxygens of CO₂ interact with two distinct cations, stabilizing the molecule within the cage.

Moreover, zeolite synthesis depends on several parameters that directly influence their structure and CO₂ adsorption capacity. The composition of the mixture (sources of silica and alumina, Si/Al ratio, cations present), the amount of water, and the

alkalinity determine pore formation and the density of charged sites. Temperature, aging time, and stirring affect crystal size and uniformity. The use of organic structure-directing agents or seed crystals helps guide growth and produce nanocrystals with regular micropores. By controlling these conditions, zeolites with pores optimized for selective CO₂ capture can be obtained [24]

The CO₂ molecule can interact in different ways with the extra-framework cations located in the supercage. The most common interactions are as follows: first, a simple mechanism where one oxygen of CO₂ binds to a cation; second, a bridging mechanism where both oxygens interact with two distinct cations, stabilizing the molecule within the cage; and third, a rarer “carbonate precursor” mechanism, in which one oxygen of CO₂ interacts with a cation while the carbon interacts with an oxygen atom of the framework [25].(Figure 16)

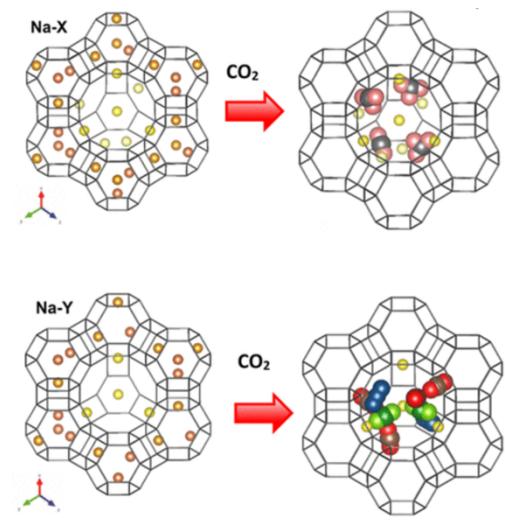


Figure 17: CO₂ adsorption on nanocrystalline Na-X and Na-Y zeolites: comparison before and after capture

In medium- and large-pore zeolites (FAU, X,Y):

CO₂ is adsorbed through electrostatic interactions with the cations (Na⁺, K⁺) located within the zeolite cages. [26]

In small-pore zeolites (CHA): The channels are very narrow, just large enough to allow CO₂ to pass, while the larger N₂ and CH₄ molecules diffuse more slowly.

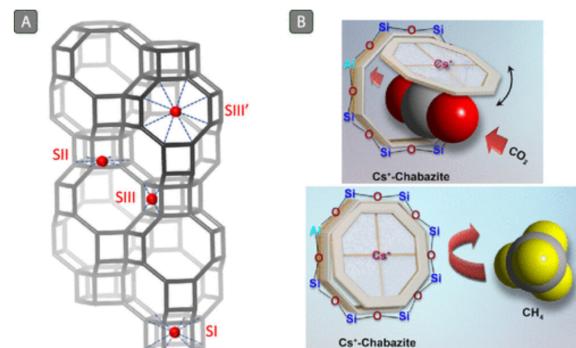


Figure 18: Selective diffusion of CO₂ in small pores

With these different types of adsorptions and small, medium, or large-pore zeolites, several challenges remain, including the use of rare or expensive cations and the optimization of the trade-off between selectivity, capacity, and diffusion rate. Large-scale deployment of these technologies therefore requires a thorough assessment of their impact on resources, human health and ecosystems while also considering economic and sustainability aspects.

Several companies are developing and commercializing zeolites for industrial-scale CO₂ capture. Among them, ZeoDAC, based in Atlanta, USA, offers zeolitic materials optimized for CO₂ selectivity and adsorption capacity. Other players, such as BASF and Honeywell UOP are also working on similar solutions aiming to integrate these materials into industrial gas or biogas treatment processes. These initiatives demonstrate that zeolite based

capture, although less widespread than amine solvents, is beginning to find practical applications in industry particularly for CO₂ separation in complex or highly humid gas streams.

In summary, CO₂ capture using zeolites relies on their unique structural and chemical properties: the size and connectivity of their pores, the nature and distribution of cations, and the electrostatic and kinetic interactions with CO₂. Zeolites offer several clear advantages. They provide high selectivity for CO₂ significant adsorption capacity, good chemical and mechanical stability, and the possibility to fine-tune their performance by controlling crystal size and cation sites. Recent advances in nanocrystal synthesis and cation design are paving the way for more efficient and sustainable industrial processes.

However, some limitations remain. Zeolites are sensitive to moisture, which can significantly reduce their performance in humid gas streams. Some compositions rely on rare or expensive cations, and there is always a trade-off between adsorption capacity, selectivity, and diffusion rates. Large-scale implementation also requires careful consideration of economic, environmental and human health impacts. Despite these challenges, zeolites represent a promising alternative to amine solvents particularly for targeted industrial applications and complex gas streams. They offer real potential for reducing CO₂

emissions and contributing to a more sustainable energy transition.

3. CO₂ Conversion into Polymers from Epoxides

The chemical valorization of carbon dioxide (CO₂) into sustainable materials is a key strategy within Carbon Capture and Utilization (CCU). Among the most promising approaches, the copolymerization of CO₂ with epoxides to synthesize aliphatic polycarbonates stands out.

This process not only sequesters CO₂ in a stable polymer matrix but also provides a pathway to biodegradable and functional materials, thereby reducing our dependence on fossil based feedstocks. The most mature and efficient approach to date for achieving this goal is the Ring-Opening Copolymerization (ROCOP) of CO₂ with epoxides (Figure 19) [27].

This reaction builds a polymer chain by perfectly alternating the two monomers, yielding aliphatic polycarbonates. The process is entirely catalytic. A catalyst, which is a cobalt-salen or zinc-based metal complex activates the epoxide ring allowing nucleophilic attack by a co-catalyst that “initiates the chain” and the subsequent insertion of a CO₂ molecule. This cycle repeats to build the polymer chain [28].

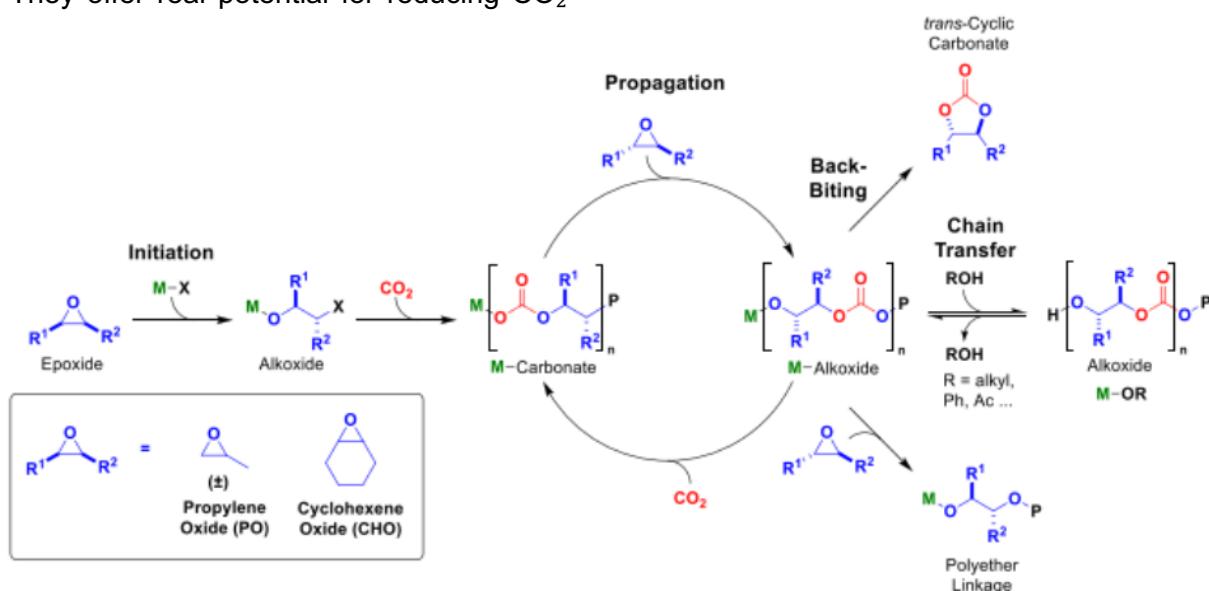


Figure 19: General reaction scheme of the Ring-Opening Copolymerization (ROCOP) of an epoxide with CO₂ to produce an aliphatic polycarbonate.

The major challenge lies in the competition between linear polymer formation and cyclic carbonate formation.

Thermodynamically, the cyclic carbonate is the favored product while the polymer is a kinetic product. This undesired phenomenon is referred to as “back-biting” (Figure 20).

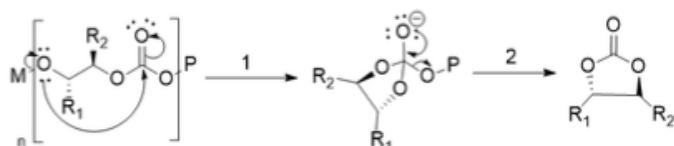


Figure 20: “Back-Biting” Mechanism

In this mechanism, the active end of the growing polymer chain folds back on itself and attacks one of the carbonate bonds already present in the chain. This reaction then breaks the chain and ejects a cyclic carbonate molecule, preventing further polymerization (Figures 19 and 20). This phenomenon therefore limits the length of the resulting polymers and reduces the overall efficiency of the process.

The crucial role of the catalyst is to lower the activation energy for linear propagation so that it proceeds much faster than this back-biting mechanism thereby enabling polymer formation despite the thermodynamic preference for cyclic carbonate. In this mechanism (Figure 20), the polymer chain end plays a key role:

- Pathway 1: The terminal oxygen atom, coordinated to the metal (M), uses its lone pair to perform a nucleophilic attack on the carbon of the carbonyl group (C=O) immediately preceding it in the chain. This attack generates an unstable tetrahedral intermediate, which quickly rearranges, regenerating the C=O double bond.
- Pathway 2: This rearrangement then breaks the C–O bond linking this segment to the rest of the polymer chain. The result is the

formation of a cyclic carbonate, a thermodynamically stable but undesirable by-product, as it interrupts polymer growth.

In industry, the most advanced application of CO₂ valorization is not the production of solid plastics but the manufacture of polycarbonate polyols which are subsequently used to produce polyurethanes.

The choice of epoxide monomer is critical, as it determines the final polymer properties. Two emblematic examples are :

- Propylene oxide (PO), which produces poly (propylene carbonate) (PPC).
- Cyclohexene oxide (CHO), which yields poly (cyclohexene carbonate) (PCHC).

Companies like Covestro, with its Cardyon® line, have already brought polyols to market that incorporate up to 20% CO₂ by mass [29]. These “green” polyols directly replace their petrochemical counterparts in a highly dynamic sector: the manufacture of polyurethane foams used in mattresses, automotive seats, and building insulation.

This application is particularly relevant as it addresses a mass market while delivering measurable environmental benefits. A Life Cycle Assessment (LCA) published in *Green Chemistry* by von der Assen and Bardow showed an 11–19% reduction in carbon footprint compared to the conventional fully fossil-based process [30].

The success of the ROCOP process, which converts epoxides and CO₂ into polycarbonates, depends mainly on the catalyst used. Its efficiency is evaluated by the Turnover Frequency (TOF), which measures how many times an active site can convert reactants into products per unit time. Ideally, the catalyst should be very fast and selective, producing mainly linear polycarbonates while minimizing the formation of undesirable cyclic carbonates.

Early successes were achieved with cobalt catalysts, particularly Co(III)-salen complexes studied by Dairensbourg's team. These catalysts are highly efficient and selective, but cobalt is toxic, expensive, and can color the polymer, complicating purification. More modern Co-salen variants, such as dinuclear complexes or those linked to quaternary ammonium groups, achieve very high reaction rates while maintaining polymer regularity, but safety concerns remain.

To overcome these limitations, researchers are turning to non-toxic metals such as zinc, or bimetallic systems combining zinc and magnesium. These catalysts provide good reaction rates even at low CO₂ pressure and remain highly selective (>99% carbonate linkages). Their operation often relies on cooperation between two metal centers, where the growing chain transfers from one metal to the other, optimizing the reaction. These modern catalysts are particularly suitable for biomedical or food applications due to their safety and lower cost [30][31].

The ROCOP process allows precise control over the final properties of polycarbonates through the choice of epoxide monomer. This structure property relationship is particularly evident when comparing two of the most studied polymers.

Poly (propylene carbonate) (PPC), formed from propylene oxide, is an amorphous and flexible material with a relatively low glass transition temperature ($T_g \approx 40^\circ\text{C}$). It is biodegradable thanks to the ester and carbonate bonds in its backbone, which can be broken by hydrolysis or enzymes. Under industrial composting conditions, it can decompose into carbon dioxide and water. This biodegradability makes it an interesting candidate for sustainable packaging and certain biomedical applications. However, its low T_g means it softens when heated, limiting its mechanical performance. For this reason, PPC is rarely used alone and is often blended

with more rigid bioplastics such as PLA or reinforced with fillers.

Poly (cyclohexene carbonate) (PCHC), derived from cyclohexene oxide, is very different: it is a rigid thermoplastic with a high glass transition temperature ($\approx 120^\circ\text{C}$). It is not biodegradable but offers increased durability and thermal resistance, making it suitable for technical applications such as polystyrene replacement plastics, durable coatings, certain food packaging, or even optical lenses.

This modularity illustrates one of the major advantages of the ROCOP process: by simply choosing the starting epoxide, it is possible to create materials with radically different properties while using CO₂ as a feedstock, combining performance and sustainability [32][33].

To evaluate the global potential of CO₂-based polycarbonates, the entire value chain must be considered. The main challenge lies in the source of the epoxide monomers, such as propylene oxide, which are still almost exclusively produced from fossil resources. This means that the final polymer is only partially sustainable, and this technology cannot be considered fully green as long as its co-reactants are not renewable.

A promising solution is to pair CO₂ with bio-based epoxides. Research is ongoing to produce epoxides from renewable resources such as glycerol (a biodiesel by-product) or terpenes derived from pine. One particularly interesting example is epichlorohydrin, historically produced from petrochemical propylene, which can now be industrially synthesized from renewable glycerol. Copolymerization of this bio-

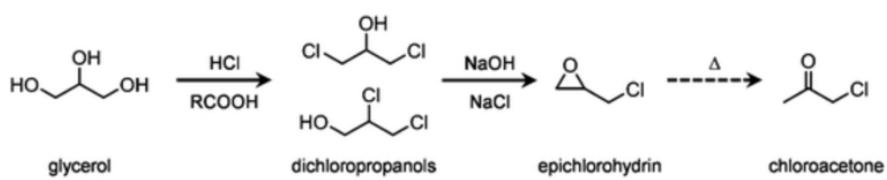


Figure 21: Synthesis of epichlorohydrin from glycerol via hydrochlorination

based epichlorohydrin with CO₂ produces a functional polycarbonate containing lateral chlorine atoms [34] (Figure 21).

These functional groups offer two main advantages: they can induce a certain degree of crystallinity in the material, improving its thermal properties, and they serve as anchoring points for further chemical modifications, paving the way for fully renewable “tailor-made” polymers.

The environmental impact is also positive. The biodegradability of PPC helps reduce plastic pollution. However, the use of catalysts must be carefully managed: if the catalyst contains toxic metals such as cobalt or chromium even trace amounts can migrate from the polymer into the environment or the human body, posing health and safety risks. Since these catalysts are dissolved in the reaction medium, complete removal after production is difficult. This is why research is focusing on non-toxic and biocompatible metals, such as zinc or magnesium, or on heterogeneous catalysts, which can be easily removed by filtration.

In summary, converting CO₂ into polycarbonates through copolymerization with epoxides is now a well established and reliable technology, already used in several industrial applications. It brings several meaningful advantages. First, it offers a smart way to lock CO₂ into useful materials, turning a waste gas into something valuable. One of its biggest strengths is its versatility: simply by changing the epoxide monomer, you can obtain very different types of polymers, from biodegradable and flexible materials like PPC to rigid and heat resistant plastics such as PCHC. Research is also moving toward safer catalysts, especially zinc and magnesium based systems making the process cleaner and more suitable for sensitive applications.

However, the technology also faces important limitations. The main issue is that most epoxides available today are still produced from fossil resources. This means that even if CO₂ is effectively used, the final polymer is not fully sustainable.

There is also the challenge of catalyst residues, especially when using cobalt complexes, which are hard to remove completely and can raise toxicity concerns. Finally, polymers like PPC, although environmentally attractive, have limited thermal and mechanical performance, which often requires blending or reinforcement to broaden their applications.

Overall, this technology holds strong potential, but its true environmental benefit will depend on our ability to develop fully bio-based epoxides. Promising candidates are already emerging, such as epoxides derived from glycerol or natural terpenes, suggesting that a truly circular value chain is within reach. If these advances continue, ROCOP could become a major route for producing efficient, low-carbon materials directly from CO₂.

4) Electrochemical Reduction of CO₂ into High-Value Chemicals

Electrochemical reduction of carbon dioxide (CO₂) has emerged as a promising strategy for converting this greenhouse gas into valuable chemicals while supporting long term carbon-neutrality goals. Among the possible products, methanol stands out because of its dual role: it is both a high-energy-density liquid fuel and a key building block in the chemical industry, used to produce formaldehyde, acetic acid, a wide range of plastics, and many common solvents. [35]

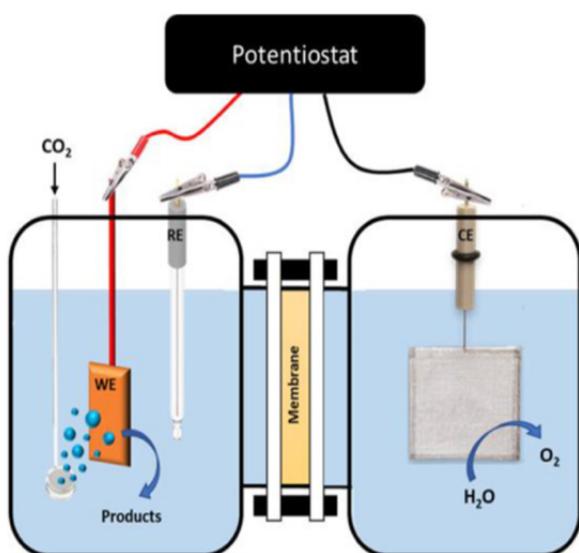
The process relies on an electrochemical cell in which CO₂ is reduced at the cathode through the supply of electrons and protons, while water is oxidized at the anode. The number of electrons transferred to each CO₂ molecule determines the final product: carbon monoxide and formic acid require two electrons, methanol six, methane eight, ethylene and ethanol twelve, and propanol

eighteen. Because of this broad spectrum of possible products, carefully controlling the operating conditions and selecting the right catalyst are essential to steer the reaction toward methanol with high selectivity. [36]

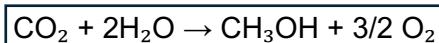
Methanol occupies a central position at the crossroads of chemistry and energy. It can serve as a cleaner fuel while also acting as a versatile feedstock for producing a wide range of industrial chemicals. If produced directly from CO₂, methanol would provide a meaningful way to recycle carbon that has already been emitted, turning a problematic greenhouse gas into a dense, easily transportable energy carrier that fits well within existing industrial infrastructures.

In this sense, CO₂-derived methanol represents not just a scientific opportunity but a practical route toward more sustainable energy and chemical systems. The most commonly used experimental setups to study CO₂ electroreduction in the laboratory are H-type cells (Figure 22).

These systems consist of two separate compartments divided by an ion exchange membrane, which allows the anodic and cathodic reactions to occur independently. In this setup, CO₂ is typically introduced into the electrolyte as fine bubbles,



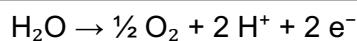
maximizing its dissolution and interaction with the catalytic surface. At the cathode, CO₂ is reduced, while water is oxidized at the anode, leading overall to the formation of methanol and oxygen according to the reaction:



While H-cells offer precise control over electrode potentials and allow the detailed study of electrode selectivity, they suffer from major limitations, particularly in terms of CO₂ mass transfer and achievable current density, which restricts their use mainly to fundamental research. [37]

To overcome these limitations, researchers have developed gas-fed flow electrolyzers. In these systems, CO₂ is supplied directly as a gas to the cathode, which is in contact with a continuously circulating electrolyte and separated from the anode by an anion-exchange membrane. This design ensures optimal interaction between the gas, electrolyte, and the catalytic electrode surface, significantly enhancing reaction kinetics and overall CO₂ conversion.

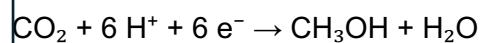
A major advantage of gas-fed flow electrolyzers is the ability to finely tune operational parameters. Gas flow rate, temperature, and electrolyte composition can be adjusted to maximize methanol production while minimizing undesired side reactions. At the anode, water is oxidized to release protons and electrons according to:



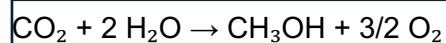
These electrons then travel through the external circuit to the cathode, where they participate in the reduction of CO₂ into

Figure 22: Schematic representation of the electrochemical H-cell.

methanol, simultaneously capturing protons from the electrolyte:



The overall reaction can thus be summarized as:



Compared to traditional H-cells, these continuous flow systems offer several important advantages. They increase current density and improve CO₂ mass transfer, while also providing better control over product selectivity, particularly toward methanol. This makes gas-fed flow electrolyzers highly attractive for scaling up, where precise reaction control is essential for both efficiency and economic viability.

Overall, the development of gas-fed flow electrolyzers represents a significant step toward the industrial valorization of CO₂. By improving the efficiency of electrochemical reduction and enabling

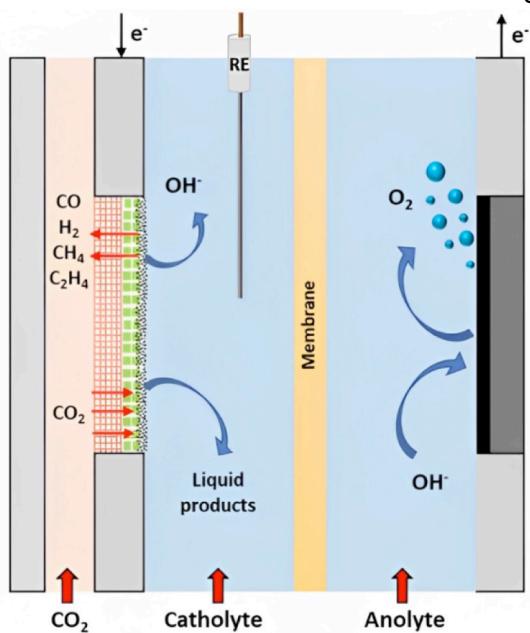


Figure 23: Schematic of a Gas-Fed Flow Cell Electrolyzer

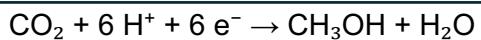
continuous methanol production, these systems bring the concept of CO₂ recycling closer to industrial implementation, while offering promising opportunities for integration with renewable energy sources. The ability to operate continuously and with greater control over selectivity highlights their potential as a cornerstone technology in sustainable chemical production.

The gas-fed flow cell electrolyzer is a technology specifically developed to overcome the limitations of traditional electrochemical systems and to improve the efficiency of CO₂ conversion into higher-value products, particularly methanol. In this configuration, CO₂ is supplied directly in its gaseous form to the cathode, where the reduction reaction takes place. By bringing the gas into direct contact with both the electrolyte and the catalytic surface of the electrode, the system significantly enhances reaction kinetics and allows for a much higher conversion rate compared with conventional setups. This design essentially reduces the constraints associated with CO₂ solubility in aqueous media, which is one of the main bottlenecks in H-cell configurations. [38]

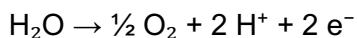
The cell itself consists of two electrodes separated by an anion-exchange membrane (AEM). This membrane plays a crucial role: it allows hydroxide ions (OH⁻) to migrate between compartments to maintain charge balance, while at the same time preventing the mixing of the reaction products formed at each electrode. A liquid electrolyte typically alkaline, such as KOH or KHCO₃ is continuously circulated through the system. This continuous flow not only ensures efficient mass transport but also helps

remove gaseous by-products, keeping the reaction environment stable and improving overall performance.

At the cathode, CO₂ undergoes electrochemical reduction to produce methanol according to:



Meanwhile, at the anode, water is oxidized to generate oxygen, protons, and electrons:



Operating the system in continuous-flow mode offers several advantages. It allows precise control over operating parameters such as CO₂ flow rate, temperature, and applied potential factors that strongly influence catalytic activity and product selectivity. By improving mass transport and optimizing the interaction between the electrode and the electrolyte, gas-fed flow cells provide a far more efficient environment for CO₂ conversion than static cells.

Overall, this configuration is increasingly viewed as one of the most promising pathways toward industrial-scale CO₂ valorization. Its ability to operate continuously, achieve higher current densities, and maintain better selectivity toward methanol makes it especially attractive for future integration with renewable energy systems and carbon-capture technologies.

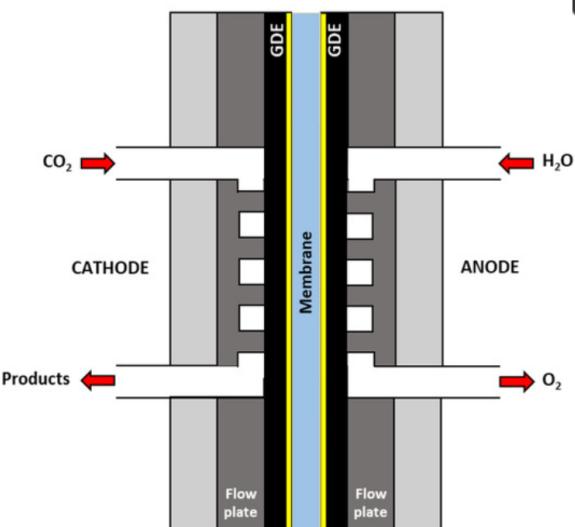


Figure 24 : Schematic representation of a polymer electrolyte membrane (PEM) electrolyzer

The polymer electrolyte membrane electrolyzer, known as PEM (Proton Exchange Membrane), relies on a solid membrane that conducts protons and serves as the electrolyte. Originally developed for hydrogen production, this technology has since been adapted for the conversion of CO₂ into methanol. Its operation is based on two coupled half-reactions: at the anode, water is oxidized to produce oxygen, protons, and electrons, while the electrons travel through the external circuit toward the cathode. The protons simultaneously migrate through the membrane to the cathode, where dissolved CO₂ is reduced to methanol. The membrane ensures selective ion conduction while preventing the mixing of gaseous products, enabling relatively pure methanol production and operation at low temperatures. [39]

Despite these advantages, the development of PEM electrolyzers for CO₂ reduction remains limited by several challenges. Competition with hydrogen evolution, electrode durability, and the chemical stability of the membrane under reductive conditions are among the main

obstacles. Nevertheless, this technology shows strong promise thanks to its high energy efficiency and compatibility with renewable energy sources.

The catalyst deposited on the cathode plays a central role in the reaction. It is responsible for activating CO₂ by facilitating the breaking of the C=O bond and stabilizing the intermediates formed during the successive hydrogenation steps. The choice of catalyst, its composition, and its nanostructure directly influence the activity, selectivity, and overall durability of the process.

Nanostructured copper is the most extensively studied material for this application [40]. Its unique ability to promote multiple hydrogenation steps allows it to generate a variety of products, including methanol. It provides efficient CO₂ adsorption and facilitates the formation of key intermediates such as CO*, HCO*, and CHO*. Its high electrical conductivity ensures optimal electron transfer between the cathode and the active sites, improving overall process efficiency. However, copper also presents limitations: it can produce a mixture of competing products, such as carbon monoxide and hydrocarbons and its nanoparticles tend to oxidize or aggregate, reducing their active surface area and effectiveness.

To overcome these limitations, zinc often introduced as zinc oxide is incorporated as a promoter in copper-based catalysts. ZnO enhances CO₂ adsorption, stabilizes oxygenated intermediates, and improves the dispersion of copper particles, preventing their agglomeration. The Cu–Zn combination creates an effective synergy: copper catalyzes the hydrogenation steps, while zinc stabilizes oxygen-containing intermediates, improving methanol selectivity. However, ZnO has limited electrical conductivity, and its performance

strongly depends on its chemical state: inappropriate reduction or oxidation can negatively affect both activity and selectivity.

Recent studies confirm that the Cu–Zn interaction is crucial for achieving an optimal balance between activity, selectivity, and stability factors essential for developing industrially viable and environmentally sustainable electrocatalytic processes. Thus, the role of the catalyst extends far beyond providing active sites, it also controls the formation of intermediates and directly influences the electrochemical behavior of the cathode. The Cu–Zn system remains one of the most promising candidates for the efficient and selective electrochemical reduction of CO₂ to methanol. [41]

After discussing the electrochemical approaches for converting CO₂ into methanol, it is important to highlight another, more mature pathway that is already widely used at an industrial scale: the catalytic hydrogenation of CO₂. This method is directly inspired by the industrial production of methanol from carbon monoxide and hydrogen (CO + H₂ → CH₃OH), a process employed for decades. In this reaction, CO₂ serves as the carbon source, while hydrogen ideally produced from renewable resources enables methanol formation through thermocatalytic conversion.

This thermocatalytic approach perfectly complements electrochemical methods: it is robust, well-suited for large-scale industrial production, and can integrate seamlessly into existing infrastructure. By combining this approach with electrolysis or direct CO₂ capture, it becomes possible to design a sustainable carbon cycle that links energy efficiency with circular chemistry. This method is particularly compelling because it integrates smoothly into existing industrial infrastructures. While electrochemical reduction offers modularity

and high selectivity at small to medium scale, catalytic hydrogenation provides high throughput, robustness and continuity, making it suitable for large-scale CO₂ utilization. In this sense, the two technologies complement each other.

The reaction relies heavily on catalysts based on Cu/ZnO/Al₂O₃, the same system used in classical methanol production. Over decades of research, the remarkable synergy between these three components has been clarified: copper serves as the primary active phase for CO₂ activation and hydrogenation, ZnO stabilizes key oxygenated intermediates and Al₂O₃ provides structural support and prevents deactivation. Numerous studies have confirmed this cooperative effect, including work by Olah and al. [42] who demonstrated that catalysts engineered to expose a high density of Cu–ZnO interfacial sites show significantly higher methanol selectivity.

More advanced catalyst designs expand this concept. For instance, the addition of spinel phases such as ZnGa₂O₄ has been shown to enhance CO₂ adsorption and hydrogenation kinetics, as reported by Wang and al. (2025) [43]. Other research avenues explore single-atom copper catalysts, where isolated Cu centers embedded within oxide matrices create highly tailored microenvironments. Such sites can stabilize intermediates formate (HCOO^{*}), formaldehyde-like species (H₂CO^{*}) and methoxy (CH₃O^{*}) that define the methanol synthesis pathway. This was elegantly demonstrated in the work of Jiang and al. (2024) [44], who showed how Na⁺ ions decorating single Cu atoms tune the local environment to favor methanol production.

Mechanistic studies converge on a hydrogenation sequence involving formate species as key intermediates. CO₂ first adsorbs at Cu–ZnO interfaces, then undergoes stepwise hydrogenation through formate and formaldehyde intermediates before forming methanol. *In situ* spectroscopic investigations particularly FTIR are crucial for probing these

intermediates under real reaction conditions. An insightful study employing Cu–ZnO/Al₂O₃ nanorods confirmed the presence of surface-bound formate species and revealed dynamic restructuring of Cu–ZnO active sites during the reaction (*In situ* FTIR study on Cu–ZnO/Al₂O₃).

The broader significance of this reaction is captured in the “methanol economy” concept proposed by Olah, Goeppert, and Prakash (2009) [45]. In their vision, CO₂ is no longer a waste product but a recyclable carbon resource. The hydrogenation of CO₂ to methanol becomes a central pillar of a circular carbon system in which CO₂ captured from industrial emissions, biomass or even air is continuously transformed into synthetic fuels and chemicals. This framework has shaped much of the modern research and policy interest surrounding CO₂ utilization.

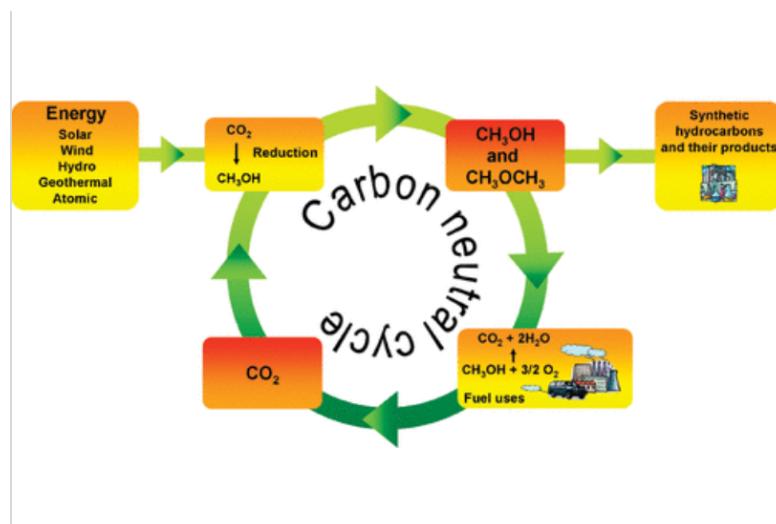


Figure 25: Schematic representation of the “Carbon Methanol Cycle” proposed by Olah, Goeppert & Prakash

Industrial applications of CO₂ hydrogenation are already emerging. Optimized Cu/ZnO/Al₂O₃ catalysts can achieve high methanol selectivity at moderate temperatures (around 200–260 °C) and pressures (50–70 bar), conditions compatible with renewable hydrogen production systems. Economic and environmental analyses confirm the potential feasibility of large-scale

deployment, especially when integrated with low-carbon hydrogen sources (*Fuel*, 2023). [46]

The valorization of CO₂ into methanol is today a promising pathway to transform this greenhouse gas into a useful product while supporting the energy transition. Two main approaches exist: electrochemistry and thermochemical catalysis. Electrochemistry directly reduces CO₂ into methanol in electrochemical cells using Cu–Zn catalysts and PEM membranes or gas-fed flow cells. This technique allows precise control over reaction conditions and good selectivity toward methanol. It also offers interesting modularity for small- to medium-scale installations. However, it remains limited by electrode durability, competition with hydrogen evolution, and energy efficiency, which still hinders its industrial scale-up. Nevertheless, companies such as Mitsubishi Gas Chemical and JFE Steel Corporation in Japan have recently launched pilot projects using captured CO₂ to produce methanol, demonstrating that this technology is beginning to move beyond the academic stage. [47]

Thermochemical catalysis, more mature, is directly inspired by industrial methanol production from CO and H₂ and uses hydrogen to convert CO₂ into methanol via Cu/ZnO/Al₂O₃ catalysts. This approach is robust, continuous, and compatible with existing infrastructure. It enables high throughput and large-scale production and can integrate hydrogen produced from renewable sources. Key intermediates, such as formate, formaldehyde, and methoxy, are stabilized by the catalysts, increasing selectivity and efficiency. Among the companies active in this field, Carbon Recycling International (Iceland) [48] and Haldor Topsoe (Danemark) [49] have developed industrial processes demonstrating the economic and environmental feasibility of this approach. The drawbacks include relatively high pressure and temperature conditions and the need for careful hydrogen management but these constraints are

well controlled thanks to several decades of industrial practice.

By combining these two approaches, it becomes possible to envision a sustainable carbon cycle. Electrochemistry provides flexibility and modularity, while thermochemical catalysis ensures robustness and continuous production. Together, they transform CO₂ into methanol, a transportable energy carrier and chemical feedstock contributing to circular chemistry and emission reduction.

In summary, methanol production from CO₂ perfectly illustrates how complementary solutions can be implemented to turn an environmental problem into an energy and chemical opportunity. Recent advances, supported by pioneering companies and catalytic innovations, show that this technology can become a central pillar of industrial carbon management strategies.

III. CONCLUSION and PROSPECTS

The issue of CO₂ capture and utilization goes far beyond a scientific topic. While working on it, we realized that behind every process, every catalyst, and every material lies a much deeper question: what kind of future do we want? The technologies we explored amine solvents, zeolites, CO₂-derived polycarbonates, and methanol synthesis show that chemistry can genuinely be part of the solution. They prove that a gas often seen as waste can actually become a valuable resource. But as we looked deeper, we also understood that none of these solutions is perfect.

Amine-based solvents are efficient, yet their regeneration requires a significant amount of energy. Zeolites are elegant and highly selective but lose efficiency when exposed to humidity. Polycarbonates made from CO₂ are an impressive step toward sustainable materials, but they still depend heavily on epoxides derived from fossil resources. And even CO₂ to methanol conversion, one of the most promising routes, can only be truly green if the hydrogen used comes entirely from renewable energy.

Through this work, we realized that these limitations should not discourage us; they should guide us. They highlight the areas where our generation will need to innovate and push further. Because we are the ones who will live with the consequences. Because the choices made today will shape our future environments, our jobs, and the health of our planet.

That is why this topic resonates so strongly with us as students. We are not just observers or technicians; we are future scientists, citizens, and decision-makers. We have the responsibility to question the technologies we develop, to educate others, and to advocate for solutions that make sense not only technically, but also ethically and environmentally. Science should not only serve industry; it should also serve society.

Fortunately, the future is rich with possibilities: electrochemical CO₂ reduction, artificial photosynthesis, bio-inspired catalysis and fully bio-based materials. These emerging directions show that a low-carbon future is not only possible but achievable. They point toward a chemistry that does not simply fix problems but rethinks how we produce and consume.

In conclusion, CO₂ capture and valorization are not miracle solutions; they are one piece of a much broader strategy. A strategy that aims not only to reduce emissions but also to rethink our relationship with energy, materials and innovation. As young scientists and citizens, we carry both the responsibility and the opportunity to help build a transition that is truly sustainable, fair and forward-looking.

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Abstract

In the context of the climate emergency, CO₂ capture and utilization have become essential levers to reduce the global carbon footprint. Amines-based absorption technologies remain the industrial standard due to their efficiency, but their high energy demands encourage the development of more sustainable and innovative solvents. Meanwhile, zeolites are emerging as highly selective materials capable of capturing CO₂ effectively under varying conditions. Converting captured CO₂ into valuable products, such as methanol through catalytic hydrogenation or polycarbonates via copolymerization, transforms a greenhouse gas into a strategic resource. These pathways provide complementary technological solutions that integrate capture, storage, and conversion. This approach links environmental performance with scientific and industrial innovation. It demonstrates how combining advanced materials and catalytic processes can address climate challenges while creating economic value.

Keywords:

CO₂ • Capture • Amines • Zeolites • Hydrogenation • Methanol • Polymers • Green Chemistry • Energy Transition