

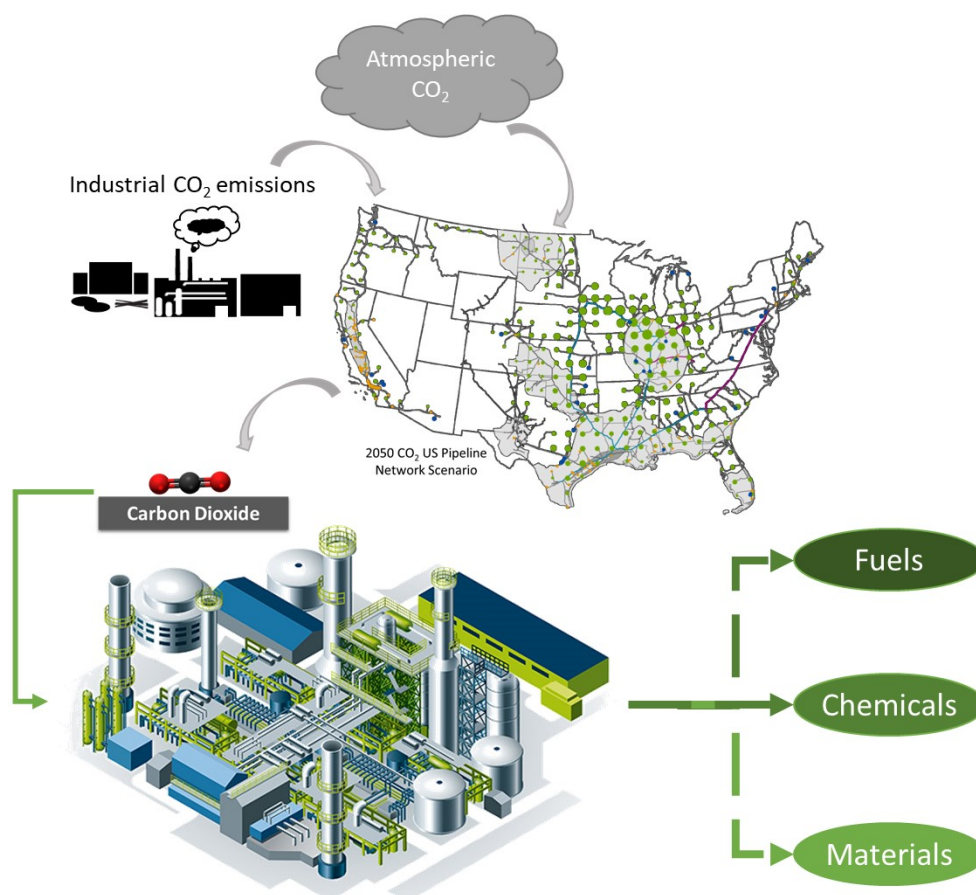
A Carbon Dioxide Refinery: The Core of a Sustainable Carbon-based Circular Economy



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Review

A Carbon Dioxide Refinery: The Core of a Sustainable Carbon-based Circular Economy

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Abstract The atmospheric carbon dioxide (CO₂) accumulation (2–2.5 ppmv/year) is the result of the enormous gap between its emissions (37 Gton/year) and its capture, storage, and utilization (<500 Mton/year). Climate has been dramatically affected due to the failure of natural sinks, in working effectively. To address this Gton-scale gap, numerous uses and applications are needed particularly, those consuming vast volumes of this compound and/or rendering longevous products or long lifecycle services. Thus, carbon utilization (CU) can be seen as the step to close the carbon cycle. Among CU, R&D on CO₂ chemical conversion has proposed a variety of processes, with different degrees of developmental maturity. These chemical process technologies could be efficiently and effectively integrated into refineries to upgrade emitted CO₂. A technology pipeline consisting of a database of these processes and the technology market status should be defined based on published scientific results and patents. Then, an innovative top-down methodology is proposed to eco-design configurations of that refinery, to warrant a sustainable carbon cycle (in terms of energy, environment, and economy) and to change the ways of producing fuels, chemicals, and materials. Additionally, the proposed methodology could be used to identify research and development gaps and needs, for orienting science and technology investments and measures. Hopefully, sustainable CO₂ refineries will be implemented to close the carbon cycle of a circular C-based economy and underpin a decarbonized chemical industry.

Keywords CO₂; CO₂ utilization; CO₂ conversion; decarbonization; CO₂ upgrading; negative emission technologies

1. Background

Carbon dioxide (CO₂) is one of the carbon-containing compounds within the greenhouse gases (GHGs) whose accumulation in the atmosphere has been held responsible for climate change. This rate of accumulation has been increasing with time, motivating 195 countries belonging to the United Nations (UN), to rule for limiting global warming not to exceed 1.5 °C by 2050, during the 26th UN Climate Conference (COP26), known as the Glasgow Climate Pact [1]. By the end of 2022, CO₂ global emissions were reported to be 37 GtonCO₂/year, reaching an atmospheric concentration of 417 ± 0.1 parts per million (ppm) [2].

The three to four orders of magnitude difference in scale of CO₂ emissions (Gton/year) [2] and its utilization (Mton/year) [3,4] calls for vast increases in the number of uses, applications, and/or conversion processes, for this compound. More particularly, processes employing or consuming massive CO₂ volumes, involving lower intensity in carbon (C) and in energy, and rendering products or services with longer longevity or lifecycle are desirable. In general, the reduction of C-intensity has been focused mainly on the mitigation of CO₂ emissions. Meanwhile, C-valorization has been gaining track as a revenue generation approach. In this case, CO₂ emissions are considered as an unlimited and renewable C-feedstock [5–7]. The R&D on CO₂-based chemistry has attained a well-advanced status to propose numerous routes of conversion pathways [8] that would lead to an improvement in the Earth's C-balance. In return, an improved C-balance will contribute to mitigating climate change effects [9]. Consequently, closing the carbon cycle by combining operations of carbon capture (CC), and storing (CS) with CU provides a platform for a carbon-based circular economy (CCE). However, only under strict conservation practices through careful management of the carbon-bearing species, compounds, and materials, sustainability might be warranted. Additionally, since utilization through C-valorization has been recognized for its significant contribution to the economic feasibility of a CCE [10], the implementation of chemical process technologies will require efficiency and effective measures.

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Highlights of Science

CO₂ emissions in the United States (US) are made readily available, by the US Environmental Protection Agency (EPA) together with other GHGs [11]. For this reason, the US data will be used here to exemplify¹ the vast proportion of available CO₂ feedstock. Thus, the map shown in Figure 1 shows the single-point static CO₂ emissions in the US and confirms the magnitude of the feedstock availability.

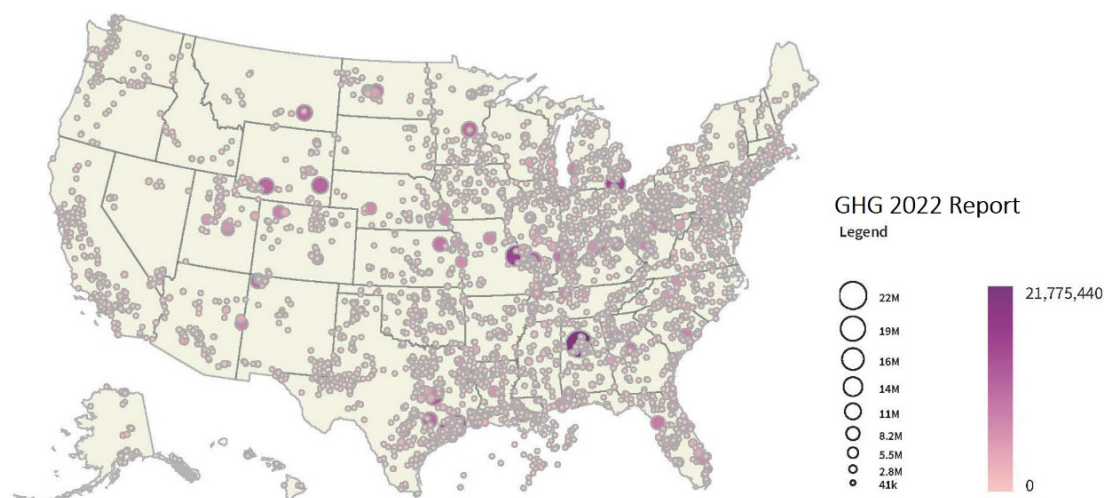


Figure 1. Carbon dioxide emissions from the US operating facilities (tonCO₂) [11].

In 2022, the total CO₂ emissions from these facilities (7586 facilities reported GHG emissions to EPA, from which 6842 reported CO₂ emissions) were nearly 2.5 GtonCO₂/year [11]. Emissions by state ranging from 13 ktonCO₂/year to nearly 350 MtonCO₂/year are shown in Figure 2. Emissions from more than 35 states exceed 20 MtonCO₂/year, as shown in Figure 2. The current strategy for industrial decarbonization is based on the belief that the composition of the emitted stream differs from one facility to another and requires either a tailor-made design solution or at least, adaptation measures of existing technologies. This atomistic view of one decarbonizing technological solution per emitting source (one-to-one strategy) not only results in high costs but also gives rise to difficulties for the implementation of any holistic criterion for carbon management. Therefore, this one-to-one carbon capture, utilization and storage (CCUS) decarbonization strategy will require nearly 7000 CCUS technologies, for the US facilities. In 2022, Texas reported to EPA GHG emissions from 853 facilities, out of which 770 emitted CO₂ amounting to 366 MtonCO₂/year. Thus, the number of technological solutions to mitigate the CO₂ problem, for the largest emitting US state, Texas (see Figures 1 and 2) is 770, if the status quo of the decarbonization one-to-one strategy is applied.

In the case of utilization, the emitted volumes as well as their composition derive from mismatches between the emission source and the use or application. Three mismatches can be identified: logistic, composition, and concentration. The logistic mismatch locates the CO₂ sources at a long (uneconomical) distance from the using location. Meanwhile, the composition and concentration of the source set a mismatch in the presence of undesirable compounds or a too-low C-content in the emitted stream from those suitable for the expected use or application. Additionally, it might also be that the dynamics of the emissions scale differ from that required by the use or by the product market size. The slowness of technology development and implementation could be associated by connecting these mismatches with the atomistic view of the one-to-one decarbonizing strategy. In solving their emission problem, larger facilities with MtonCO₂/year-scale of CO₂ emissions could benefit from both the economy of scale and the advantages of CU valorizing technologies. Instead, under these circumstances, smaller facilities with ktonCO₂/year-scale of CO₂ emissions will incur costs no longer competitive with those of a larger facility, to address decarbonization.

¹ Recurrently along this work, US data and information will be used to exemplify specific cases and situations and to illustrate particularities, facilitate discussion and/or derive potential implications, which may become valid globally. However, regardless of these regional examples, one needs to keep in mind that emissions are a global problem, technologies are globally applied and feedstocks and products are globally commercialized.

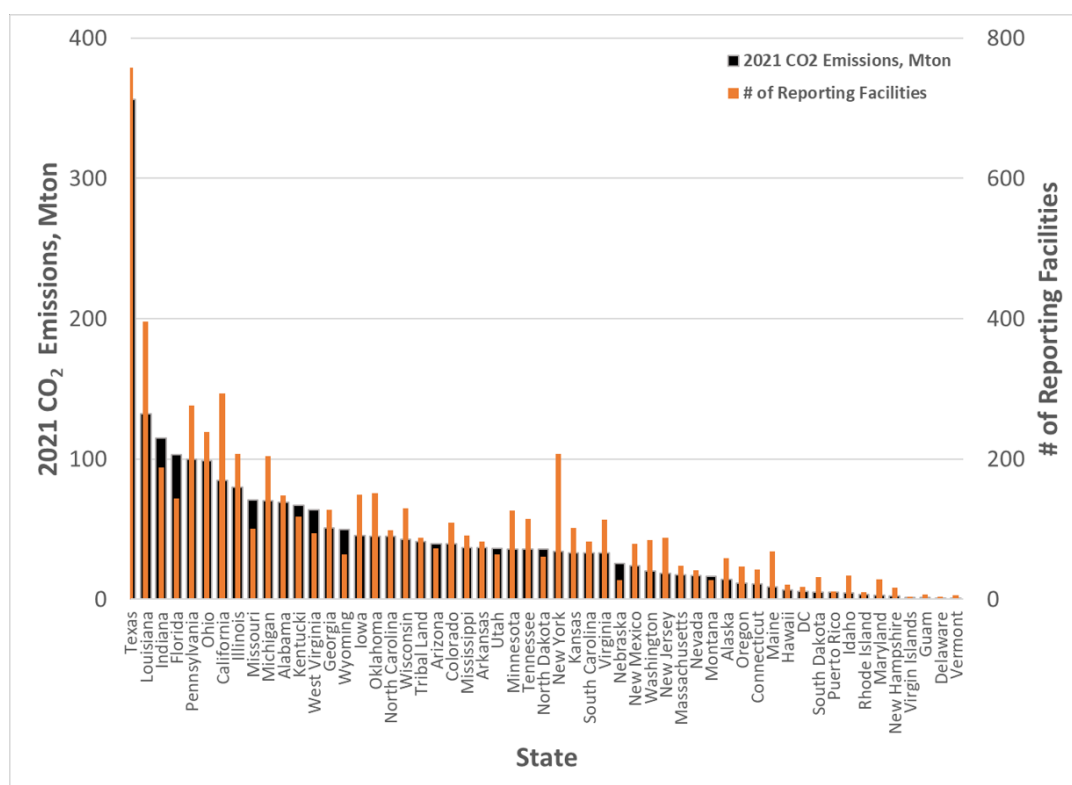


Figure 2. CO₂ emissions by state (from data reported in [11]).

Regarding CC, there are several commercially available technologies while only a few technologies are commercially available, for using and/or utilizing CO₂ (CU). Current industrial use of CO₂ demands about 2000 Mton/year [12], which represents consumption of about 5% of the yearly global emissions [13]. Historically, the most extensive use of CO₂ is in enhanced oil recovery (EOR) [14], in which it would remain geologically stored. Therefore, this is a combined utilization and storing technology, with an upstream capture system (CCUS). Meanwhile, C-valorization remains at the lowest implementation rate, partially because of the high costs associated with transforming CO₂ into economically valuable molecules [15]. The costs involved in the chemical conversion of CO₂ are associated with its high thermodynamic stability ($\Delta G^\circ_f = -396$ kJ/mol), responsible for its low reactivity. Thus, the CO₂ thermodynamic stability leads to chemical processes demanding high temperatures and/or heat fluxes for its conversion and then, resulting in high energy intensity. In order to address this energy intensity issue, energy efficiency and effectiveness have been focused. Catalysis [16] together with process intensification [17] and the use of low-carbon energy sources play important roles in this search for more efficient, effective, and intensified process technologies. On one side, the development of better catalysts needs to be based on the fundamental knowledge of reaction pathways and mechanisms, as well as on the overcoming of identified technical barriers and limitations [18].

This work revises relevant literature to support a proposal for a methodology to eco-design configurations of a CO₂ refinery, to warrant a sustainable carbon cycle (in terms of energy, environment, and economy) and to change the ways for producing fuels, chemicals, and materials. That configuration of the CO₂ refinery efficiently and effectively integrates chemical process technologies to upgrade emitted CO₂. Since the developmental stage and maturity of these chemical processes differ from each other, a technology pipeline consisting of a database of these processes and the technology market status needs to be defined based on published scientific results and patents. The lessons learned from the historical development or configuration design of oil refineries and biomass refineries (biorefineries) were also incorporated in the conceptualization of the methodology.

2. Technology Integration towards Carbon Dioxide Valorization

The valorization of emitted CO₂ would require its capture before its subsequent utilization, which for the present work will be focused on its conversion. Clearly then, CC plays a vital role

in whether the carbon gets sequestered or converted into value-added products. However, according to the Intergovernmental Panel on Climate Change (IPCC), CCS will not be enough to achieve the net zero goals and additional components should be added to the strategy, such as reduction of energy demand, improvements in energy efficiency, and deploying of innovative conversion processes [19]. Although IPCC [19,20] and IEA [21] projections studies showed different trends in the carbon and energy intensities, their models and scenarios assume a predominant role in efficiency improvements, demand reduction measures, electrification, and increased CCS deployment. For emissions reduction purposes, the relative importance of the recommended approaches differs in their respective predictions on the deployment of CCS, in the level of fuel switching and efficiency improvements, and the effect of structural and behavioral changes. Nevertheless, some emissions have been recognized as being difficult to cost-effectively avoid, such as from the aviation sector; long-distance freight by ships; industrial process emissions (e.g., cement, steel, refining, etc.); high-temperature heat (e.g., $>1000\text{ }^{\circ}\text{C}$); etc. [22]. Although technological solutions exist for these cases, these technologies are unsustainable, absurdly expensive, or have not reached commercial scale [23]. In this regard then, the well-accepted role of CCS in decarbonization can be complemented with CU to valorize CO_2 , create revenues, and balance economic sustainability. Additionally, CU could be seen as the integrating vehicle that could close the C-loop in the anthropogenic C-cycle, as represented in Figure 3, even though sustainability indicators and metrics must be kept in mind permanently. Probably, closing the carbon cycle might be the most challenging aspect of establishing a CCE. However, circularity per se does not warrant sustainability: other measures need to be put in place, together with tools and methods for assessing, monitoring, and measuring the implementation of the circularity strategy [24].

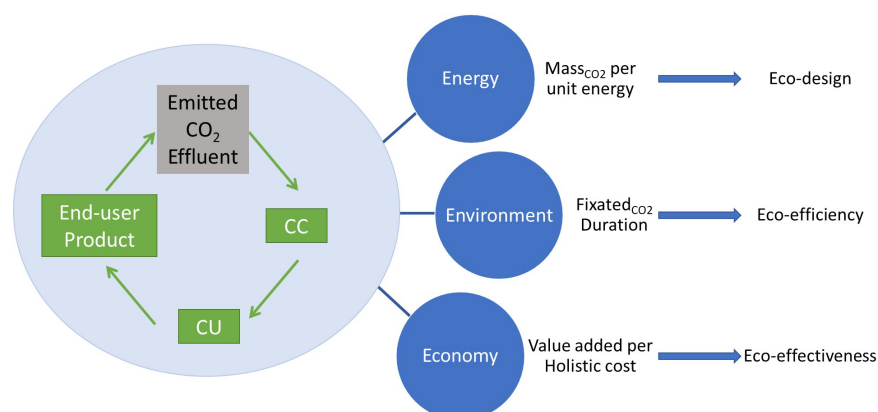


Figure 3. CU as a C-loop closing step and sustainability metrics (adapted from [24]).

The 17 Sustainable Development Goals (SDGs) of the 2030 Agenda were defined by the UN, in 2015 [25]. The four pillars of sustainability namely, human, economy, environment, and social were articulated to achieve a sustainable future in which human development is promoted. For this work, two of these sustainability pillars will be considered together with energy for its prevalent role in development and its impact on the environment (see Figure 3). Thus, the three considered pillars regard energy consumption, environmental impact, and economic feasibility. Figure 3 also includes some metric indicators to monitor and assess sustainability, for those three pillars. Each of these indicators can be associated with the concepts listed on their right namely, eco-design, eco-efficiency, and eco-effectiveness [26]. Eco-design is defined as the use and application of methods, tools, processes, and/or measures to reduce resource consumption and waste generation. Eco-efficiency is the minimization of volume, flow rate, and toxicity of materials flowing through the systems, per unit energy spent/consumed. Eco-effectiveness is the transformation of products and their associated material flows without impact on ecological systems or economic growth. In summary, these concepts represent the principles framework to support sustainability. These principles include (i) resource optimization, (ii) efficiency optimization, (iii) effectiveness optimization, and (iv) waste prevention. Regarding (i) and (iv), maximizing the volume of CO_2 consumed and minimizing its emissions call for the use of low-carbon energy sources and the production of long-lasting products (not fuels). In the case of CO_2 , new eco-efficient chemical processes enabling its valorization are needed, making CCE an urgent essential that

cannot be prolonged into a future prospect. The total substitution of today's linear “take, make, dispose” economy, by eco-efficient/eco-effective closed-loop approaches to production processes has to be emphasized [10].

The 2018 energy demand of more than 175 EJ by the industrial sector led to 30% of the global CO₂ emissions (~11 Gton) [27]. Approximately 60% of this industrial energy demand comes from energy-intensive, C-intensive, or both industries, such as steel, cement, chemicals, oil and gas, etc. [28]. Their share of direct CO₂ emissions is even higher, close to 70% [29]. By 2023, according to the International Energy Agency (IEA), the situation has worsened to make industry the most energy-consuming and CO₂-emitting end-use sector. It accounts for 38% of total final energy consumption and 47% of CO₂ global emissions [30]. Industrial heat and steam generation are the main responsible for this level of emissions since energy is provided from the combustion of fossil fuels [31]. The vast majority of the industrial emissions correspond to streams with CO₂ concentration lower than 15%, and only a few industries (<2%) emit streams that exceed 95% of CO₂ concentration [32]. In summary, the industrial and energy sectors are the main ones responsible for CO₂ emissions, which could be an unlimited source of carbon-based feedstock. However, the industry seems to believe that the individual factors defining the emissions are responsible for the above-described mismatches of the CO₂ resources. The magnitude of the emitted volumes, their CO₂ concentration, and even their transportation feasibility are defined by different factors, such as the origin of fossil sources of electricity and of fossil-derived heat, the high-temperature heating requirements, energy inefficiencies, energy intensity or high consumption, fossil or carbon-containing raw materials, inefficient and ineffective material flow, waste generation and/or disposal, etc. On the other hand, most of the published literature on CO₂ conversion does not consider real feedstocks (emitted streams), nor surrogate mixtures of the real feeds, or even the effect of potential contaminants present in these feeds. If during development and scaling up these processes do not fill these gaps, then, for demonstration or implementation purposes, any of them would need to be considered in conjunction with either a recovery process that yields a concentrated CO₂ stream or a pretreatment step that removes undesired contaminants.

Valorizing technologies to convert massive amounts of CO₂ into value-added products are preferred for the already mentioned benefits: (i) the large consumption of this noxious compound, (ii) being retained in a long lifecycle material, and/or (iii) the economy of scale. In addition to CO₂ consumption, the energy intensity of the current process-producing technology should be considered as well since most processes for CO₂ conversion are typically energy-intensive [33]. In this regard, The U.S. Department of Energy (DOE)'s Advanced Manufacturing Office (AMO) published an analysis study reporting the energy intensity and the domestic production of 74 individual chemicals [34]. Then, based on different rationalized reasons, 10 of those chemicals were excluded. A summary is presented in Table 1 to show the top 20 chemicals that can be directly or indirectly produced or derived from CO₂. Thus, Table 1A lists these chemicals in decreasing order of energy intensity while Table 1B lists those in decreasing order of production.

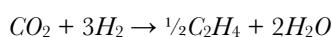
Table 1. Top 20 chemicals with higher (A) energy intensity and (B) production capacity in the United States (data from [34]).

A			B		
Chemical	Energy Intensity (kJ/kg)	Production (ton/y)	Chemical	Energy Intensity (kJ/kg)	Production (ton/y)
Benzene	18289	6	Ethanol	10799	30
Ethylene	16436	24	Ethylene	16436	24
Polycarbonate	15590	1	Propylene	3140	14
Ammonia	13591	10	Ammonia	13591	10
Methanol	11392	1	Polypropylene	1432	8
Ethanol	10799	30	Polyethylene, HD	2410	8
Carbon Black	8937	2	Polyethylene, LLD	2025	6
Styrene	8779	4	Benzene	18289	6
Phenol	8510	2	Urea	1959	5
Xylenes, Paraxylene	5906	3	Ethylbenzene	2729	4

Table 1. (Continued)

PET	5325	4	PET	5325	4
Polystyrene	5262	2	Styrene	8779	4
Terephthalic Acid	5153	3	Isobutylene	7580	4
Propylene	3140	14	Cumene	1209	3
Ethylbenzene	2729	4	Xylenes, Paraxylene	5906	3
Polyethylene, LD	2657	3	Terephthalic Acid	5153	3
Polyethylene, HD	2410	8	Polyethylene, LD	2657	3
Hydrogen	2206	3	Hydrogen	2206	3
Polyethylene, LLD	2025	6	Ethylene Oxide	4454	3
Urea	1959	5	Polystyrene	5262	2
Polystyrene	1478	1	Phenol	8510	2

From Table 1, among the top five compounds, ethylene is the only carbon-containing compound in both A and B lists. Therefore, ethylene was selected, to exemplify one of the current technologies that should be replaced by another one converting CO₂ into a product that not only has a large market but also the current producing process is within the top five in energy intensity. Thus, regarding ethylene, it could be produced through thermocatalytic (e.g., [35–43]) or electrocatalytic (e.g. [44–50]) hydrogenation of CO₂, by the reaction shown below:



Global demand for ethylene is about 150 Mton/year [51]. Therefore, about 500 Mton/year of CO₂ would be consumed to satisfy the entire ethylene global market, if it is produced through this reaction. Consequently, the production of one of the largest market-demanded single products represents just 1.3% of the consumption of the current 37 GtonCO₂/year of global emissions [2]. Since it cannot be expected that current producers would shut down their existing facilities, nor that the olefins market would grow to catch up on the CO₂ emissions to consume the remaining 36.5 GtonCO₂/year then, it is clear that more than one conversion technology is needed for valorizing the massive quantities of emitted CO₂ and for rendering a larger variety of products and to satisfy broader market needs. Therefore, a suite of process technologies is needed for producing this wide variety of value-added products, from the immense source of carbon, emitted as CO₂ from static facilities.

Numerous reactions leading either to value-added products or to render a service or application of CO₂ have been proposed, studied, and developed. Production of fuels, bulk or commodity chemicals including inorganic materials and specialty products with biological activity such as health products and medicines have been suggested. Additionally, the production of functionalized polymeric materials in which carbon is fixated during prolonged times has been studied to a lesser extent though besides the economic benefits will bring environmental advantages, as well. These reactions and in general, CU and/or CO₂ conversion have been the subject of multiple review articles, chapters, and books [5,9,52–97]. In Table 2, relevant examples are given to emphasize the number and type of reactions studied, the level of progress or maturity [98] and the covered time. Some references for patents and patent applications are also included, though these are just a few examples and cannot be taken as a measure of intellectual property efforts.

Table 2. Some examples of chemical reactions of CO₂ and their marketable products.

Product	Reaction	Chemical Reaction	References	Patents (e.g.)
Carbon Monoxide	RWGS	$CO_2 + H_2 \rightarrow CO + H_2O$	[99–116]	[117]
	Electrocatalytic reduction		[118–150]	[151–162]
	Reversed Boudouard	$C + CO_2 \rightarrow 2CO$	[163,164]	
Carbon	Sorption Enhanced Boudouard	$CaO + CO_2 \leftrightarrow CaCO_3$	[165]	
		$CaO + 2CO \leftrightarrow CaCO_3 + C$		
	Boudouard	$2CO \rightarrow C + CO_2$	[163–165]	

Table 2. (Continued)

Syngas	Dry Reforming	$\text{CO}_2 + \text{CH}_4 \rightarrow 2\text{CO} + 2\text{H}_2$	[166–219]	[220,221]
	Via Chemical Looping (1)	$\text{CH}_4 (\text{CO}, \text{H}_2) + \text{MeO} \rightarrow \text{CO}_2 + \text{H}_2\text{O} (\text{CO}_2, \text{H}_2\text{O}) + \text{Me}$	[222–225]	
		$\text{Me} + \frac{1}{2}\text{O}_2 \rightarrow \text{MeO}$		
	Via Chemical Looping (2) w/integrated CC	$4\text{MeO} + \text{CH}_4 \rightarrow 4\text{Me} + \text{CO}_2 + 2\text{H}_2\text{O}$	[56,226–254]	
		$2\text{Me} + 2\text{CO}_2 \rightarrow 2\text{MeO} + 2\text{CO}$		
		$2\text{Me} + 2\text{H}_2\text{O} \rightarrow 2\text{MeO} + 2\text{H}_2$		
		$\text{CH}_4 + \text{CO}_2 \rightarrow 2\text{CO} + 2\text{H}_2$		
		$2\text{Me} + \text{O}_2 \rightarrow 4\text{MeO}$		
	Iron looping	$\text{Fe}_2\text{O}_3 + 3\text{CO} \rightarrow 2\text{Fe} + 3\text{CO}_2$	[255,256]	
		$3\text{Fe} + 4\text{H}_2\text{O} \rightarrow \text{Fe}_3\text{O}_4 + 4\text{H}_2$		
		$4 \text{Fe}_3\text{O}_4 + \text{O}_2 \rightarrow 6 \text{Fe}_2\text{O}_3$		
	(X + CO ₂) Co-electrolysis or Combined electrocatalytic		[130,257–280]	[153,281–293]
Methane	Sabatier	$\text{CO}_2 + 4\text{H}_2 \rightarrow \text{CH}_4 + 2\text{H}_2\text{O}$	[88,294–371]	[372,373]
	Hydrogenation w/steam	$\text{CO}_2 + 2\text{H}_2\text{O} \rightarrow \text{CH}_4 + 2\text{O}_2$	[374–376]	
Methanol	Catalytic Hydrogenation	$\text{CO}_2 + 3\text{H}_2 \rightarrow \text{CH}_3\text{OH} + \text{H}_2\text{O}$	[80,87,377–459]	[460–468]
	Electrocatalytic		[469–500]	[283,284,460,501–503]
DMC & Organic Carbonates	Carboxylation	$\text{CO}_2 + 2\text{ROH} \rightarrow \text{RO-COO-R} + \text{H}_2\text{O}$	[504–519]	
Polycarbonates	DMC Polymerization		[6,520–539]	
Polyurethanes			[528,540–545]	
Carboxylic acids or Esters	Hydrocarboxylation of olefins or alcohols		[546]	
Polyacids or Polyesters	Polymerization		[547,548]	
Inorganic carbonates	Carbonation	$2\text{H}^+ + \text{H}_2\text{O} + (\text{Ca}, \text{Mg}, \text{Fe})\text{SiO}_3 \rightarrow \text{Ca}^{2+}, \text{Mg}^{2+}, \text{Fe}^{2+} + \text{H}_4\text{SiO}_4$	[55,549–575]	[576–580]
		$\text{H}_2\text{CO}_3 + \text{Me}^{2+} \rightarrow \text{MeCO}_3 + 2\text{H}^+$		
		$\text{Mg}_2\text{SiO}_4 + 2\text{CO}_2 \rightarrow 2\text{MgCO}_3 + \text{SiO}_2$		
		$\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4 + 3\text{CO}_2 \rightarrow 3\text{MgCO}_3 + 2\text{SiO}_2 + 2\text{H}_2\text{O}$		
		$\text{Ca}_2\text{SiO}_4 + 2\text{CO}_2 \rightarrow 2\text{CaCO}_3 + \text{SiO}_2$		
		$\text{CO}_2 + \text{NH}_3 + \text{H}_2\text{O} \rightarrow (\text{NH}_4)\text{HCO}_3$		
Urea	Two steps process	$2\text{NH}_3 + \text{CO}_2 \rightleftharpoons \text{H}_2\text{N-COONH}_4$	[581–586]	[587–591]
		$\text{H}_2\text{N-COONH}_4 \rightleftharpoons (\text{NH}_2)_2\text{CO} + \text{H}_2\text{O}$		
Urea polymers	Polymerization	w/CH ₂ O	[592–605]	
		w/melamine	[588–591,606–615]	

The current trend (not necessarily reflected in Table 2) of devoted R&D efforts [616] is mainly associated with reactions of the CO₂-H₂ pair [617] and the development of electrocatalytic processes [485,618]. The technical advances reached in most of the reactions collected in Table 2 were summarized in [619], including the identified gaps, needs, and challenges. The Technology Readiness Level (TRL) of CCUS and CU technologies was reviewed in [98] and [97], respectively. In the case of the CU, there was a high degree of uncertainty in TRL determination. As can be concluded from the examination of Table 2, the amplitude of the CU technology portfolio based on CO₂ conversion reactions is quite large though currently, most of these products cannot yet reach the market since the low TRL of the corresponding technologies. A graphical representation of this technologies maturity, spanning from TRL 1 through TRL 9 [97,98] is presented in Figure 4, along the curve of the S-curve of technology research, development and demonstration (RD&D). Only a few products (methanol, CO₂-based polycarbonates, polyols, polyurethanes, salicylic acids, and urea) and some inorganic carbonates have reached TRL 9, particularly for cases where CO₂ is used in the production of intermediates. Methanol from CO₂

is an exception since a commercial demonstration plant has been operating in Iceland for almost two decades [620,621]. The other exceptions are urea and salicylic acid, which together with polycarbonates consume 26% (130 MtonCO₂/year) [622] out of the reported utilized 500 MtonCO₂/year.

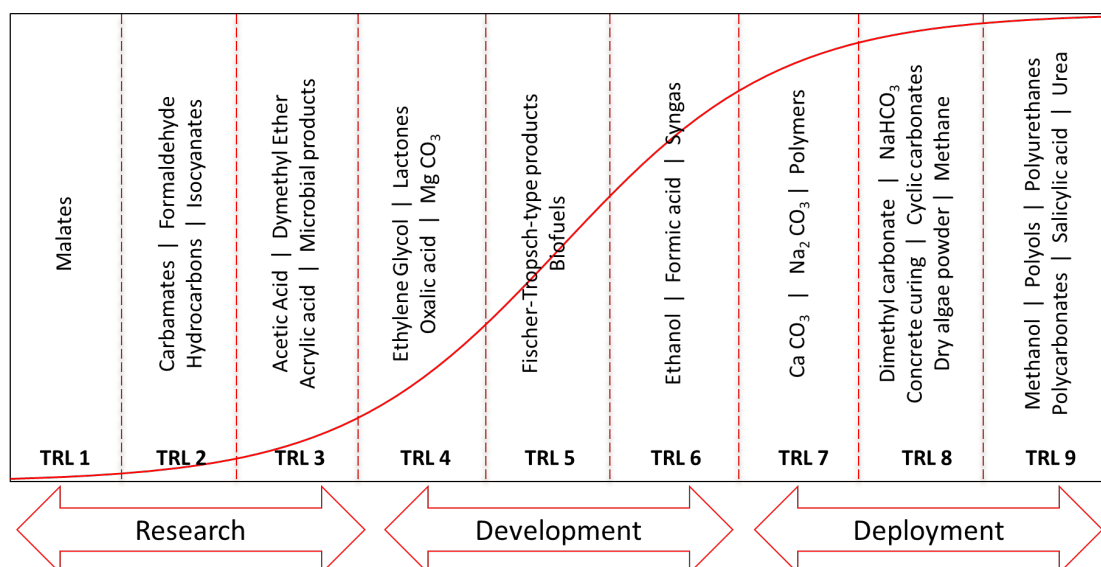


Figure 4. TRL status of technologies rendering referred products (from the information reported in [97]).

As can be deduced, the reaction options are quite broad but the reported efforts and results on these reactions are even larger since only references relevant to this work have been included in this table. Thus, the examination of this vast literature cited indicates the three particular trends within the R&D efforts that could be emphasized, namely, (i) synthesis of highly demanded compounds, (ii) pathways (reactions) that may consume large CO₂ amounts, and (iii) products that may retain CO₂ moiety for long periods (i.e., fuels production/uses are avoided).

The much greater impact of large-scale production based on CO₂ conversion, as summarized in Table 2, on the overall emissions reduction, has been recognized by Dziejarski et al. [98]. The incorporation of CU or CO₂ valorization, for the effective closure of the C-cycle requires the articulation of several strategies, among which become relevant to this work: wastes valorization, effective treatment and/or processing of flue gases, generation of high value-added products for diverse applications, integrated approach to multi-waste treatment as green solutions, business models based on industrial eco-designed parks and eco-effective integration for zero waste ([623] and references therein). Within the approaches followed to achieve decarbonization, three particular cases could interplay favorably towards a sustainable closure of the C-cycle: (1) long-lasting products immobilizing CO₂; (2) improvements in process efficiency; (3) deployment of decarbonized (renewable or low-C) energy, including electricity and fuels. Examples of CU falling within these approaches might include industrial wastes for carbonation, CO₂ hydrogenation, and electrochemical processes [624]. Therefore, new more efficient and effective chemical processes, for CO₂ conversion into longer lifecycle products need to be developed. Ideally, these processes would convert massive volumes of CO₂, with high selectivity. The optimal use of resources points towards the integration of processes and energy. These highly integrated plants for the processing of large volumes of CO₂, yielding a product slate that satisfies a broad range of market demands, fall in the category of a refinery, i.e., a CO₂ refinery.

3. Sustainability Considerations

In regards to sustainability, Artz et al. [8] presented two ways of addressing this topic. Typically, CU is seen as a way to mitigate CO₂ emissions by reducing its flow to the atmosphere from the emitting facility. On the other hand, the carbon balance and other critical environmental factors could be greatly improved by implementing CO₂-based chemistry for the production of fuels, chemicals, and materials to substitute those of fossil origin. This review article questioned the rare appearance of life cycle assessment (LCA), for evaluating the potentialities of the studied conversion pathways and stressed the importance, relevance, and complexity of this type of

analysis. Although they concluded that the highest potential of CO₂ conversion resides in the replacement of energy-intensive feedstocks by CO₂, here the energy intensity of CO₂ activation and reaction should be recognized and will rather point towards lowering the energy intensity of the conversion process and improvements in their energy efficiency.

Recently, a concept of a CO₂ biorefinery for the production of fuels and bulk chemicals has been proposed and found financially attractive and environmentally beneficial [625,626]. The first work proposes and discusses multiple pathways to be considered for CO₂ upgrading [625]. A brief mention of the chemical conversion routes (thermochemical, photochemical, and electrochemical) focuses on methanol production. Most of the work discusses biological means for both fixation and conversion, by terrestrial plants, microalgae, and microorganisms. The authors identified challenges closely related to the lack of sustainability of the reported and discussed results, which emphasized the importance of attaining technical and economic feasibility. These challenges include the development of high-performance catalysts and overcoming the high cost of conversion process into biofuels [625]. The second CO₂ biorefinery concept was centered on two core pathways described as Schemes 1 and 2 in [626]. The reactions considered in Scheme 1 include CO₂ reduction and hydrogenation, into fuels or fuel precursors, via catalytic and electrocatalytic processes. Meanwhile, Scheme 2 considers the production of bulk chemicals by reactions with basic molecules such as ammonia or small organic compounds. Most of the chemical processes considered in Scheme 2 have reached commercial demonstration and/or implementation while those in Scheme 1 are mostly bench-top proof of concept. Scheme 1 has the potential of consuming nearly 30% of the global CO₂ emissions but to produce fuels, which in turn will be responsible for equivalent CO₂ emissions. In terms of CO₂ consumption, Scheme 2 might take 3–4% of the global emissions though it might be more economically favorable due to the higher value of the products [626]. Neither of these CO₂ refinery concepts incorporates sustainability considerations, which have to be brought to the plate early, i.e., during the definition of the configuration. A third CO₂ biorefinery concept has been suggested to be an electro-biorefinery and be centered on microbial electrochemical synthesis (MES) [627]. Although this work offers to describe and discuss the potentialities of a CO₂ electro-biorefinery, it rather describes the R&D status of the MES process and identifies the existing needs and gaps within its scaling up towards industrialization. Furthermore, the status of the development precluded the authors from concluding whether MES could be the core process of a CO₂ electro-biorefinery or just a process in any other type of CO₂ refinery.

In fact, configuring the CO₂ refineries could advantageously apply the principles derived from the eco-design, eco-efficiency, and eco-effectiveness concepts, delineated above. Additionally, the historical growth of oil refining as well as the experience gained from the failures during the commercialization of second-generation (2G) biorefineries offer many lessons learned that should be considered and examined. In both cases, refining [628] and biorefining [629] configurations have been defined following a bottom-up approach (Figure 5a).

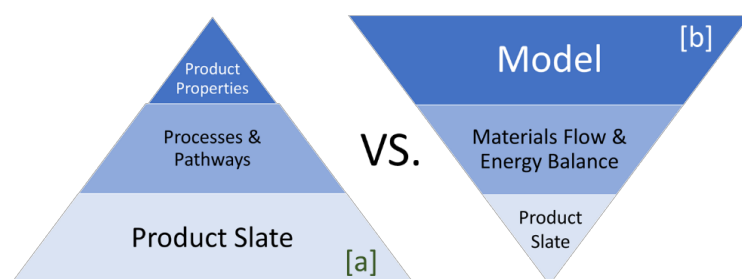


Figure 5. Approaches for refinery configuration design: (a) Bottom-up vs. (b) Top-down.

In the bottom-up approach, process technologies are first developed to render product(s) either to satisfy a market need or to resolve an end-user problem. Then, these process technologies are integrated to attain viability, technical, and in some instances economical, at least in [628]. Regarding the economy, 2G-biorefining has not been proven to be economically sustainable [630] and oil refining has passed through margin struggling periods [631]. Furthermore, both biorefineries [78] and oil refineries are CO₂ emitters, resulting in environmentally unsustainable or at least with some environmental limitations [632,633]. Moreover, the urgency imposed on decarbonization and the immediate need for increasing the magnitude of CU are driving forces

that cannot wait for the historical evolution of concepts and models to start configuring and designing CO₂ refineries. The process technologies should be integrated using eco-efficient and eco-design criteria to fulfill net-zero goals. Additionally, a sustainably designed refinery would optimize the management of the carbon resources to make sure of optimum settings for carbon efficiency, energy efficiency, levelized costs, durability, and robustness. The CO₂ refinery configuration could be eco-designed by following a top-down approach, from a model underpinned by eco-efficiency and eco-effectiveness criteria (Figure 5b). Contrary to the bottom-up approach, the top-down approach is centered on circularization and starts with the development of an integral TEA-LCA model. The model would allow to perform materials flow analysis to identify uses, applications, and post-processing of intermediate and stranded streams, and to define potential circular configurations. The definition of key economic, social, and environmental performance metrics for technology comparison will be key in evaluating sustainability metrics, e.g., water footprint, life cycle cost, global warming potential, ecological footprint, and health-adjusted life years. Selected eco-design configurations can be compared based on TEA-LCA assessments to quantify technology performance, identify technical gaps, and forecast improvements. Applying the three principles (eco-design, eco-efficiency, eco-effectiveness) will promote technologies and practices that minimize resource squandering and waste generation, maximize value creation, and design for reintegration of materials into natural bio-, geo- or chemical processes at their end-of-life. Circularization through eco-design is based on the “dematerialization” transformative change, replacing current practices with measures, policies, or technologies to decrease the quantity of consumed resources and minimize waste generation. The strong need for dematerialization practices has been stressed since improved cleaner technologies, resource recovery, more efficient conservation, and recycling will not be enough to achieve sustainability.

For the CO₂ refinery, the required energy should be provided by a low-carbon power plant and the feed can be supplied either from static emitting sources (biorefinery, oil refinery, power plant, etc.) either through a pipeline network or by integration (chemical or physical). Construction of pipeline networks to interconnect and collect multiple emissions from different emitting facilities has started already. In the case of the U.S., about 8500 km of pipelines were already constructed by 2021, which were transporting an average of 80 MtonCO₂/year [634] (globally a capacity for transporting more than 240 MtonCO₂/year has been built or is under advanced development [14]). The defined infrastructure plans have been based on the analyses of different scenarios, mostly connected either to sequestration or to enhanced oil recovery (EOR). Some of these plans and scenarios consider the construction of 20,000–110,000 km of pipelines, connecting the emitting facility to the sequestration or utilization field [635]. Nevertheless, it is worth mentioning that feeding a CO₂ refinery from pipeline networks would underpin larger capacity refineries, in a preferred scenario benefitted from the economy of scale. Since a refinery can be built at a more convenient location, the issues derived from the logistic mismatch could be minimized and even overcome.

The products manufactured in this wholly integrated CO₂ refinery will contribute to fulfilling the net-zero goals set by the “The 26th UN Climate Change Conference (COP26)” [1] and to achieve sustainability. Preferred process technologies fall within the definition of the so-called “negative emission technologies, NETs”. Each of the conversion processes listed in Table 2 are not necessarily NETs though technologies consuming large volumes of CO₂ to convert it to long-lasting products, using low-carbon energy sources have great potential for becoming NETs. However, the needed measures within a net-zero scenario, at a close approach to COP26 2050 committed goals require that the implementation of the developed technologies takes place with the sustainability principles of a circular economy (eco-design, eco-efficiency, and eco-effectiveness criteria, as mentioned above).

The R&D attention for CCUS technologies development has been diverted to specific topics, due to the broad range of composition of industrial emissions (one of the mismatches mentioned above). The variability of these streams brings about different scenarios and situations, for which no universal solution exists. As per similarity with oil refining, a given scenario—situation would be solved with an optimal configuration, constituted by a specific NETs portfolio. Feasibility, effectiveness, and side impacts considered holistically are the critical performance parameters to be assessed. In the bioenergy technologies portfolio, it has been found that only *an optimized technology portfolio timely deployed* will sustainably contribute to mitigating climate change [636]. In this case, suboptimal solutions resulted in infeasible, ineffective, and even created larger problems. In this study, the most promising NETs in the area were evaluated: afforestation and reforestation,

biochar, bioenergy with CCS, direct air carbon capture and storage, enhanced weathering on land and in oceans, ocean fertilization, and soil carbon sequestration [636].

As mentioned above, and according to several discussions and revisions [4,5,9,53,54,69,97,98], the developed process technologies have reached different levels of research, development, and maturity, and clearly, gaps and needs for further R&D in the corresponding topics still exist. For these reasons, it is necessary to establish a technology pipeline that positions the processes' availability in terms of a timeframe. The definition of this technology pipeline starts with a technology analysis based on the published patent literature. The importance of this type of analysis has been indicated by Bisotti et. al. [394] though their study concerns the conventional methanol synthesis technology and the development of a better kinetic model for such reaction, which falls outside the scope of the present work. Additionally, their technology analysis associates technological evolution with the results of a bibliometrics analysis of the patents. Furthermore, the high number of new catalysts developed was associated with the growth of the methanol market. One of the first remarks that should be made is that the technology market is not the market of the technology product(s). Then, it is fair to say that the vast majority of patented catalysts rarely reach commercial scale. Finally, according to technology licensors and/or catalysts vendors, the commercially available methanol synthesis catalysts have been optimized to perform and to last for their corresponding process. Many of these catalysts are part of the technology package that the licensor offers to the user and the process operator cannot change it without losing the guarantees. The reader interested in the real status of the technologies for methanol synthesis from CO₂ could refer to [9,33]. Further details of the thermocatalytic and electrocatalytic work for these processes can be found in the references cited in Table 2 (85 articles and 15 patents or applications). The tech pipeline consists of a database of the processes with potential for integration within a CO₂ refinery. It should contain the description and state of the art of the technology market as well as their relevant data for the design and development of the proposed model to configure CO₂ refineries. Particular emphasis should be placed on technical data concerning carbon-efficiency (conversion, product slate, and yields), energy efficiency (consumption, balance, and losses), and material balances (waste generation, losses, and by-products). Besides building the model, the collected information could also be used to identify needs, challenges, and gaps to guide R&D, e.g., to accelerate or kill a given development attempt.

An integral model combining TEA, LCA, and optimization tools is desirable to understand the performance and benefits of the different configurations and to generate insights into the impact on the sustainability of feedstock compositions (see Figure 6). Several issues have been reported on C-accounting that invalidate the C-intensity and efficiency results [637], stressing the importance of method selection, i.e., consequential or attributional, which are key for carbon and GHG accounting, and for life cycles. The issues associated with C-accounting for NETs have been pointed out since these technologies not only remove carbon but also, they might emit it. The recommendation was to report emissions and removals when and where they actually occur, providing the appropriate accounting method is used. Thus, for NETs incentivization or for supporting decision-making consequential methods were recommended while attributional methods were better employed in the descriptions of possible net zero static scenarios [637]. As mentioned already, addressing the magnitude of the CO₂ emissions calls for multiple gigaton solutions. Examples of the "removal" capabilities (or emissions reduction capability) of some technologies were provided by Pacala & Socolow [638], for instance, reduction of 1 Gton of CO₂ emissions can be attained with 1000 zero-emission 500 MW coal-fired power plant with CCS, by installing 50 times the current global capacity of wind energy or by planting energy crops 15 times the size of Iowa. Net zero goals require 37 times these types of solutions, leading to the cost-effectiveness barriers found in the implementation and deployment of CCS solutions [639].

Configuration complexity, a well-known term in oil refining, needs to be defined and assessed for the newly introduced CO₂ refinery concept. The degrees of freedom increase with complexity. Likewise, feedstock flexibility and product slate broadening also increase with complexity. Therefore, it could be expected that complexity would impact the sustainability indicators. Regarding feedstock flexibility, the real composition of industrial emissions is practically unknown. Their chemical characterization, as well as their volumetric quantification, are needed for the most concerning CO₂-containing industrial feedstocks particularly, from the larger emitters, e.g., (fossil-fired) power plants, cement, steel, and/or refining. The optimum management of materials involves the full valorization of the feedstock, even including contaminants. Closing all material flow loops warrants zero waste processing and maximizes eco-efficiency. Feedstock composition

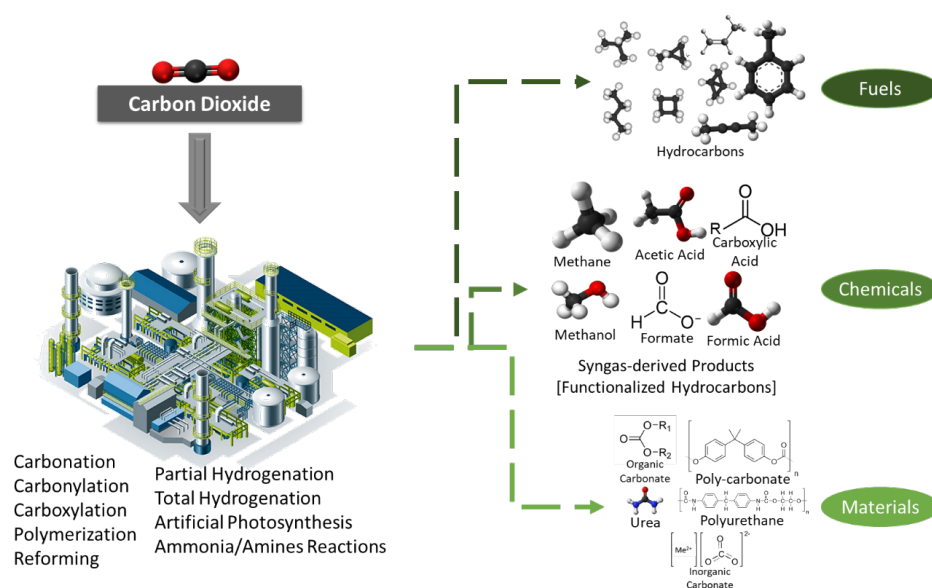


Figure 6. Value-added products from the conversion of CO₂ in an eco-designed refinery.

also determines the need for pretreatment, separation, and/or conversion units that might turn into increasing complexity. Further increments in the number and type of compounds present in the feedstock lead to increases in refinery complexity. Additional increases in complexity might arise from product slate broadening and from producing materials with a longer lifecycle or service life. Even though increasing the configuration complexity will bring technical advantages, it will nonetheless impact the economy of the refinery. A simple configuration of a CO₂ refinery has been recently economically and environmentally evaluated [640]. This configuration was defined following a bottom-up approach and includes methanol synthesis, from DAC-CO₂ as feed, integrated to methanol-to-olefins (MTO) and methanol-to-aromatics (MTA). The quantified refinery benefits were highly limited by the high cost of green hydrogen and the atmospheric CO₂ [640]. Furthermore, these results can be also taken as an indication of the failure of the bottom-up approach, and of the need for incorporation of the three eco-principles and for a circularization strategy that optimizes resource use and minimizes waste generation. Finally, although it shows the advantages of the early conceptualizing of the configuration, that concept missed the relevance of maximizing the volume of CO₂ consumed, the inclusion of long-lasting products (not fuels), and the use of a low-carbon energy source and/or more energy-efficient processes.

The application of “the one problem-one solution approach” requires tailor-made solutions for each emitting facility, which is far from sustainable. Meanwhile, few CO₂ refineries, such as the one proposed here might be more prompt to attain the COP26 goals and become sustainable, particularly when the configuration has been eco-designed following eco-efficiency and eco-effectiveness criteria, with the top-down suggested approach and the considerations discussed above.

4. Final Remarks

In order to close the gap between carbon dioxide (CO₂) emissions (Gton/year) and its utilization (Mton/year), neither a single product nor an exclusive pathway are holistic solution. Instead, processes consuming massive CO₂ volumes, exhibiting lower intensity in carbon- (C-) and energy, and rendering products or services with longer longevity or lifecycle are needed. Therefore, a suite of process technologies should be configured efficiently and effectively, into an integrated CO₂ refinery, to change the way and pathways for producing fuels, chemicals, and materials (Figure 6), from the unlimited source of carbon, emitted as CO₂. The knowledge and learnings from refining and biorefining can be applied together with the novel concepts of eco-design, eco-efficiency, and eco-effectiveness to configure such CO₂ refineries. This work presents and discusses the author’s views on an innovative top-down methodology that could lead to a sustainable carbon cycle (in terms of energy, environment, and economy). Research and development gaps and needs could be identified using the suggested methodology before any further investment. Therefore, eventually, a sustainable CO₂ refinery will be the vehicle to close the carbon cycle of a CCE and underpin a decarbonized chemical industry.

In summary, this review has served to establish that (i) CU complements CCS, (ii) CCS and CU are not enough to fulfill net-zero goals, (iii) CU and CCS can be integrated to create a “Circular Carbon Economy, CCE”, (iv) circularization is not a warranty for sustainability, (v) CU should focus on the use of massive volumes of CO₂, (vi) conversion into value-added products create revenues, (vii) manufacture of long-lasting products can mitigate atmospheric accumulation, and (viii) eco-principles could lead to sustainable technological solutions. Therefore, the proposed CO₂ refinery can holistically be seen as a “negative emission technology package”. Hopefully, the application of the eco-principles, the circularization, and the conversion of massive volumes of CO₂ into long-lasting/value-added products, using low-C energy sources would all sum up to an effective carbon reduction strategy, underpinning a CCE.

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Data Availability

No new data were created or analyzed in this work. Data sharing is not applicable to this article.

Conflicts of Interest

The author has no conflict of interest to declare.

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