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Recent Advances and Future Perspectives of Low-Concentration CO_2 Enrichment and Emerging Applications

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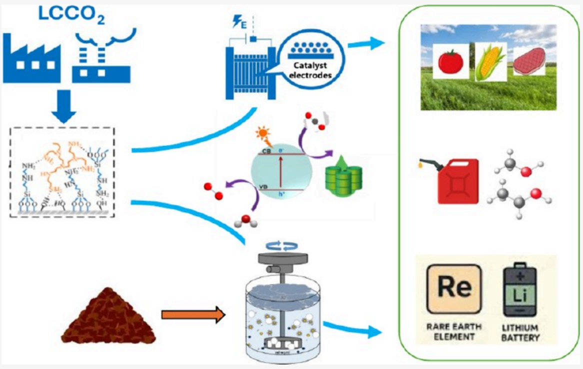

ABSTRACT: Low-concentration carbon dioxide ($LCCO_2$) streams, ranging from ambient air to industrial flue gas, represent a significant yet underutilized carbon resource. This review examines recent advances in technologies for enriching $LCCO_2$ and enabling its integration into a wide range of applications. It covers emerging materials and processes for CO_2 capture, including immobilized amines, metal oxides, polymers, porous carbons, and zeolites, with a focus on performance metrics, regeneration strategies, and process intensification. Beyond capture, the review explores the direct utilization of $LCCO_2$ through reactive capture pathways enabled by novel catalysts and processes, as well as nonreactive applications, such as greenhouse cultivation, enhanced weathering, and sustainable agriculture. Additionally, this review highlights the role of $LCCO_2$ in enhancing critical metal recovery. Finally, the review outlines crucial future research directions necessary to fully realize $LCCO_2$ enrichment's potential within circular carbon economies and critical material supply chains, paving the way toward a more energy-innovative and resource-resilient industrial sector. The findings are significant for researchers in academia, industry, and government, as well as the public interested in extending the value of $LCCO_2$.

1. INTRODUCTION

New opportunities to utilize carbon dioxide (CO_2) in the production of chemicals, foods, and critical materials have attracted significant attention from researchers, engineers, and policymakers. CO_2 can be separated and concentrated from a range of diluted sources, including atmospheric air with ~ 400 ppm and industrial flue gas streams with CO_2 concentrations typically ranging from 4−20 vol %.[1−3] These sources represent a substantial, underutilized carbon resource, yet the energy-intensive nature of CO_2 capture and purification remains a key barrier to widespread implementation.

Conventional CO_2 capture processes typically aim for high-purity CO_2 (≥95 vol %) to meet transportation, storage, or chemical conversion requirements. Achieving such high purity often involves multistage separation processes, such as amine scrubbing, cryogenic distillation, or membrane separation. This approach incurs significant energy penalties due to the regeneration of CO_2 capture media and the compression of CO_2, increasing both the capital and operational expenses for producing CO_2-derived products. Furthermore, transporting the purified CO_2 to the conversion site adds to the overall energy consumption. These processes can consume significant amounts of energy, often making them economically unfeasible for low-concentration sources.[4−8] Compared to traditional high-purity CO_2 (≥95%) systems that dominate current CCS/CCU practices, low-concentration CO_2 ($LCCO_2$) sources such as flue gas, ventilation exhaust, or ambient air offer both techno-economic challenges and novel opportunities. While high-purity CO_2 capture relies on energy-intensive separation, compression, and transport infrastructure, $LCCO_2$ can be accessed and valorized more locally—often without full purification. This enables decentralized, modular, and potentially lower-energy utilization pathways, such as direct flue-to-fuel reactors, greenhouse enrichment, or reactive mineralization. As summarized in Table 1, $LCCO_2$ systems generally exhibit lower capture costs and energy intensity, and they support a broader range of applications in distributed or low-margin contexts.

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Table 1. Comparison of Low-Concentration CO_2 ($LCCO_2$) and High-Purity CO_2 Systems in Terms of Capture, Energy Demand, and Applications

category	low-concentration CO_2($LCCO_2$)	high-purity CO_2 (≥95%)
capture cost	\$15−100/t CO_2 (varies by technology and concentration)	\$40−200/t CO_2 (including compression and purification)
energy demand	1−3 GJ/t CO_2 (adsorption, membrane, hybrid systems)	3−6 GJ/t CO_2 (scrubbing + compression)
infrastructure needs	decentralized, modular setups	large-scale, centralized plants
utilization pathways	greenhouse enrichment, flue-to-fuel reactors, mineralization	methanol synthesis, EOR, polymer/chemical feedstocks
integration potential	high with renewables and distributed systems	moderate, requires CO_2 hubs or pipelines

$LCCO_2$, defined broadly as CO_2 with a concentration from several thousand ppm up to 90 vol %, can be used directly without additional purification in a variety of systems. In response, there is growing interest in leveraging $LCCO_2$ streams,[9,10] wherefrom CO_2 can be directly fed or enriched at significantly lower energy cost and still provide functional value in certain industrial and environmental applications. The utilization of $LCCO_2$ is gaining increasing attention as a critical strategy for achieving and fostering a more sustainable future. Recent research has explored diverse technological approaches, ranging from innovative capture and conversion methods to direct utilization in various industrial and biological processes.[9−16] Shen et al.[17] provided a comprehensive overview of the behavior and mechanisms of $LCCO_2$ in various energy media, including solid and liquid absorbents, as well as catalytic surfaces. Their review underscored the importance of advancements in thermal, electrochemical, photo, plasma, and biocatalysis. Several studies focus on novel methods for capturing and converting $LCCO_2$ into valuable products. Building on these themes, Kar et al.[18] introduced a promising gas-phase dual-bed reactor that directly captures CO_2 from ambient air using a silica-amine adsorbent. This captured CO_2 is then converted into syngas (CO + H_2) under concentrated sunlight, offering a decentralized and energy-efficient approach to renewable fuel production. Electrocatalytic approaches also show significant potential. Stanley et al.[19] developed a sorbent-mediated electrocatalytic system that efficiently reduces dilute CO_2 to methane. By concentrating CO_2 at the catalyst surface using a sorbent material, the system overcomes the challenges associated with low CO_2 concentrations and enhances reaction rates. Biocatalytic systems offer alternative routes for converting $LCCO_2$ into valuable chemicals under mild conditions. Kita and Amao[20] developed a visible-light-driven system that produces 3-hydroxybutyrate from acetone and $LCCO_2$, coupling photocatalytic NADH regeneration with biocatalysts to transform waste streams into a biodegradable plastic precursor. Similarly, Takeuchi and Amao[21] reported a multienzyme system that converts pyruvate and low-concentration CO_2 (<15%) into fumarate, a precursor for biodegradable plastics. To further improve the efficiency of $LCCO_2$ conversion, Yamazaki et al.[22] explored the use of molecular catalysts assisted by CO_2-capturing additives or reaction media. Their work demonstrated that incorporating CO_2-capturing agents can significantly enhance CO_2 conversion rates, making it possible to utilize even highly dilute CO_2 streams. In chemical manufacturing, low- to moderate-purity CO_2 serves as a reactant for producing methanol, urea, and synthetic hydrocarbons via catalytic or electrochemical routes.[6,23]

Beyond conversion technologies, $LCCO_2$ can also be directly utilized in various applications. Enriching CO_2 in greenhouses to levels between 800−1,200 ppm with ex-situ controlled-environment agriculture, for example, has been shown to significantly increase plant biomass and yield.[9,15] Supercritical CO_2 ($scCO_2$) extraction techniques, often enhanced with cosolvents, have also been explored to recover valuable elements from electronic wastes[24,25] and mine tailings,[26,27] showcasing their potential as a selective, low-energy recovery method. Similarly, $LCCO_2$ plays a key role in mineral carbonation processes,[11] which offer a dual benefit of carbon storage and critical metals recovery. $LCCO_2$ can enhance the leaching of lithium from minerals like spodumene and facilitate the carbonation of magnesium- and calcium-rich tailings, stabilizing carbon while simultaneously liberating rare earth elements (REEs) or other valuable metals.[28] Alternatively, one study using molecular dynamics simulations showed that CO_2 enriched to at least 70% from a direct air capture (DAC) process can be directly sequestered rather than further purified for other applications. Further in this study[29] Zhang et al. referenced that among DAC technologies, the lower end of \$94/t CO_2 capture cost for achieving purities presumably ≫70% was markedly higher than a \$10/t CO_2 onshore storage cost. Although not conclusive on absolute cost effectiveness compared to CO_2 utilization, this work highlights CO_2 storage as a viable option to handle $LCCO_2$.

Importantly, the growing body of research in CO_2 enrichment underscores the potential of $LCCO_2$ to reduce the energy intensity and cost of carbon capture, while simultaneously opening new pathways for sustainable manufacturing, carbon circularity, and resource recovery. Integrating $LCCO_2$ into chemical production processes presents a particularly promising route to sustainable manufacturing, with advancements in CO_2 capture, electrocatalysis, biocatalysis, and catalytic conversion technologies paving the way for efficient utilization of dilute CO_2 streams. These innovations not only contribute to carbon mitigation but also create economic value by producing valuable chemicals and fuels from waste CO_2.

To our knowledge, there are few reviews in the literature aimed at $LCCO_2$ enrichment. These reviews have focused largely on the needs for greenhouse crop production,[30] including for free-air CO_2 enrichment (FACE) crop generation, which utilizes a previously CO_2-enriched gas stream for accelerated vegetation growth,[31] and sorbent-based DAC processes designed and assessed based on greenhouse agriculture applications.[32] The review presented herein examines the technological innovations, material developments, and process strategies that enable the utilization of enriched $LCCO_2$ streams in varied applications. Particular attention is given to emerging classes of sorbent materials, as well as process intensification approaches that minimize the energy requirements for CO_2 separation from dilute sources. Beyond materials and capture systems, this review also addresses the direct utilization associated with integrating CO_2 enrichment with downstream applications. These include considerations regarding the nonthermal reaction pathways, kinetics, yields, and selectivity for conversions, as well as the energy transition. Ultimately, the review explores the broader implications for sustainable carbon management, especially in the context of national priorities for critical materials supply chains and energy innovations. The goal is to provide a comprehensive assessment of the role LCCO$_2$ enrichment can play in shaping future industrial systems that are both carbon-conscious and resource-resilient.

At present, review articles on CO$_2$ capture and conversion typically focus on either upstream sorbent design or downstream catalytic processes in isolation. However, as industrial applications demand systems that are more integrated, selective, and adaptable to varying CO$_2$ concentrations, there is a growing need to understand how materials and process strategies can be codesigned to enable efficient, localized carbon valorization. The synergistic coupling of capture and conversion within a single unit operation represents a process intensification strategy that not only enhances energy efficiency and process productivity but also advances sustainability and scalability in industrial applications. This review addresses this emerging paradigm by bridging the gap between capture technologies and catalytic functionality, with a focus on reactive systems and concentrated CO$_2$ streams. By surveying recent innovations in multifunctional catalysts, tandem reactor configurations, and energy-coupled systems, the article offers a timely perspective on the next generation of CO$_2$ utilization platforms.

2. RECENT ADVANCES IN CARBON CAPTURE FOR LCCO$_2$ ENRICHMENT

Fundamentally, LCCO$_2$ has the distinct advantage of higher thermodynamic efficiency in concentrating CO$_2$ to a lower than to a higher purity. Figure 1 shows the second law efficiency for different processes of various target species, whereby the efficiencies were associated with processes designed to optimize capital costs that usually increase efficiency, and operating costs that usually decrease efficiency.[33]

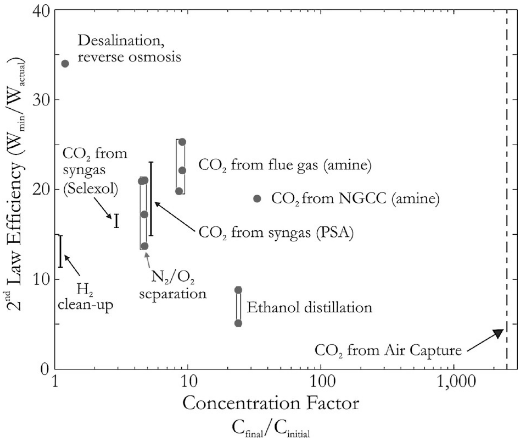


Figure 1. Relationship between second law efficiencies and concentration factors of different separation processes. Reproduced with permission from ref 33. Copyright 2010 National Academy of Sciences.

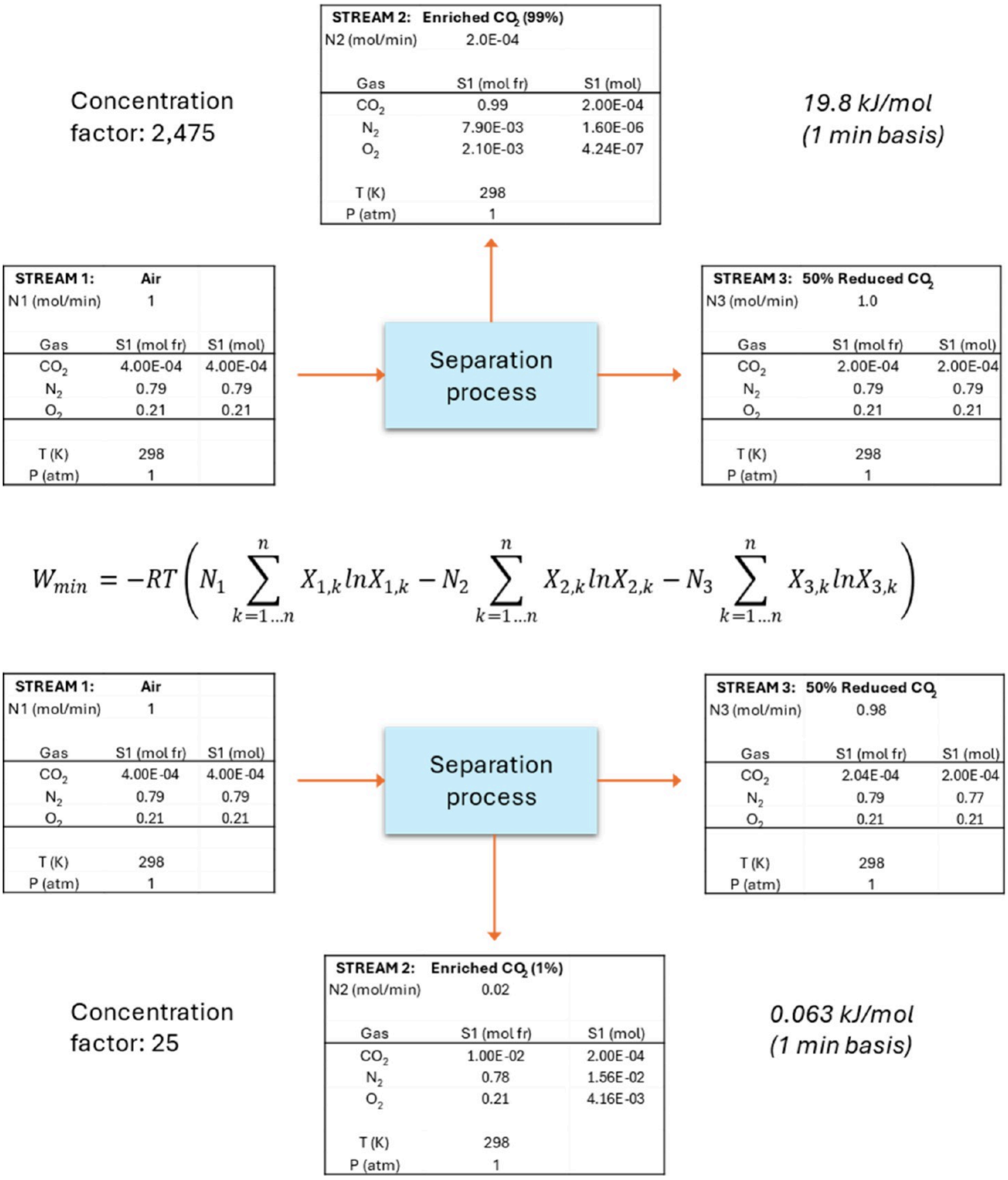
Overall decreasing thermodynamic efficiencies with increasing concentration factors indicate greater resource utilization in enriching analytes, like CO$_2$. House et al. described that greater work is required to handle and process an increasing amount of material and further stated that DAC processes are likely to give <10% efficiency, being closer to 5%. In line with this, rising energy requirements are inherent in essentially all systems where increasingly smaller amounts of material are added or removed due to the decreasing driving force. Offsetting the added costs for increased CO$_2$ purity, high sale prices of pure CO$_2$ should be matched to proportionally "sophisticated" markets, such as those involving medical-grade applications and instrumentation calibration.

According to the isothermal, isobaric, ideal gas assumptions used by House et al, Figure 2 shows the calculated minimum work required to achieve 99% and 1% purity CO$_2$ from the starting 400 ppm (0.04%) purity of the inlet air at 1 atm and 298 K.[33] There, $W_{\min}$ is the minimum work; N_j represents the total molar flow rate of each stream; $X_{i,j}$ is the mole fraction of species k of n total species in stream i = 1, 2, or 3. Replicating House's values, a minimum of 19.8 kJ/mol CO$_2$ is required to enrich CO$_2$ by 2475× to high purity. Conversely, a substantially lower 0.06 kJ/mol CO$_2$ is required to enrich the CO$_2$ concentration by 25× to 1% purity. These basic thermodynamics justify practical research into LCCO$_2$ enrichment. A simple analysis as this is typically used to first determine if a separation process is thermodynamically feasible. Accounting for process inefficiencies by correcting the theoretical minimum work needed would be the next "back-of-the envelope" step in evaluating the economic feasibility of a CO$_2$ enrichment process.

A broad range of materials and processes has been investigated for removing CO$_2$ from air and point sources. Many studies have further explored the potential of LCCO$_2$ enrichment, aiming to produce desorbed CO$_2$ concentrations suitable for various applications. Table 2 summarizes recent studies (primarily within the last 5 years), highlighting sorbent type, process conditions, and CO$_2$ capture-release performance. A key focus is on developing effective and economical methods for enriching CO$_2$ from low-concentration sources like ambient air and industrial waste streams. This involves a variety of sorbent materials, generally classified into five categories: immobilized amines, metal oxides, polymers (e.g., resins), carbons (e.g., activated carbon), and zeolites. Amine-functionalized silicas, MOFs, and hybrid composites show promise for achieving reasonable CO$_2$ uptake capacities under dilute conditions. While the desorbed CO$_2$ concentration is a crucial metric for downstream utilization, with some studies reporting high purities (>90%) and others lower concentrations (20−50%), significant challenges remain. These include improving CO$_2$ selectivity in the presence of moisture, reducing energy consumption for sorbent regeneration, and developing scalable, cost-effective LCCO$_2$ enrichment and conversion processes. To address these challenges, emerging approaches like electrically (Joule heating) and microwave (MW)-heated swing adsorption are being actively explored for enhanced sorbent regeneration, alongside conventional methods such as temperature, pressure, and vacuum swings.

2.1. Novel Sorbents for LCCO$_2$ Enrichment.

Solid sorbents, in general, are a prime technology relative to liquid processes to capture and enrich CO$_2$ due to rapid uptake and release kinetics; simpler process operations; and lower process energy − ex. no heat of liquid H$_2$O vaporization, unlike the benchmark liquid monoethanolamine process. These overall characteristics make for an economic process that rapidly captures CO$_2$ and then subsequently releases it in a more



Concentration factor: 2,475

STREAM 2:	Enriched CO_2 (99%)	
N2 (mol/min)	2.0E-04	
Gas	S1 (mol fr)	S1 (mol)
CO_2	0.99	2.00E-04
N_2	7.90E-03	1.60E-06
O_2	2.10E-03	4.24E-07
T (K)	298	
P (atm)	1	

19.8 kJ/mol (1 min basis)

STREAM 1:	Air	
N1 (mol/min)	1	
Gas	S1 (mol fr)	S1 (mol)
CO_2	4.00E-04	4.00E-04
N_2	0.79	0.79
O_2	0.21	0.21
T (K)	298	
P (atm)	1	

Separation process

STREAM 3:	50% Reduced CO_2	
N3 (mol/min)	1.0	
Gas	S1 (mol fr)	S1 (mol)
CO_2	2.00E-04	2.00E-04
N_2	0.79	0.79
O_2	0.21	0.21
T (K)	298	
P (atm)	1	

$$W_{min} = -RT\left(N_1\sum_{k=1\ldots n}^{n} X_{1,k}lnX_{1,k} - N_2\sum_{k=1\ldots n}^{n} X_{2,k}lnX_{2,k} - N_3\sum_{k=1\ldots n}^{n} X_{3,k}lnX_{3,k}\right)$$

STREAM 1:	Air	
N1 (mol/min)	1	
Gas	S1 (mol fr)	S1 (mol)
CO_2	4.00E-04	4.00E-04
N_2	0.79	0.79
O_2	0.21	0.21
T (K)	298	
P (atm)	1	

Separation process

STREAM 3:	50% Reduced CO_2	
N3 (mol/min)	0.98	
Gas	S1 (mol fr)	S1 (mol)
CO_2	2.04E-04	2.00E-04
N_2	0.79	0.77
O_2	0.21	0.21
T (K)	298	
P (atm)	1	

Concentration factor: 25

STREAM 2:	Enriched CO_2 (1%)	
N2 (mol/min)	0.02	
Gas	S1 (mol fr)	S1 (mol)
CO_2	1.00E-02	2.00E-04
N_2	0.78	1.56E-02
O_2	0.21	4.16E-03
T (K)	298	
P (atm)	1	

0.063 kJ/mol (1 min basis)

Figure 2. CO_2 enrichment from air, targeting two different outlet concentrations—top, 99% purity; bottom, 1% purity.[33]

purified form. Basic immobilized amine sorbents (BIAS) are among the most common sorbent types for capturing CO_2. Wilfong et al.[34,35] have developed and tested a collection of silica-supported BIAS technologies. The quintessential feature of the BIAS that differentiates this sorbent type from other types is the strong affinity of the BIAS' tertiary, -N (humid conditions); secondary, -NH; and primary -NH_2 basic amine groups for acidic CO_2. Strong CO_2–N bonds of adsorbed CO_2, as largely ammonium-carbamates embodied by solid BIAS can give binding energies of around 89 (dry) to 143 (humid, hydronium carbamates) kJ/mol, lending to high CO_2 capture capacities at room temperature at low ppm concentrations compared to lower CO_2 capture capacities of physisorbents, like activated carbon.[36] This high CO_2 affinity makes BIAS ideal for CO_2 enrichment. Strong binding facilitates a "most-at-once" CO_2 release scheme, where a large portion of the adsorbed CO_2 can be released upon reaching a certain minimum higher temperature. This assists in the enrichment of captured CO_2. BIAS form factors include particle, pellet, plus solid and hollow fiber morphologies. The first patented Class 4 BIAS particle sorbent, designated as 32D or T28/P_L12, was comprised of ∼27 wt % polyethylenimine (PEI)/N-(3-(trimethoxysilyl)propyl)-ethylenediamine (TMPED) on silica, with a TMPED/PEI weight ratio of 28/12.[34] The 32D hypothesized structure, shown in Figure 3, exhibits a surface layer of hydrogen-bonded PEI and grafted TMPED, whose anchoring was catalyzed by PEI. A polymerized TMPED network encircling and stabilizing PEI domains through hydroxyl-amine hydrogen bonds is tethered to the surface layer and pervasive throughout the bulk layers. This unique

Table 2. Summary of Recent Studies on CO_2 Capture Sorbents and Processes for Removing and Enriching CO_2 from Point Sources and the Air

sorbent name	material shape	type	reactor configuration	uptake conditions	uptake: (mmol CO_2/g-sorb)	desorption	desorbed CO_2 conc. (%)	ref
MIL-160(Al)	granule/pellet, 1 mm diameter	metal organic framework (MOF)	fixed bed	318 K; 4.5 bar; CO_2/N_2-15/85; 200 s	3.05 mmol/g*h (productivity)	N_2, 0.2 bar	67.3	38
triphenylmethanol	particle	Hyper-cross-linked polymer	fixed bed	40 °C; 25 bar; 20/80-CO_2/N_2; (with preadsorbed water)	6.6 (28.9 wt %)	1 bar	>50	39
TEMED-DQ-CO32-	particle, 38−75 μm	anion exchange resin	chamber	1 bar; 25 °C; 400 ppm of CO_2 air; 620 min	3.41	99% humidity, < 750 ppm of CO_2	not determined	40
	cylindrical pellets, 1−2 mm diameter	amine-functionalized silica/fly ash	TGA	35 °C; 60/40-CO_2/N_2; 30 min	1.3	105 °C, N_2	not determined	41
BIAS	particles	amine-functionalized silica	Microwave cell, 100 W	1 atm; 22 °C; 405 ppm of CO_2/N_2; 24 h	2	microwave (100 W), N_2	1.5−60	42
PB-10	pellets, 30−600 μm diameter	Al-promoted CaO-based	fluidized bed	650 °C; 14.9/3.6/8.8/72.7-$CO_2/O_2/H_2O/N_2$; 20 min	5.4	900 °C, N_2, 5 min	not determined	43
CaO/HcATP	particles	CaO loaded on HCl-modified attapulgite (ATP)	TGA	25 °C; 400 ppm of CO_2/N_2; 30 min	0.67	200 °C, N_2	not determined	44
Lewatit VP OC 1065	particles	benzylamine-functionalized,porous polystyrene	moving bed	18.9 °C; 39% RH; air-515 ppm of CO_2;	1.35	90−105 °C, N_2	3−3.5	45
	flat sheet	amine-functionalized cellulose acetate/silica composite	tubular fiber sheet module	27 °C; 50% RH; air-~475 ppm of CO_2, 4000 min	1.9	100 °C, purge gas	20	46
S-NaY	monolith	zeolite-coated monolith	rotary adsorption system	25 °C; 15% CO_2; 20−30 min	1.2	(a) no vac; 120 °C air; (b) −60 kpa vacuum; 120 °C; air	(a) 54 (b) 79	47
	pellet, 2 mm diameter	activated carbon	microwave cell, 200 W	25 °C; 15% CO_2/N_2	0.81 (30 °C, 0.15 bar)	(a) microwave, 200 W, N_2, 145 °C (b) conventional, N_2, 145 °C	(a) 41.1 (b) 16.1	48
commercial pitch-based active caron fiber (ACF)	not mentioned	ACF	fixed bed configuration coupled with electrodes	pure CO_2	OG-A7 ACF, 0.7	electric swing adsorption, 25 V,	78	49
commercial MAST activated carbon monolith	honeycomb (length: 0.2 m; diameter: 0.02 m)	activated carbon	fixed bed configuration coupled with electrodes	100 kPa, 323.15 K, CO_2/N_2-15/85	1.9	12A, 80 s heating, 600 mL/min N_2 purge	52	50
commercial Norit SC1G activated carbon	3D printed activated carbon monolith	activated carbon	fixed bed configuration coupled with electrodes	CO_2/CH_4-30/70 (biogas mimic)	0.6	150 °C, 8 V, N_2 purge 10 N mL/min for 15 s	87.3	51
AC-NaUSY-70	monolith	hybrid activated carbon and zeolite monolith	fixed bed configuration coupled with electrodes	293.15 k, CO_2 partial pressure 20 kPa	2.23	100 °C, 109 s, 50 W, 600 mL/min N_2 purge	80%	52
CNT-zeolite or SiC-PEI	fibers	hybrid zeolite incorporated nanotube or hybrid PEI impregnated silicon carbide	fixed bed configuration coupled with electrodes	25 C, 15 vol %CO_2	CNT-zeolite: 102.2 SiC-PEI: 8.3	140 °C, 0.5 bar, 80 s	81	53
sorbent-coated carbon fibers	fibers	hybrid sorbent-coated carbon fibers	fixed bed configuration coupled with electrodes	100 sccm of 400 ppm of CO_2, 30 °C, 2 h	0.48	7 V, 1A, 10 min	95	54
3D printed IER/Ag coated glass flakes sorbent	3D printed structure 25 × 25 × 10 mm^3	hybrid amine-based ion-exchange resin and silver-coated glass flakes	fixed bed configuration coupled with electrodes	0.04 kPa, 2 h, 25 °C,	1.48	3A, 70 °C, 1 min	not determined	55
Na_2CO_3 based structured material assembly (MSA)	6″ × 6″ × 3″ MSA	monolith substrate coated with sorbent and heating element	MSA connected with Joule Heating under ESA mode	413 slpm, 50 RH%, 45 min	>3 wt % over 233 cycles	4 slpm N_2, 8−16 °C/min, 100 °C, 2 h	not determined	56

Table 2. continued

sorbent name	material shape	type	reactor configuration	uptake conditions	uptake: (mmol CO_2/g-sorb)	desorption	desorbed CO_2 conc. (%)	ref
PCS–OH	2 cm × 1 cm	OH-charged activated carbon cloth	fixed bed configuration coupled with electrodes	30 °C, 60 min, 400 ppm dry CO_2	0.2	7–8 V, 90 °C, 20 min Joule heating	not determined	57

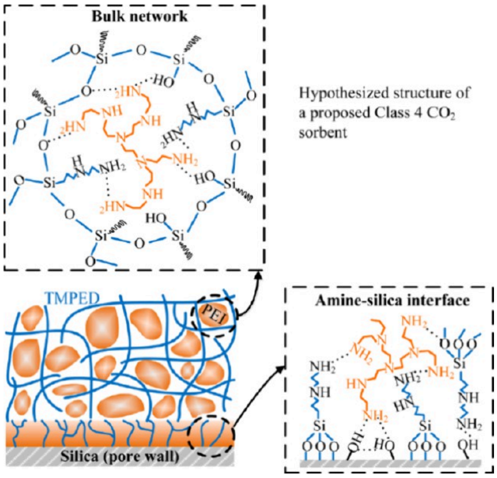


Figure 3. Hypothesized structure of a class 4 BIAS sorbent—T28/P_L12, also known as 32D. Reproduced with permission from ref 34. Copyright 2016 American Chemical Society.

distribution of H-bonded stabilized PEI endowed the sorbent with superior liquid and H_2O vapor stabilities.[35] and stable ~1.8 mmol CO_2/g capture capacity over 100 humid adsorption (50 °C)– desorption (105 °C) cycles.[37] Stability of BIAS, which is directly affected by its chemical composition, during cycling is required to produce economically and reliably enriched CO_2 over the long-term. Therefore, understanding the fundamental formulation characteristics contributing to this parameter is relevant to the topic of CO_2 enrichment. Although CO_2 enrichment was not explored in this work, previous studies with pelletized BIAS materials demonstrated that steam regeneration via a single steam pulse could give an outlet CO_2 concentration of 37 vol % following a previous air pulse purge at the adsorption temperature. Moreover, a concentration of 85% could be reached by a steam pulse following a pure CO_2 pulse purge to remove the other gases.

Advantages of the pellet over the particle shape is the reduced pressure drop resulting from greater void space between the larger sorbent size. Among different binders tested, like poly(vinyl alcohol), polystyrene, and polychloroprene; the latex version of the chlorinated binder was superior regarding strength and processability.[58−60] Shaping the particles into rods or spheres with polychloroprene latex eliminated the complex milling step and subsequent high-temperature dissolution of the pure polymer analog. Pellet binder choice is critical to CO_2 enrichment because it directly affects the pellet's scalability and longevity – ex. polyvinyl chloride degrades the amine during cycling. Lab-scale batches of pellets (10 g/batch) were used to capture CO_2 under simulated postcombustion conditions of 15% CO_2 at 50 °C. Scale-up of pellet production to kg scale was completed by a commercial partner, whereafter the pellets were used to capture 1.2 mmol CO_2/g under simulated landfill gas conditions – 60% CO_2 at 35 °C.[41]

Hollow fiber amine sorbents represent a second evolutionary stage of BIAS technology that further reduce system pressure drop. These fibers are prepared by mixing a Torlon or cellulose acetate-based polymer dope solution, containing solid silica or sorbent, and a bore solution, then extruding both through a dual-outlet hollow fiber nozzle into H_2O to precipitate a solid fiber.[61,62] Hollow fibers enable switchable flows of cooling and heating water through the fiber tube-side to both rapidly

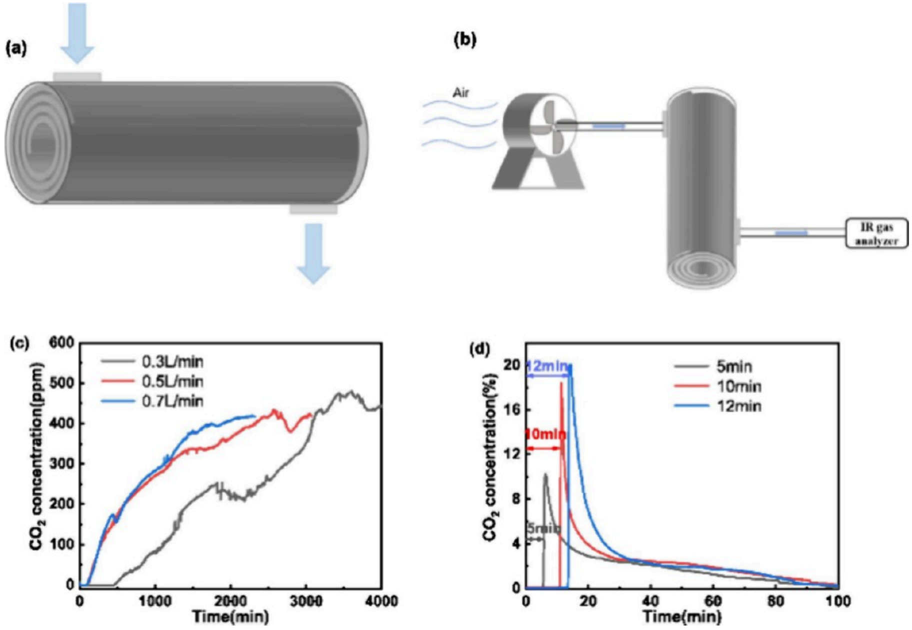


Figure 4. Performance of the spiral-wound membrane module during CO_2 capture from air. Adsorption conditions: 27 °C, 475 ppm of CO_2, 50% relative humidity. Desorption at 100 °C in purge gas flow. Reproduced with permission from ref 46. Copyright 2025 Elsevier.

remove the heat of CO_2-amine adsorption and deliver the heat of CO_2 desorption, respectively.[62] One RTSA process utilizing PEI-based fibers in the literature enriched CO_2 from 13% to ~50% by passing 110 °C water through the fiber, then flowing N_2 to purge desorbed CO_2. Despite this good enrichment, hollow fiber synthesis is complex, and the resulting materials may underperform compared to newer flat sheet materials and their corresponding reactor configurations. These features make the hollow fiber a less desirable form factor for CO_2 enrichment.

Two-dimensional flat sheet sorbents recently gained attention for their easy preparation and low-pressure drop characteristics, amenable to parallel gas flow across plate-like and spiral-wound module configurations. One of the first materials demonstrating this setup utilized a preparation method seemingly derived from fiber sorbent synthesis − casting a cellulose acetate/silica-based polymer dope onto a glass plate, followed by phase inversion of the liquid film in water, then submersion of the precipitated membrane into a PEI-based impregnation solution for amine functionalization.[46] The solid adsorption-based membrane was rolled and placed into a tubular module with parallel direct flow, as shown in Figure 4. The sorbent demonstrated a high 1.9 mmol CO_2/g capture capacity after 4000 min (66.7 h) at 27 °C in air containing 475 ppm of CO_2 and 50% relative humidity. Desorption of adsorbed CO_2 at 100 °C in a purge gas flow enriched the CO_2 concentration from 475 ppm at the inlet to about 20% at the outlet when the sorbent was allowed to age at high temperature for 12 min.

Additional polymers for CO_2 enrichment include metal organic frameworks (MOF's), which represent one of the most widely explored categories of materials used for CO_2 capture. A recently developed and bioderived Al-centered MIL-160(Al) was reported to have an economical scale-up and was shaped/pelletized for practical fixed-bed operation.[38,63] Originally developed for water adsorption, researchers tested it for CO_2 enrichment. CO_2 capture was performed at 318 K with a 4.5 bar CO_2/N_2-15/85 mixture for 200 s. Desorption was performed first with a pure CO_2 rinse step, followed by a blowdown by reducing the pressure from 4.5 to 0.2 bar, then purging with N_2 under the vacuum. This process generated a 67.3% CO_2 stream during desorption, with an overall 99.1% recovery of CO_2. Despite the good CO_2 performance, the affinity of the sorbent for water raises questions about its efficacy under humid conditions, specifically regarding the competitive binding between CO_2 and H_2O.

Metal oxide-based sorbents, like CaO promoted by aluminum (Al) or loaded onto acid-modified attapulgite clay, were used for CO_2 capture under different conditions.[43,44] A typical calcium looping process is predicated upon the simple reaction of gaseous CO_2 with CaO to form $CaCO_3$, releasing about 178 kJ/mol of energy.[64] Similar to the BIAS sorbent, the strong binding of CO_2 for CaO meets an important aspect for successful sorbent-based CO_2 enrichment − high CO_2 affinity and capture and low CO_2 concentrations. In one study, the scalability of an Al-promoted CaO sorbent was demonstrated, and its CO_2 capture performance was validated. Al served as an inert support that resists sintering, which can appreciably reduce the performance of the sorbent. A 6 kg batch of Al-CaO powder was pelletized and tested for cyclic CO_2 capture in a fluidized bed under the following conditions: adsorption − 650 °C in a flowing $CO_2/O_2/H_2O/N_2$-14.9/3.6/8.8/72.7 mixture for 20 min; desorption − 900 °C in flowing N_2 for 5 min. After 30 cycles, the sorbent maintained 94.7% of its initial 5.7 mmol CO_2/g capture capacity. Because of the excessively high adsorption and regeneration temperatures, researchers loaded CaO onto attapulgite clay to reduce both these temperatures and their associated energy consumption values. Because of the high energy cost involved in raising the process temperature to at least 650 °C, the CaO looping process is less attractive than other sorbent processes. Furthermore, although CaO sorbent formulations were adjusted to operate under lower temperatures, these formulations still operate at temperatures higher than most other sorbent processes and have not been fully explored for their long-term feasibility.

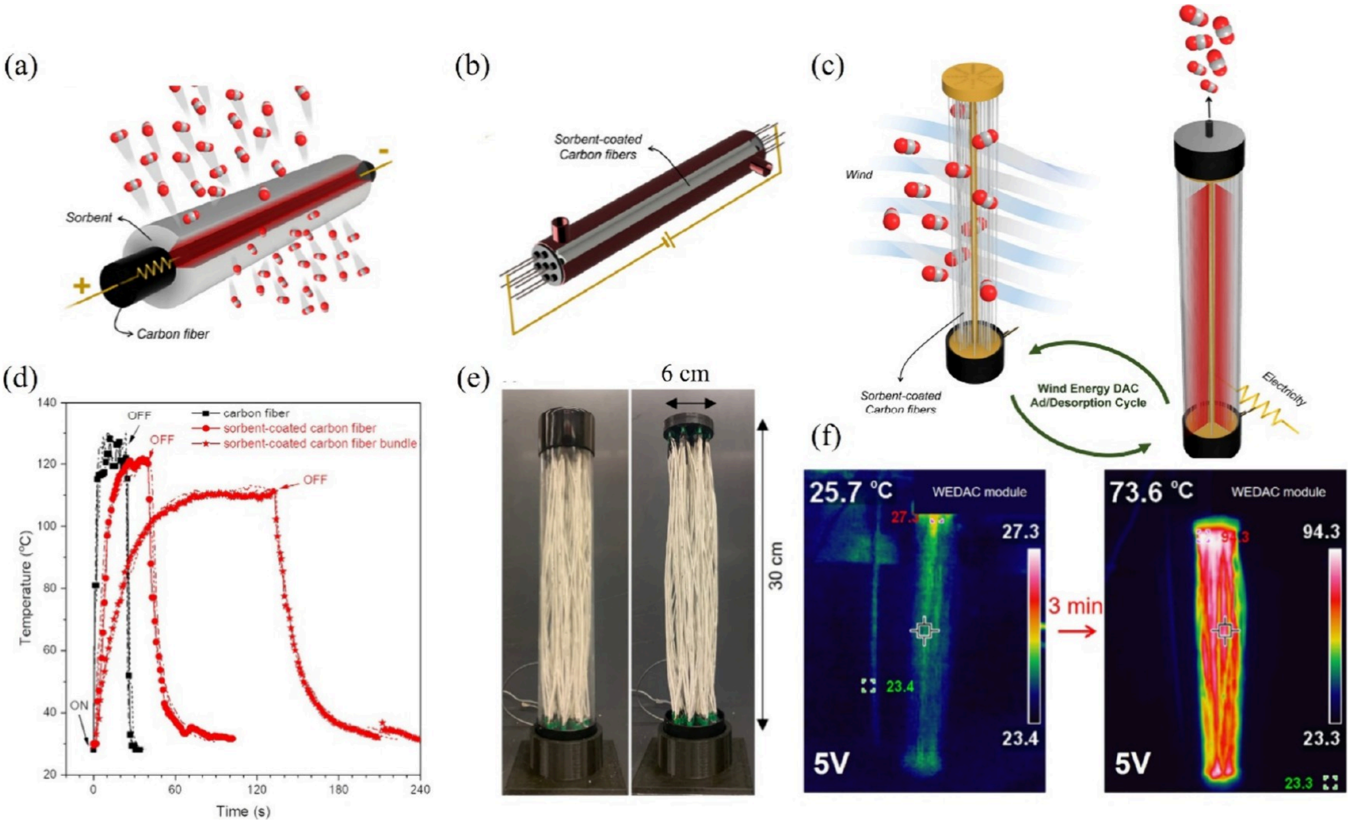


Figure 5. Sorbent-coated Carbon Fibers for DAC applications. (a) Schematic of a single sorbent-coated carbon fiber. (b) Schematic of a lab-scale DAC module for testing fiber performance. (c) Schematic scaled-up WEDAC module operation. (d) Rapid temperature swing of the sorbent-coated carbon fibers by Joule heating. (e) WEDAC module with 120 sorbent-coated carbon fibers in open (left) and closed (right) status for wind-driven adsorption and vacuum-assisted electrothermal desorption, respectively. (f) Thermal images of the open WEDAC module at 5 V for 3 min. Reproduced with permission from ref 54. Copyright 2023 Cell Press.

2.2. Electrified Processes for $LCCO_2$ Enrichment.

Thermal swing desorption (TSD) using a steam sweep is a common method for regenerating CO_2 sorbents. This process involves heating the sorbent bed while introducing steam, which enhances CO_2 release by providing heat and lowering the CO_2 partial pressure. This process is applicable to materials like solid amines and zeolites, where the performance depends on optimizing factors like steam temperature, flow rate, and reactor design. However, challenges exist around balancing water management, sorbent stability, and energy consumption for steam generation. Recent studies demonstrated that electrothermal swing adsorption (ESA) processes like microwave and Joule heating offer significant advantages over steam sweep methods in terms of energy efficiency, process control, system miniaturization, and potential for integration with renewable energy. While challenges related to material properties, scalability, and capital costs remain, these technologies hold great promise for the development of more sustainable and cost-effective $LCCO_2$ applications.

Microwaves take advantage of local molecular polarity, targeting species with a high dielectric loss factor. Contrary to conventional oven heating, microwave heating selectively and rapidly heats only those materials embodying a changing dipole moment, like CO_2 adsorbed ammonium-carbamate ionic species. This molecular feature is the fundamental property of the sorbent that makes microwave-based desorption technically feasible. A 25 wt % novel nonaqueous slurry comprised of PEI-functionalized silica and silicone oil was loaded with a 15% CO_2 stream at 65 °C and irradiated with 300 W to 105 °C for CO_2 release.[65] About 3× as much CO_2 desorption flux was observed for the microwave irradiated sample as was observed for the conventionally heated sample at 105 °C, an even greater 10X flux was observed for the microwaved sample when evaluating desorption at 65 °C. It was estimated that the MW irradiation reduced the apparent activation energy of CO_2 desorption from 31.1 kJ/mol for thermal heating to 24.6 kJ/mol. One important note is that microwave heating for desorption at an elevated temperature of 90−105 °C (95 °C steady state) produced hot and cold zones within the reactor. This inhomogeneous temperature distribution would limit the power output, and hence enrichment capability, of the microwave and could accelerate sorbent degradation. Further work is needed to improve the reactor design and operation of microwave desorption.

Unlike microwave heating, which uses electromagnetic radiation to heat materials volumetrically based on their dielectric properties, Joule heating generates heat selectively within a conductive material by passing an electrical current through it. Fundamentally, a voltage is applied across a resistor, causing electrical current to flow and lose energy as heat while passing through the resistor.[66] Typically, a resistor metal like tungsten or a carbon-based material, is coated or contacted with the sorbent, which releases adsorbed CO_2 quickly after current is initiated. For the process to be successful at a fundamental level, a material must have sufficient maximum dielectric voltage and current density, such that heat is generated fast enough at the applied voltage without failure of the resistor to release CO_2 quickly enough for successful

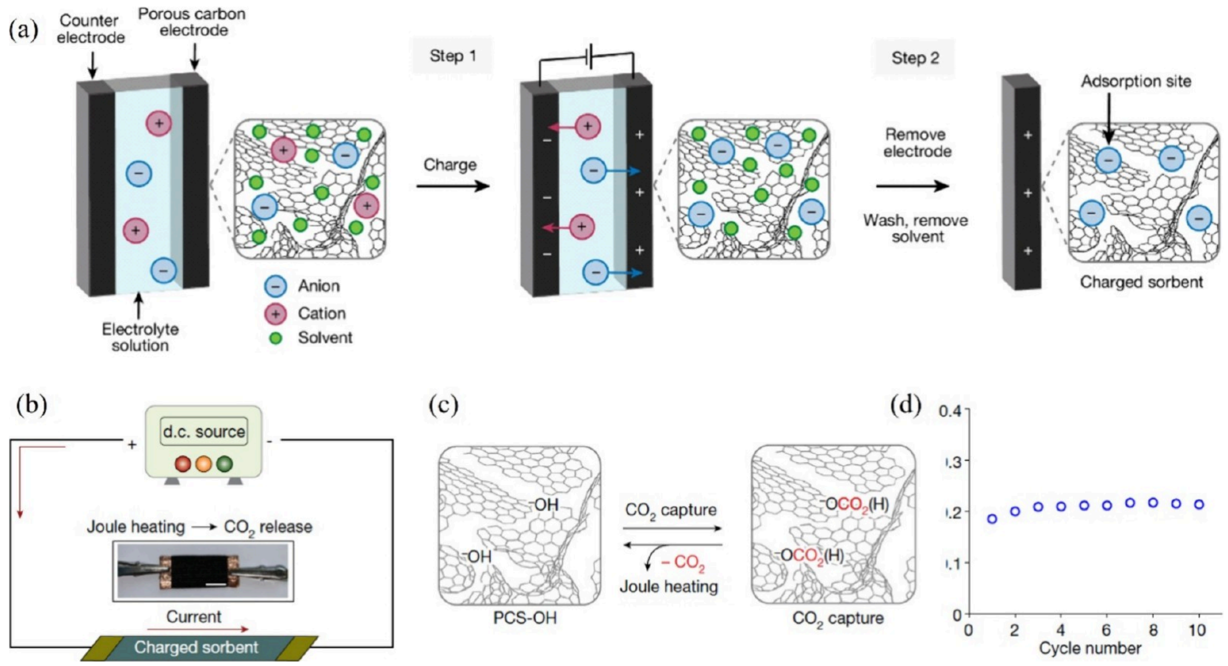


Figure 6. Charged sorbent for CO_2 DAC application. (a) Preparation of charged sorbents. (b) Schematic of Joule heating for CO_2 release (scale bar, 0.5 cm, sample dimensions 2 × 1 cm). (c) Mechanism for CO_2 capture and release by Joule heating. (d) TGA DAC cycling capacities for PCS–OH (adsorption: 30 °C, 400 ppm of CO_2 in dry air; desorption: 130 °C, 60 min). Reproduced with permission from ref 57. Copyright 2024 Nature.

enrichment.[67] This method has emerged as a promising regeneration strategy for CO_2 sorbents due to its rapid response, energy efficiency, and compatibility with structured adsorbent designs.

Activated carbon (AC) based sorbent materials, such as monoliths, fibers, and powders, have been widely explored for joule heating applications due to their intrinsic electrical conductivity, thermal stability, and high surface area. Moon and Shim used activated carbon fiber (ACF) in an early demonstration of joule-heating for CO_2/CH_4 separation.[49] This early proof-of-concept study showed that conductive sorbents could be directly heated via electrical current, enabling efficient desorption of CO_2 without an external heating system. Zhao et al. further employed activated carbon monoliths and investigated the impact of electrical and flow parameters on CO_2 capture performance.[50] The sorbent achieved a CO_2 purity of 52%, a recovery of 76%, and a working capacity of 1.9 mmol/g, under optimal Joule heating conditions (12A, 80 s heating, 600 mL/min purge). Verougstraete et al. introduced several designs in Joule heating sorbent and process.[51] By employing 3D printing in the fabrication of activated carbon monoliths, the resulting sorbents were of good structural flexibility and great thermal response. These improvements resulted in high CO_2 purity and high recovery (up to 85 and 87.3%, respectively), as well as faster and more uniform heating under lower voltages, demonstrating significant progress in practical Joule heating system development.

Novel hybrid or functionalized conductive materials aim to preserve the thermal and electrical advantages of carbon frameworks, while significantly improving CO_2 affinity and separation efficiency. An optimized balance between material conductivity and resistivity must exist to economically convert enough electrical energy to heat energy for CO_2 desorption without overheating and degrading the sorbent. For $LLCO_2$ capture, such designs minimize energy losses and enable faster desorption, a key factor in reducing the cost of continuous operation. Lee et al. presented a multiphase, hierarchically structured Joule-heating sorbent that combines a conductive carbon monolith, a microporous particle coating layer, and amine functionalization (Figure 5).[54] In this architecture, the carbon backbone serves as the electrically conductive substrate for Joule heating. A secondary coating layer was composed of microporous silica particles applied to the monolith surface, onto which PEI was impregnated to introduce chemisorption capability. This design effectively decouples the sorption and heating functions: the PEI provides strong CO_2 affinity via carbamate formation, while the underlying carbon maintains electrical conductivity for rapid heating. Moreover, this work developed a roll-to-roll process for fabricating sorbent-coated carbon fibers. The system achieved ∼1.2 mmol CO_2/g-fiber capacity under simulated ambient CO_2 adsorption conditions and >80% CO_2 purity upon CO_2 release. A techno-economic analysis (TEA) projected a CO_2 capture cost of ∼$160/ton, with only ∼7% of electricity input lost to ambient convection. This study showcases a promising route to scalable, electrically regenerable sorbents optimized for $LCCO_2$ enrichment.

In addition to activated carbon-based conductive frameworks, silver-containing flakes can be added to the sorbent matrix to improve the energy efficiency in Joule-heating systems, even at a relatively low volume fraction.[56]

An illustrative example of a structured chemisorbent system compatible with Joule heating was reported in a US DOE project led by Susteon Inc. (DE-FE0032118).[54] The study developed a modular Structured Material Assembly (MSA) that integrates a Na_2CO_3-based sorbent, a carbon-based resistive heating layer, and a low-pressure-drop cordierite monolith. The sorbent was dispersed onto a high-surface area alumina support and prompted to enhance CO_2 uptake via $NaHCO_3$ formation. Direct Joule heating enabled rapid and energy-efficient regeneration, with heating rates up to 95 °C/min (structured sorbent can reach a temperature of 120 °C in <1 min) and a desorption rate of >0.652 mol CO_2/kg-min (completed in 2 min). For $LLCO_2$ applications, the rapid heating capability improves recovery efficiency from dilute sources. In a 1 kg/day bench-scale test unit, the SMA demonstrated stable performance over 230 adsorption−desorption cycles, maintaining a working CO_2 capacity of ∼ 3.1 wt % using ambient air. The embedded carbon heating layer remained effective over 500 thermal cycles, highlighting the system's mechanical robustness-a critical requirement for long-term $LLCO_2$ operation. TEA projected a CO_2 capture cost of \$349.4/ton with transportation and storage included, with potential reduction to below \$207/ton through further improvements in adsorption rate, capacity, manufacturing, and risk-reduction methods. These cost figures are especially relevant for LLCO2 utilization scenarios, where the economic viability is sensitive to regeneration energy demand, sorbent lifetime, and large-scale production feasibility. This work highlights the feasibility of integrating alkali-based chemisorbents and embedded conductive structures for scalable, low-energy DAC applications.

Beyond conventional Joule-heating composites that combine conductive substates with functionalized sorbents, recent advancements have introduced intrinsically charged sorbents that integrate sorption sites and Joule-heating frameworks in a single porous architecture. Charged sorbents work by electrochemically inserting reactive ions (e.g., hydroxide) into the pores of conductive substrates that enable field-driven CO_2 adsorption and desorption, often regenerated at low voltages. Li et al. demonstrated hydroxide-functionalized sorbents regenerated from ambient air via low-voltage operation.[57] Figure 6 shows that the sorbent was prepared using a battery-like charging setup in which a porous carbon electrode was immersed in an aqueous electrolyte (KOH solution) and positively polarized. The confined OH- ions act as reactive sites for CO_2, forming bicarbonate or carbonate species via chemisorption at low partial pressures. Regeneration is achieved using direct Joule heating of the conductive matrix, which rapidly releases CO_2 at ∼90 to 130 °C. The resulting material demonstrated a capacity of 0.2 mmol/g under DAC conditions and excellent cycling stability.

One important aspect of joule- and microwave-heating is sorbent degradation, primarily oxidation of BIAS-based formulations. Oxidation is expected to occur primarily to the amines either functionalized within the porous solid supports (microwave) or coated on the conductive solid matrix (joule). Localized heating, owing to the nonuniform electromagnetic field strength within the microwave chamber, is the accelerating cause of sorbent oxidation within this system. Overheating for joule units due to improper current control, or an unexpected increase in resistance of the heating element, both of which would generate more heat, are expected to accelerate amine oxidation. Several studies investigating amine oxidation explored the molecular mechanisms causing breakdown of the amine structure into forms that have minimal CO_2 affinity.[68] Typically, amines are transformed into amides and imides while releasing ammonia and CO_2 as gaseous byproducts.[69] Oxidation of amines was found to be dependent not only on temperature, but also on O_2, CO_2, and H_2O concentrations.

3. $LCCO_2$ UTILIZATION FOR AGRICULTURE AND FOOD APPLICATIONS

3.1. Direct $LCCO_2$ Utilization for Enhanced Crop Growth. CO_2 is a fundamental building block for plant life, serving as the primary carbon source for photosynthesis. In natural environments, however, CO_2 concentrations can often be a limiting factor for plant growth, particularly in densely vegetated areas or under conditions of high photosynthetic demand. The advent of controlled-environment agriculture (CEA), including greenhouse and vertical farming systems, has allowed for the manipulation of environmental factors, including CO_2 concentration, to optimize crop yields. While CO_2 enrichment (typically to levels of 800−1200 ppm) is a well-established technique[70,71] the source of CO_2 is often derived from fossil fuel combustion, creating a sustainability challenge.

In greenhouse agriculture, the most widely adopted CO_2 enrichment approach involves the combustion of fossil fuels such as natural gas, propane, or biomass, inside or near the greenhouse, to generate $LCCO_2$.[72−74] In themselves, processes inherently generating $LCCO_2$ effluents, e.g., coal combustion, require no further concentration and can be considered as self-enriching. This inherently enriched CO_2 is achieved by introducing flue gas from combustion sources (typically containing less than 20% CO_2 by volume) into greenhouses, either through ventilation systems or by direct injection. Although this method requires careful monitoring and control of the flue gas composition to prevent the introduction of harmful substances, it can be a more cost-effective option compared to using pure CO_2 sources, particularly if a suitable flue gas source is readily available nearby.

Direct CO_2 enrichment, using solid sorbent or liquid solvent technology to capture CO_2 directly from the ambient air (∼400 ppm), involves injecting the CO_2 into the greenhouse environment after capture. This method offers precise control over CO_2 levels and a sustainable CO_2 source, but can be energy-intensive and costly depending on the DAC technology employed. Novel approaches include the integration of CO_2 enrichment with novel structured materials, solar energy, heat pumps, or energy storage technologies[75] to improve efficiency. Figure 7 shows direct greenhouse CO_2 enrichment using a metal−organic framework (MOF) integrated with solar cells and heat pumps.[76] According to a recent techno-economic analysis (TEA)[77] of a zeolite 13× sorbent-based greenhouse CO_2 enrichment process, the $LCCO_2$ enrichment cost could potentially be as low as \$15 per ton. However, these cost estimates are sensitive to assumptions concerning materials costs, sorbent lifetime, process efficiency, and local energy prices.

3.2. Electro-Agriculture Conversion of $LCCO_2$ for Food Production. Recent studies[78] suggest that electro-agriculture (electro-ag), as shown in Figure 8, is revolutionizing food production by integrating CO_2 electrolysis with biological systems, offering a transformative alternative to the inherent efficiency limitations of photosynthesis.

The core system consists of vertically stacked layers, incorporating solar photovoltaics for renewable energy, tandem CO_2 electrolyzers that convert CO_2 into acetate, and heterotrophic cultivation systems where crops consume the acetate in the absence of light. This innovative design promises several benefits: it boosts energy efficiency by at least 4-fold, drastically reduces land usage by up to 88%, and stabilizes food

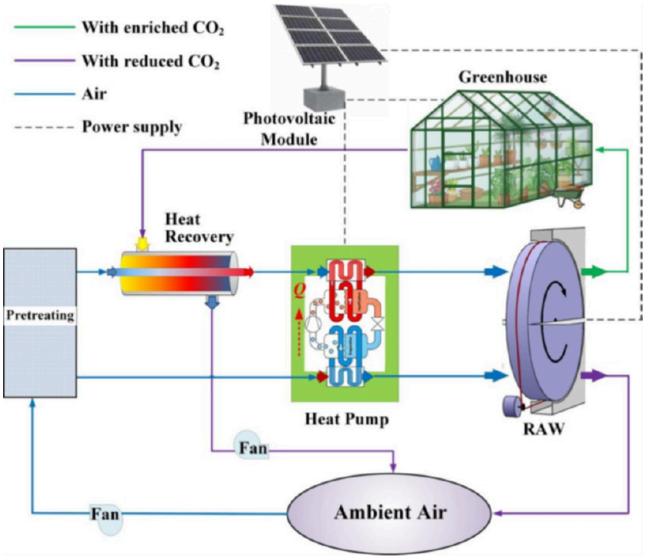


Figure 7. Schematic diagram of a greenhouse CO_2 enrichment system utilizing a rotary regenerative adsorption wheel. Reproduced with permission from ref 76. Copyright 2022 Elsevier.

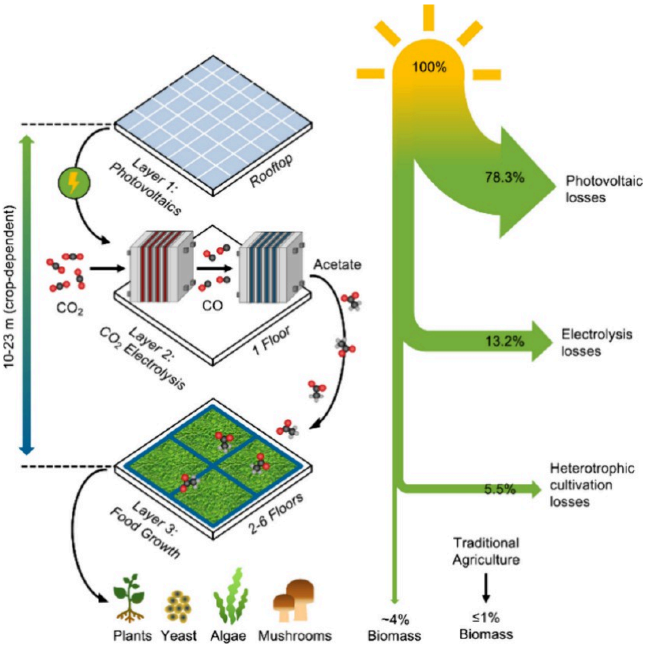


Figure 8. Schematic of a vertical electro-ag system and energy efficiency. Reproduced with permission from ref 78. Copyright 2024 Cell Press.

prices by lessening reliance on weather behavior. The use of enriched $LCCO_2$ in electro-ag presents a promising yet still developing solution for food production.

Further studies by Wang et al.,[79] indicate that electro-biological hybrid systems present a promising alternative for producing protein from CO_2, water, and electricity—bypassing the need for photosynthesis, as shown in Figure 9. This approach, using meat production as a case study, demonstrates a sequestration potential of 13.11 kg of CO_2 per kilogram of protein produced, showcasing its capacity to meet global protein needs and its potential for substantial resource savings compared to traditional agriculture. Replacing all U.S. beef protein production with electro-agriculture could result in a net removal of approximately 308.23 million metric tons of CO_2 annually.

3.3. Indirect $LCCO_2$ Utilization via Enhanced Weathering for Crop Production. Enhanced weathering (EW) accelerates natural geological processes by reacting enriched $LCCO_2$ with finely ground silicate rocks, such as basalt, to form stable carbonate minerals. When applied to agricultural lands, EW offers multiple cobenefits beyond carbon removal. It improves soil quality by regulating pH levels and releasing essential nutrients, which can reduce the reliance on chemical fertilizers. Additionally, EW contributes to improved air quality by lowering surface ozone and particulate matter levels, which in turn can increase crop yields—particularly for maize, soybean, and wheat—resulting in substantial economic gains through avoided yield losses.

Building on a series of studies by Beerling and colleagues,[80−82] EW has emerged as a scalable and promising carbon dioxide removal strategy with agronomic benefits. Their pioneer study[82] demonstrated the global potential of applying ground silicate rocks to croplands to enhance chemical weathering, sequester CO_2, and improve soil health. In 2024, a follow-up study examined the U.S. Corn Belt, showing that EW can simultaneously boost crop productivity and reduce fertilizer runoff. Most recently,[80] they quantified the national-scale impact, indicating that widespread adoption of EW across U.S. agriculture could achieve gigaton-scale CO_2 removal while promoting sustainable farming practices (Figure 10). These studies demonstrated that EW primarily involves the application of crushed silicate rocks to agricultural soils to capture atmospheric CO_2 via natural weathering. There is no indication that high-purity CO_2 is required for this process, suggesting that EW can effectively utilize $LCCO_2$ streams, enriched from direct air capture or point sources.

Reported concern for the kinetics of natural enhanced weathering prompted studies into a more engineering approach. Xing et al. conducted a modeling assessment of a counter-current trickle flow packed bed reactor process, involving calcite dissolution in a CO_2-water solution.[83] Among the studied parameters, CO_2 concentrations in the range of 410 ppm to an enriched 20% were evaluated. Interestingly, results showed that raising the CO_2 concentration from 410 ppm to 0.5% lowered the energy and water consumption by 80%. Using even more enriched CO_2 feedstock was shown to further decrease the demands of these two parameters. Although utilizing enriched CO_2 for enhanced weathering has been rarely used, if at all, these results give strong support for advancing a technology that uses an enhanced form of a natural process.

4. INTEGRATED REACTIVE CAPTURE FOR $LCCO_2$ VALORIZATION TO FUELS AND CHEMICALS

Traditional carbon capture and storage (CCS) strategies have largely focused on the permanent sequestration of CO_2 to prevent its release into the atmosphere.[84] However, growing interest in circular carbon economies has led to the emergence of carbon capture and utilization (CCU) pathways,[85−87] in which CO_2 is activated and transformed into value-added products via tailored catalysts and integrated process systems.[86] While some of these efforts target high-purity CO_2 streams, increasing attention is being paid to low-concentration CO_2 sources such as ambient air or flue gas, which pose additional kinetic and thermodynamic challenges. As reported in a recent study,[88] Figure 11a compares the

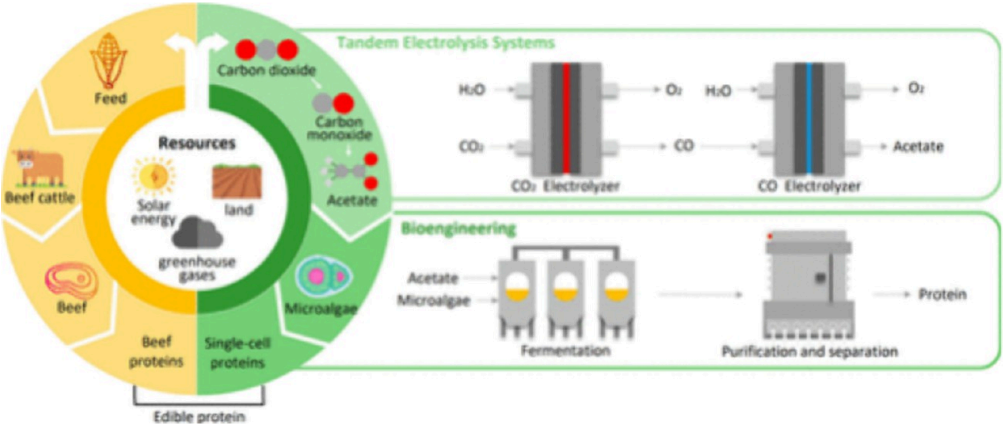


Figure 9. Schematic of the conventional agricultural pathway (yellow background) and the electro-agriculture pathway (green background). Reproduced with permission from ref 79. Copyright 2025 American Chemical Society.

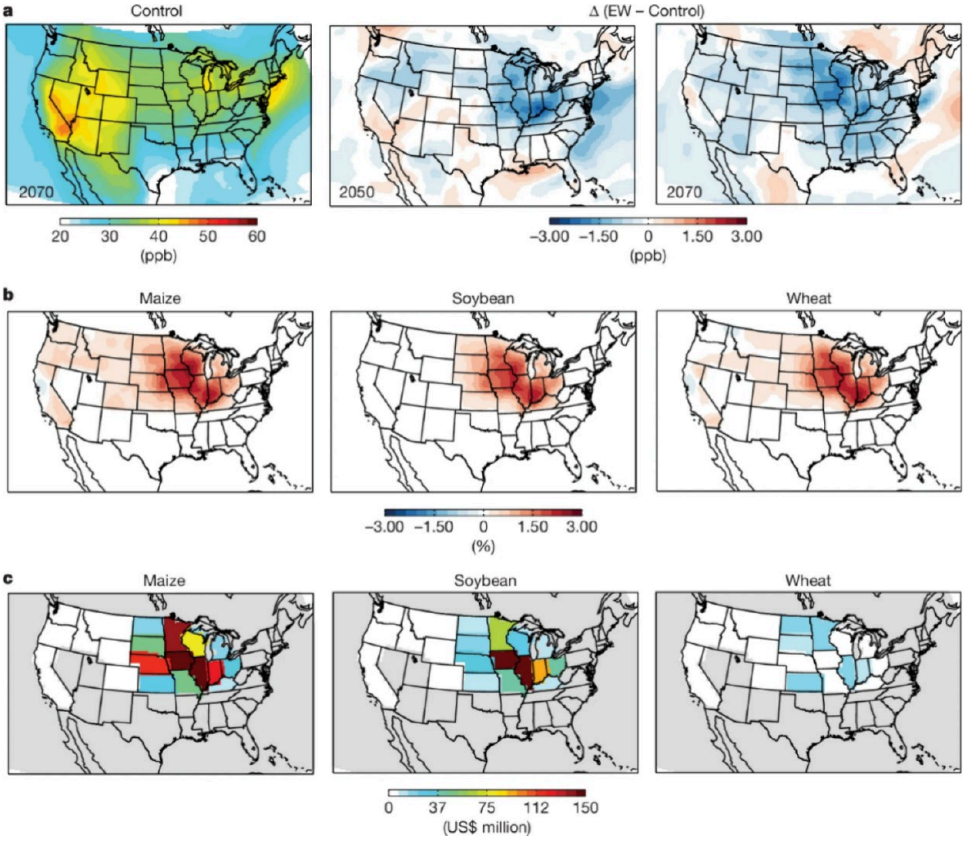


Figure 10. Benefits of enhanced weathering for surface ozone and crop production. (A) Simulated summer surface ozone (O3) for 2070 (control; anthropogenic emissions + biomass burning + present-day biogenic emissions, no EW effects), with widespread reductions by 2050 and 2070 due to EW lowering soil nitrogen trace gas emissions. (B) Average calculated increases in yields of maize, soybean, and wheat for 2070 of three ozone exposure–crop yield functions. (C) Calculated avoided economic yield losses for maize, soybean, and wheat per state due to lower surface O3 exposure levels in 2070 with EW. Reproduced with permission from ref 80. Copyright 2025 Nature.

energy intensity for methanol production from air and H_2 using two methods: DAC followed by CO_2 conversion, and reactive capture. This analysis, based on solid sorbent DAC systems, indicates that reducing energy intensity in CO_2 capture and conversion directly lowers the carbon intensity of the final product. The reactive capture method could reduce methanol production energy intensity by approximately 50% compared to the DAC and conversion processes. Similarly, Figure 11b estimates potential capital cost savings for the reactive capture approach in methanol production. Using a solvent-based DAC system and thermochemical CO_2-to-methanol conversion, reactive capture is estimated to yield capital expenditure savings of $23 per metric ton of methanol produced over the plant lifetime, representing a 38% reduction compared to DAC and conversion.

As illustrated in Figure 12,[22] overcoming the low CO_2 conversion efficiency requires systems that can both enrich $LCCO_2$ locally and drive catalytic reduction reactions under solar or electrochemical energy inputs. In such configurations, porous materials, photoactive semiconductors, or electro-

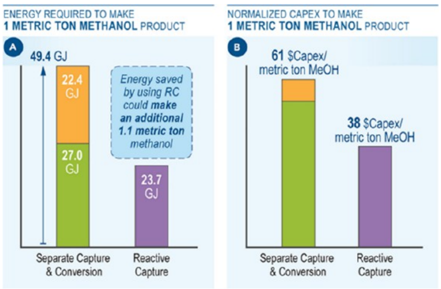


Figure 11. Assessment of reactive capture versus DAC and conversion for methanol production. (a) Energy required to produce 1 metric ton of methanol. (b) Normalized capital cost to produce 1 metric ton of methanol. In both panels (A) and (B), green bars represent the capture portion, orange bars represent the conversion portion, and purple bars denote reactive capture. Reproduced with permission from ref 88. Copyright 2023 Cell Press.

catalytic frameworks are codesigned to achieve simultaneous $LCCO_2$ enrichment, concentration, and conversion, forming the basis of modern low-carbon chemical platforms.[89]

From a theoretical perspective, the valorization of $LCCO_2$ requires overcoming both thermodynamic and kinetic constraints inherent to dilute systems. At low partial pressures, the chemical potential of CO_2 is significantly reduced, resulting in limited driving force for adsorption and subsequent reaction. Moreover, the low collision frequency of CO_2 molecules suppresses the intrinsic reaction rate. To address these challenges, functional materials are designed to locally enrich and activate CO_2 molecules. Basic oxides and alkali promoters stabilize CO_2 through carbonate or bicarbonate formation, while oxygen vacancies and surface defect sites lower the activation barrier for C═O bond cleavage.[90−92] In addition, single-atom catalytic centers and tailored metal−support interactions provide highly localized electronic environments that facilitate proton-coupled electron transfer and enhance catalytic turnover under dilute conditions.[91−94] Beyond material design, energy-coupled systems—such as photothermal, electrochemical, or plasma-assisted processes—introduce external fields that supply the activation energy otherwise absent in low-pressure environments.[19,95] Together, these theoretical principles explain why emerging materials and integrated processes can achieve effective CO_2 capture and conversion even from highly dilute sources.

Among the most promising approaches are systems that enable in situ conversion of $LCCO_2$ streams, such as those found in industrial flue gas or high-purity CO_2 reservoirs. These approaches rely heavily on the coevolution of functional materials, including hybrid adsorbents, catalytic frameworks, and multifunctional composites. These not only bind CO_2 effectively, but also activate it toward reduction, coupling, or insertion reactions.[96] Concurrently, process-level advancements, such as solar-assisted reactors, dual-zone catalytic beds, and electrified DACCU (Direct Air Capture and CO_2 Utilization) systems, provide a technological foundation for the practical deployment of such materials.[19]

The following section reviews recent progress in this field, focusing first on reactive $LCCO_2$ capture platforms (Section 4.1), which integrate separation and transformation into a single functional material or device. It then transitions to novel catalytic systems for in situ chemical synthesis using concentrated CO_2 (Section 4.2), highlighting structure−activity relationships, tandem reaction schemes, and emerging hybrid technologies.[20] Together, these advances represent a significant step toward energy-efficient, modular, and scalable CO_2 utilization strategies.[97]

4.1. Reactive Capture Technologies for $LCCO_2$.

The traditional approach to CO_2 utilization is largely modular: CO_2 is first separated from dilute gas streams, purified, compressed, and then subjected to chemical conversion.[90] Each step is energy- and capital-intensive, particularly when dealing with low partial pressures such as ambient air or postcombustion

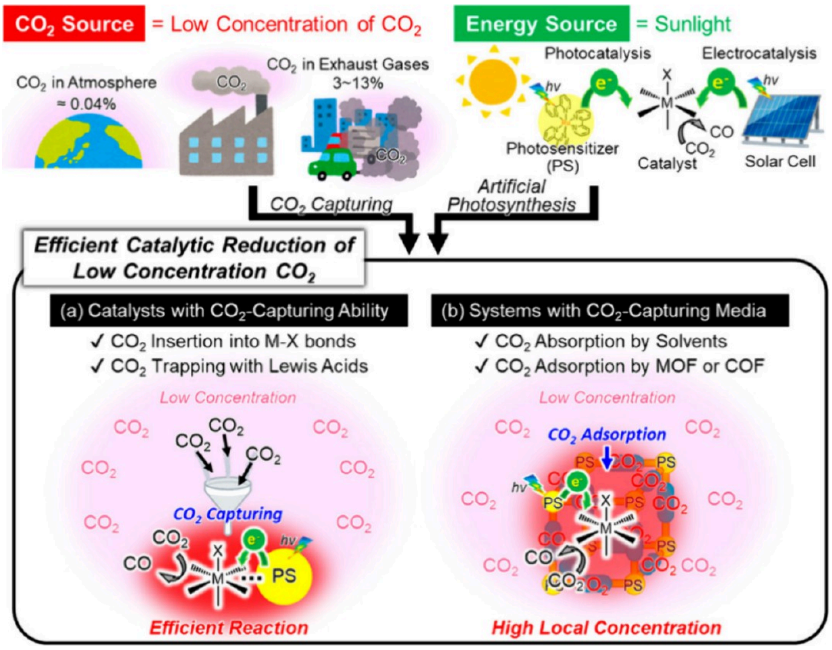


Figure 12. Conceptual representation of catalytic strategies for reducing $LCCO_2$ (0.04−13%) by integrating adsorption and activation pathways. Local $LCCO_2$ enrichment is achieved via porous or functionalized materials, enabling photothermal or electrochemical reduction under visible light or solar energy. Reproduced with permission from ref 22. Copyright 2022 American Chemical Society.

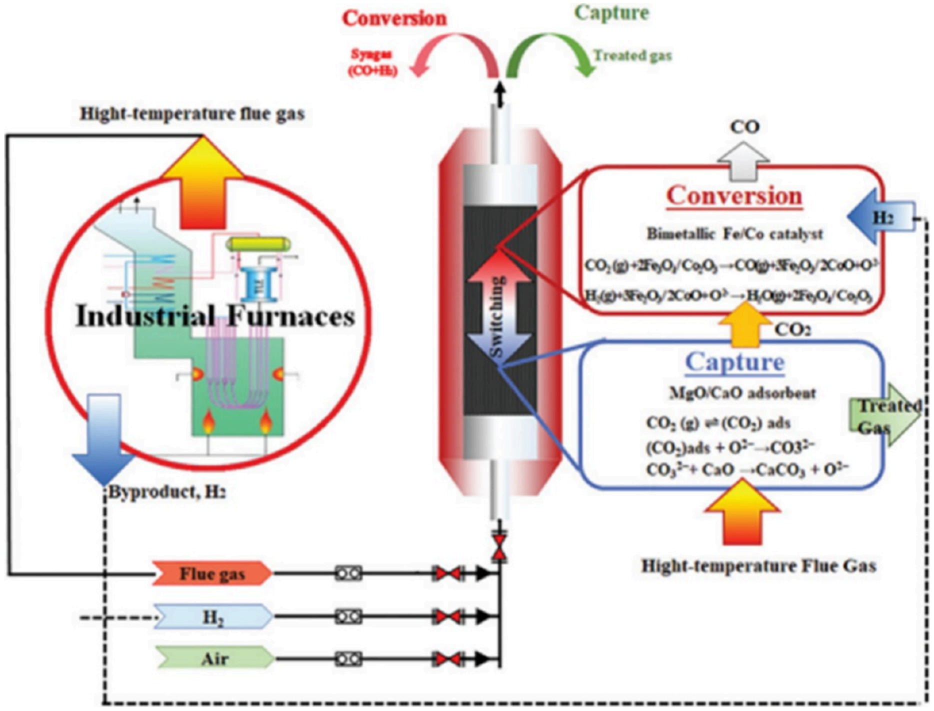


Figure 13. Integration of CO_2 capture and in situ conversion in one fixed-bed column with high-temperature industrial flue gas (for example, from ethylene manufacturing). Reproduced with permission from ref 90. Copyright 2021 Royal Society of Chemistry.

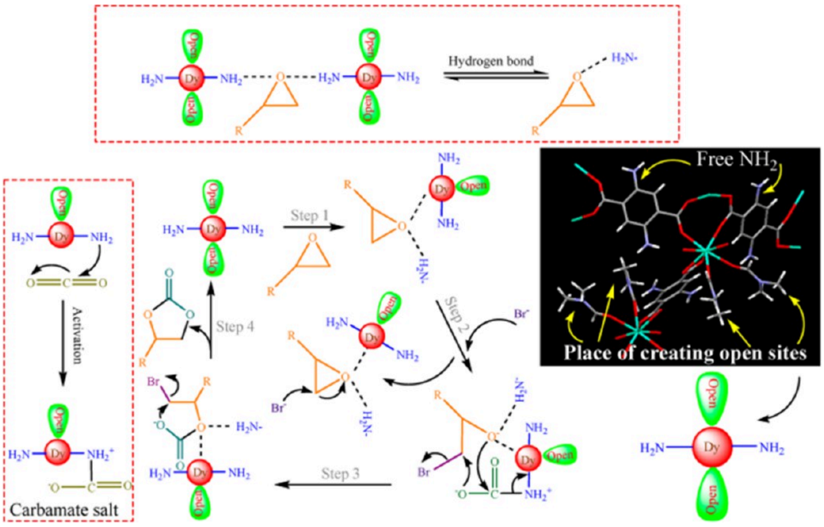


Figure 14. Simplified schematic representation of a possible mechanism for the cycloaddition of CO_2 to epoxide catalyzed by NH_2-TMU-73. Reproduced with permission from ref 102. Copyright 2021 American Chemical Society.

flue gases.[98] In response, the concept of reactive capture for $LCCO_2$ has emerged as a process intensification strategy that seeks to integrate adsorption and conversion into a single, multifunctional system. This approach reduces energy input, simplifies system architecture, and enables localized carbon valorization, particularly in scenarios where energy efficiency and modularity are critical.

Reactive capture for $LCCO_2$ systems functions by combining materials that can both adsorb CO_2 and catalyze its subsequent transformation. These platforms fall into three major categories. First, dual-function materials (DFMs)[90,99] physically integrate a CO_2-binding component,[100] typically a basic oxide such as CaO or MgO, with a transition metal catalyst (e.g., Ni, Ru, Fe)[100,101] that facilitates downstream hydrogenation reactions. Second, hybrid porous materials, such as metal−organic frameworks (MOFs)[102,103] and covalent organic frameworks (COFs),[104] can host both CO_2-philic functional groups (e.g., amines, Lewis bases) and active catalytic centers (e.g., transition metals or metal clusters). Third, system-level integrations, including solar-assisted or electrochemically driven modules, combine CO_2 capture and utilization across colocated unit operations powered by renewable energy.[18,105]

The first and most established strategy involves dual-function materials (DFMs),[99] typically based on basic metal oxides such as CaO or MgO that serve as CO_2 sorbents through carbonate formation. These oxides are physically or chemically combined with transition metal catalysts—such as

Ni, Fe, or Ru—that facilitate hydrogenation reactions. One illustrative case is the Fe−Co/CaO composite developed by Hu et al.,[90] as shown in Figure 13, which captures CO_2 from flue gas via calcium looping and concurrently converts it into CO through the reverse water−gas shift (RWGS) reaction at 650 °C. The system exhibits excellent cycling stability and exploits the thermal energy present in flue gases to drive both sorption and reaction steps, highlighting its potential for industrial integration.

A second approach focuses on porous hybrid frameworks, particularly metal−organic frameworks (MOFs)[102] and covalent organic frameworks (COFs), which offer molecular-level tunability for hosting both adsorption and catalytic sites. In these materials, functional groups such as amines or carbonyls act as CO_2-binding motifs, while embedded metal centers (e.g., Zn^{2+}, Dy^{3+}, Ru nanoparticles) serve as catalytic sites. For instance, Abazari et al. (2021) reported a Dy(III)-based MOF containing both open metal sites and pendant amine groups, enabling cooperative CO_2 capture and chemical activation, as shown in Figure 14.[102] Similarly, Lu et al. reviewed a range of COFs with metalloporphyrin or ketoamine building blocks that exhibit high activity in photocatalytic CO_2 reduction under visible light.[104] These frameworks are particularly suited to low-pressure CO_2 environments and solar-driven conversion processes. Though challenges remain in maintaining their chemical and thermal stability under reactive conditions, as shown in Figure 15.[104]

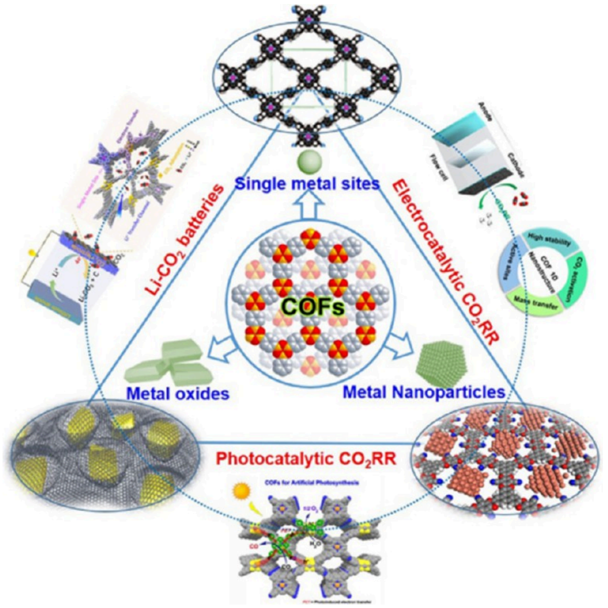


Figure 15. Synthesis of COF-based functional materials and chemical conversion of CO_2 using COF-based functional materials as catalysts. Reproduced with permission from ref 104. Copyright 2022 Wiley.

A third and increasingly prominent strategy in reactive CO_2 capture involves system-level integration, where spatially and functionally distinct capture and conversion units are combined into a modular, flow-through architecture. This is exemplified by solar-assisted direct air capture and utilization (DACCU) systems, which employ solid amine-functionalized sorbents, such as polyethylenimine (PEI) grafted onto mesoporous SBA-15, to extract CO_2 from ambient air. As illustrated in Figure 15, the captured CO_2 is subsequently released through mild thermal regeneration, often using solar-derived heat, and fed directly into a photothermal catalytic reactor for transformation into CO or other intermediates. The CO_2 utilization (CO_2U) unit typically incorporates visible-light-responsive catalytic materials such as cobalt porphyrins supported on $TiO_2/SiO_2-Al_2O_3$ composites, enabling solar-driven hydrogenation reactions under continuous flow.

This dual-bed design offers both diurnal adaptability and system modularity. During nighttime operation, the DAC unit operates passively, adsorbing atmospheric CO_2 under ambient conditions. In the daytime, solar concentrators activate the CO_2U unit and regenerate the sorbent, completing a self-contained, carbon-neutral loop.[18] The clear physical separation between the DAC and CO_2U units (Figure 16a−d) enables thermal and material optimization in each subdomain, while maintaining efficient coupling at the system level.[18]

A related and more advanced configuration is depicted in Figure 17, which shows a solar photovoltaic−thermal hybrid (PV-T) platform for fully renewable CO_2-to-methanol synthesis.[105] In this approach, solar thermal energy regenerates the DAC module, while photovoltaic electricity powers water electrolysis to generate green hydrogen. The captured CO_2 and hydrogen are then combined in a thermos-catalytic hydrogenation reactor to produce electro-methanol. The system is compact, scalable, and grid-independent, offering strong potential for the distribution of fuel production and off-grid carbon utilization.

Together, these integrated systems represent a powerful evolution of reactive CO_2 capture—extending the concept from material-level innovation to process-level architecture, where solar energy not only drives sorbent regeneration and reaction chemistry but also enhances overall system autonomy. While still in early development, such designs demonstrate a feasible pathway toward decentralized, carbon-neutral chemical manufacturing enabled by renewable energy and air as the only feedstock.

Despite their different structural embodiments, these three approaches share common challenges. Chief among them is the need to balance adsorption strength with catalytic turnover, as strong CO_2 binding can hinder subsequent reaction steps. Material stability under cycling conditions, especially in the presence of moisture and flue gas contaminants, remains a critical limitation—particularly for MOFs and COFs.[102,104] Moreover, effective thermal and kinetic matching between the sorption and conversion steps is essential to maximize efficiency. Reactor design, material shaping, and techno-economic viability will also influence the success of each strategy as these systems move toward industrial deployment.

Beyond laboratory-scale research, several companies have already advanced reactive capture concepts toward pilot and demonstration levels. For example, Climeworks has operated demonstration plants such as Orca in Iceland, with a capture capacity of about 4000 tCO_2 per year, representing one of the first large-scale field deployments of DAC technology. Similarly, Carbon Engineering commissioned a DAC pilot plant in Squamish, Canada, which captures approximately 1 tonne of CO_2 per day and integrates with hydrogen electrolysis and fuel synthesis modules to produce liquid fuels. These cases illustrate that the transition from laboratory experiments to early demonstration is already underway, underscoring the practical relevance of reactive capture approaches for diluted CO_2 streams.

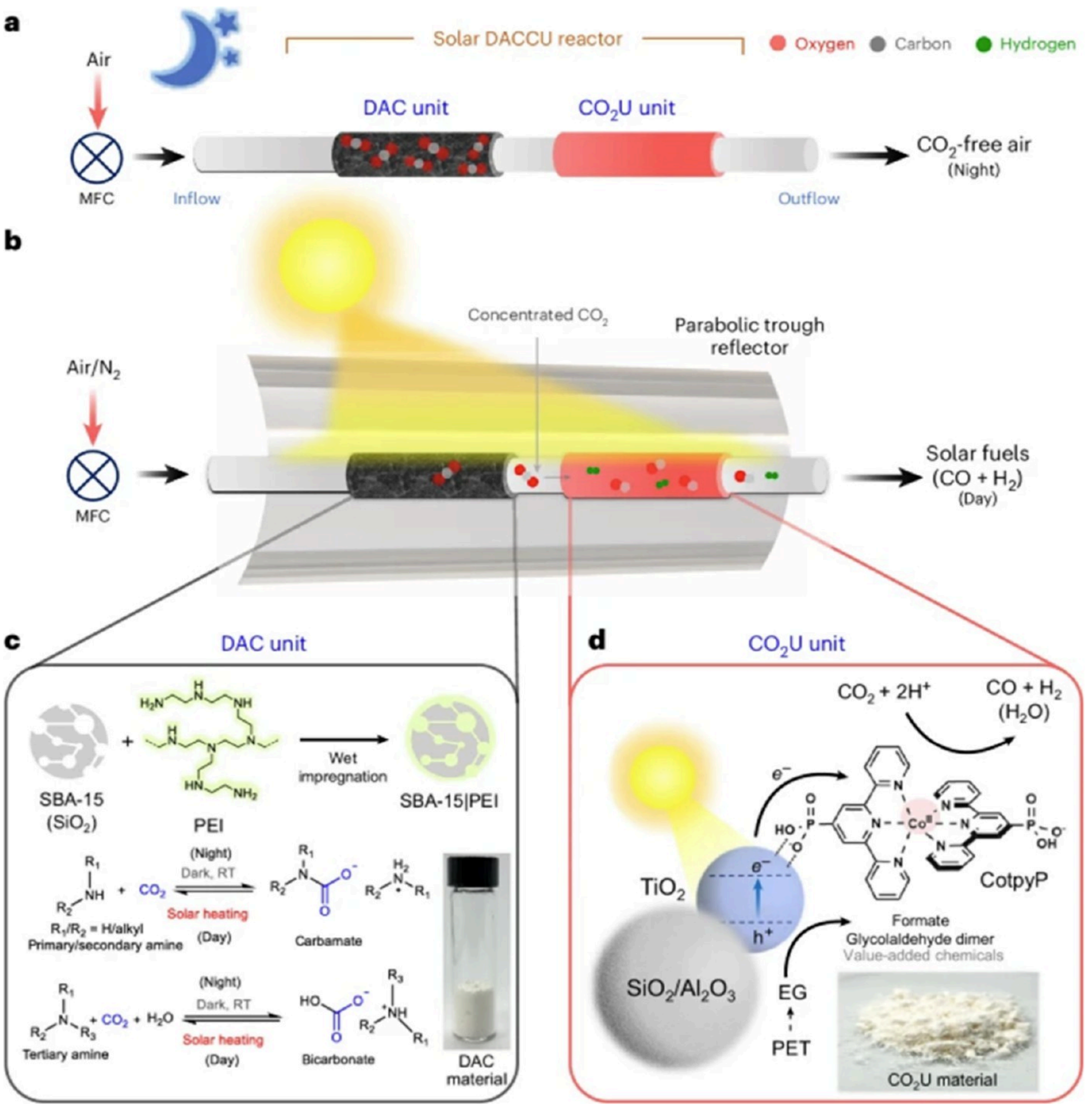


Figure 16. DACCU through a dual-bed flow reactor consisting of DAC and CO_2U units. Reproduced with permission from ref 20. Copyright 2025 Springer Nature.

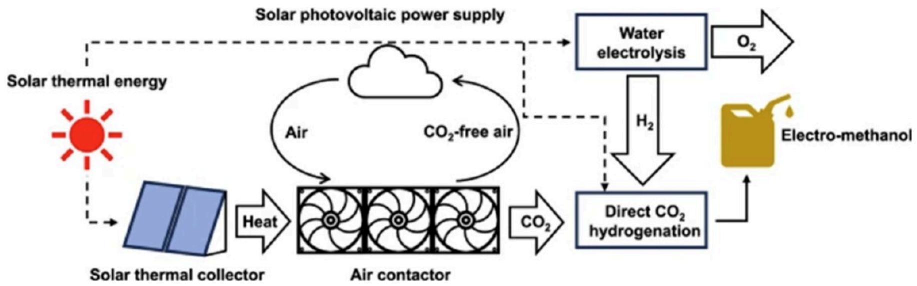


Figure 17. Schematic diagram of solar thermal energy-assisted DAC for sustainable CO_2-to-methanol transformation. Reproduced with permission from ref 106. Copyright 2024 Springer Nature.

In summary, reactive CO_2 capture represents a transformative shift from traditional carbon capture and storage toward active, integrated utilization. By unifying adsorption and catalysis within tailored materials and systems, this approach enables more efficient use of low-grade CO_2 sources while opening pathways to renewable and distributed chemical production. Continued innovation in material chemistry, interface engineering, and process integration will be essential to unlocking the full industrial potential of this promising concept.

4.2. Novel Catalysts for In Situ Synthesis Using $LCCO_2$. The efficient transformation of $LCCO_2$ into valuable chemicals and fuels represents a critical pillar in the future carbon-neutral chemical industry.[107] Industrial gas streams—such as syngas conditioning, biogas upgrading, and CO_2-rich flue emissions—offer readily available CO_2 sources with elevated partial pressures. This allows for in situ chemical synthesis without the need for intermediate separation or purification.[97,108] To meet the demands of this reaction environment, recent efforts have focused on engineering catalysts that are highly active, selective, and robust under realistic conditions.[109]

A promising strategy for CO_2 valorization under capture-integrated conditions involves the use of single-atom catalysts (SACs) with well-defined local environments.[94] In a recent study, Neves-Garcia et al. demonstrated that atomically dispersed nickel (Ni) species, when selectively deposited on conductive substrates such as Au or CNTs, can directly convert CO_2 carbamate species—the primary product of amine-based CO_2 capture—into methane under electrochemical conditions.[10,93] This integrated platform bypasses the energy-intensive CO_2 stripping step, enabling one-step electro conversion from capture solution to fuel.

Through a combination of operando spectroscopy, isotope tracing, and control experiments, the authors established that CH_4 production is strongly correlated with the presence of carbamate, and not with dissolved CO_2 or bicarbonate.[110] Density functional theory (DFT) calculations further confirmed that on Ni single-atom sites (e.g., Ni@Au), carbamate undergoes a favorable proton-coupled electron transfer pathway, leading to CH_4 formation via sequential C−O and C−N bond cleavage steps. Notably, the catalytic activity and turnover frequency of SACs were significantly higher than those of bulk Ni surfaces, underscoring the unique reactivity of dispersed Ni atoms (Figure 18).[93] These findings mark the first experimental validation of direct hydrocarbon production from carbamate on a well-defined SAC, offering a scalable route for coupling CO_2 capture and conversion.[111]

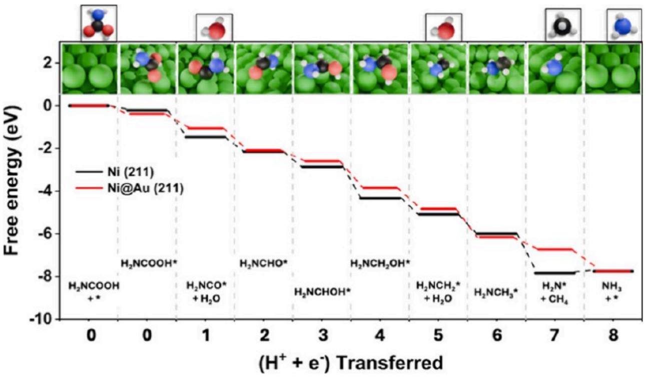


Figure 18. Comparison of calculated free energy diagram on Ni(211) and Ni@Au(211). The black pathway represents carbamic acid reduction on Ni(211) at −0.83 V vs RHE, and the red pathway represents the reaction pathway on Ni@Au(211) at −0.83 V vs RHE. Note that * denotes either a surface site or an adsorbed species on the surface. Here, nickel is represented in green, hydrogen in white, oxygen in red, carbon in black, and nitrogen in blue. Reproduced with permission from ref 93. Copyright 2024 American Chemical Society.

Beyond atomically precise systems, bimetallic and heterostructured catalysts offer synergistic reactivity by combining distinct functional domains.[112] In Fe−Co systems (Figure 19),[91] for instance, Fe may enhance CO_2 dissociation, while Co facilitates H_2 activation and hydrogenation. Similarly, mixed-metal frameworks such as Zn−Zr MOFs enable cooperative stabilization of formation or CO intermediates, thus guiding reaction selectivity. Zhang et al. illustrated how spatial proximity of Zn and Zr sites in a MOF can improve CO_2 hydrogenation performance, offering tunable pathways toward methanol or higher hydrocarbons.[92] These dual-function and synergistic designs are particularly relevant in tailoring product distribution under high-pressure CO_2 conditions.

To address the formation of C_2+ products such as ethylene or ethanol, researchers have explored multifunctional and tandem catalyst architectures.[21,113] These systems strategically integrate multiple active sites or reactors to steer the reaction through sequential steps—such as CO_2 methanation followed by C−C coupling. Wang et al. designed a composite catalyst featuring proximity-controlled metal oxide and zeolite domains, allowing the intermediate CH_3* species to undergo coupling to form ethanol with high selectivity.[114] In another example, Ou et al. reported a red phosphorus catalyst decorated with atomically dispersed gold, which promoted photoinduced C−C−C bond formation from CO_2, as shown in Figure 20.[115] These examples demonstrate how tandem catalysis and multisite cooperation can overcome the intrinsic limitations of product selectivity dictated by the Anderson−Schulz−Flory (ASF) distribution.

In addition to structural strategies, energy-coupled catalysis has emerged as a means to enable CO_2 activation at lower temperatures and in more sustainable configurations.[17,21] Plasma-assisted systems, such as Cu/γ-Al_2O_3 under non-thermal plasma, have shown improved CO_2 hydrogenation rates and methanol selectivity, as reported by Cui et al. (Figure 21).[95] The plasma environment activates CO_2 vibrationally and promotes surface hydrogenation reactions via Eley−Rideal pathways.[95] Alternatively, photothermal[13] and electrocatalytic[116] systems built on covalent organic frameworks (COFs) offer high tunability, excellent charge transport, and compatibility with visible-light irradiation. Lu et al. reviewed a wide range of COFs incorporating metalloporphyrins or ketoamine functionalities that exhibited strong performance in photocatalytic CO_2 reduction, achieving high selectivity for CO or HCOOH under mild conditions.[104]

5.0. RECENT DEVELOPMENTS AND APPLICATIONS OF $LCCO_2$ MINERALIZATION

$LCCO_2$ sources utilized in mineralization and advanced separation processes include a range of industrial and natural emissions. Flue gas from industrial facilities such as coal-fired power plants, cement kilns, and waste incinerators provides a CO_2 stream (4−15% CO_2), suitable for mineral carbonation of alkaline solid wastes, including slags or mine tailings.[117−120] Reddy et al. reported on the reaction of coal fly ash with CO_2 flue gas in a circulating fluidized bed. After a reaction time of 120 min under conditions of 43−54 °C temperature and 88−114 kPa pressure, the carbonation conversion rate was only 2.34%.[121] Power et al. found that the reaction of flue gas with ultramafic mine tailings from the Baptiste nickel deposit in British Columbia, Canada, would offset ~50% of a mine's emissions based on year-round reaction, and when extrapolated to the scale of the proposed tailing facility (5 km^2).[122] Regarding CO_2 from ambient air (~0.04% CO_2), Ragipani et al. investigated that coupling DAC with mineral carbonation of coal fly ash in alkaline sodium carbonate solutions under ambient conditions achieves up to ~80% CO_2 conversion in 1 h.[123] Collectively, these technologies provide flexible, regenerable CO_2 sources that lower the energetic and material barriers to mineralization and advanced separations, thereby accelerat-

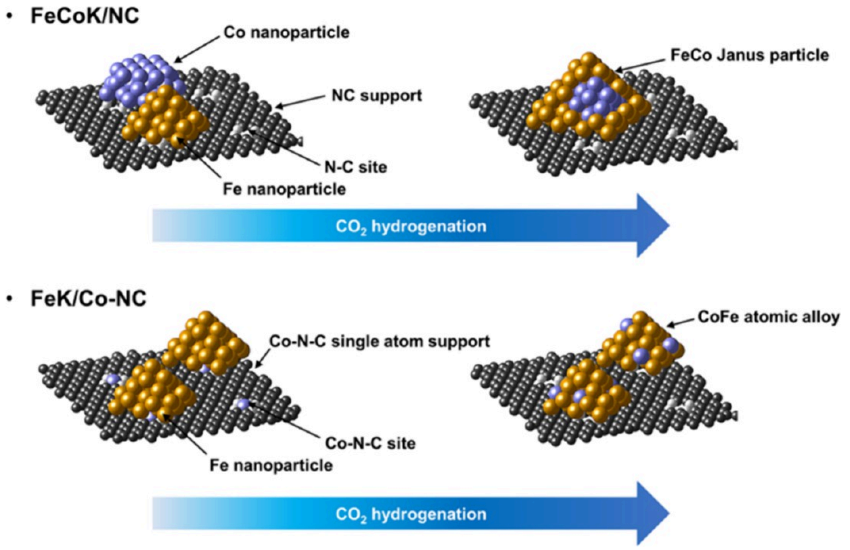


Figure 19. Proposed structures of the FeCoK/NC and FeK/Co−NC catalysts after CO_2 hydrogenation. Reproduced with permission from ref 94. Reproduced with permission from ref 91. Copyright 2021 American Chemical Society.

Figure 20. Interface-assisted catalytic C−C Coupling of Au single atoms over red phosphorus. Photocatalytic reduction of CO_2 to ethane. Reproduced with permission from ref 115. Copyright 2022 American Chemical Society.

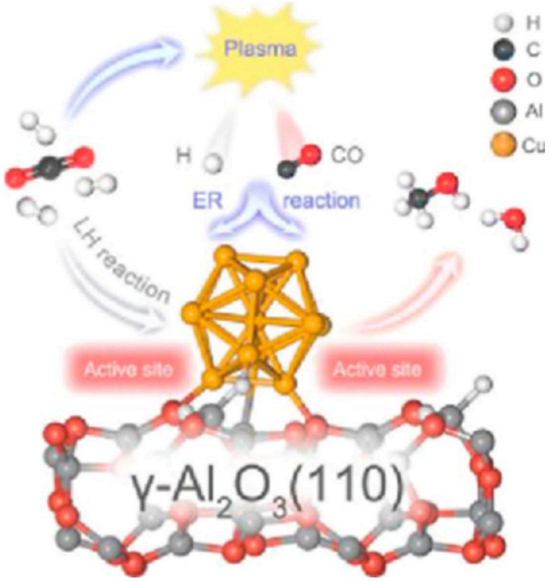


Figure 21. Plasma-catalytic methanol synthesis from CO_2 hydrogenation over a supported Cu cluster catalyst: Insights into the reaction mechanism. Reproduced with permission from ref 95. Copyright 2022 American Chemical Society.

ing the deployment of negative-emission and circular-carbon processes.

5.1. Enhanced CO_2 Mineralization.

Natural mineral carbonation occurs over geological time scales, which is too slow to effectively reduce current atmospheric CO_2 levels. Accelerated CO_2 mineralization is categorized into three major pathways—passive, in situ, and ex-situ carbonation—based on the operational environment and process design.[124]

Passive CO_2 mineralization involves ambient CO_2 reacting with alkaline rocks or industrial residues like ultramafic mine tailings left in situ, without added heat, pressure, or chemicals.[125] This method leverages natural weathering, requiring negligible energy and capital, making it a low-cost option for large-scale deployment near waste streams.[126] For example, passive air-weathering of Dumont Nickel's ultramafic tailings increased inorganic-carbon content from 0.1 to 4 wt %, potentially sequestering ~21 kt CO_2 annually (16% of site emissions) at a rate of 1.4 kg CO_2 t^{-1} yr^{-1} for the mine's 15 Mt yr^{-1} of tailings.[127] Although naturally slow, simple measures like drilling holes and pumping air into tailings can significantly accelerate this carbonation process.[128,129]

In-situ CO_2 mineralization involves injecting CO_2 underground into reactive rocks, where it dissolves in formation water and mineralizes within the reservoir (Figure 22).[130] The CarbFix pilot in Iceland injected 2,200 t CO_2 yr^{-1} into basalt.[131] Despite this, reaction kinetics are slow, taking months to years. Enhancement methods include drilling, hydraulic stimulation, and CO_2 preheating.[132] Drilling increases surface area and accesses deeper deposits. For example, Reykjavik Energy injected 200 t of CO_2 and water into a 500-m drill hole in basalt, with over 95% of the CO_2 mineralizing into stable carbonates within two years. Hydraulic stimulation improves rock conductivity and contact surface area for CO_2 injection. CO_2 preheating enhances reaction kinetics, with optimal conditions for olivine mineralization reported at 180 °C, CO_2 partial pressure >70 bar, and pH ~ 8 at the bottom hole.[133]

Ex-situ CO_2 mineralization, unlike passive or in situ methods, involves the surface mineralization of CO_2 with natural minerals and industrial alkaline wastes, completing reactions in hours for effective climate mitigation. A key advantage is the production of valuable carbonate products usable in cement, construction, and other industries.[134]

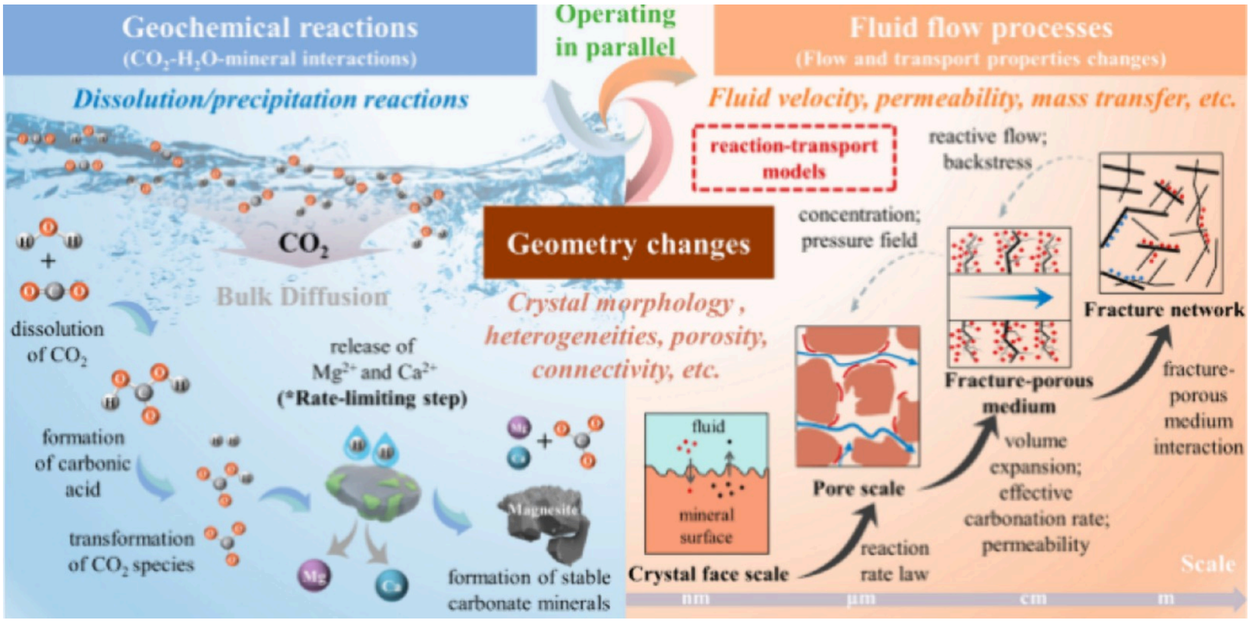


Figure 22. Schematic view of in situ CO_2 mineralization. Reproduced with permission from ref 130. Copyright 2025 Springer.

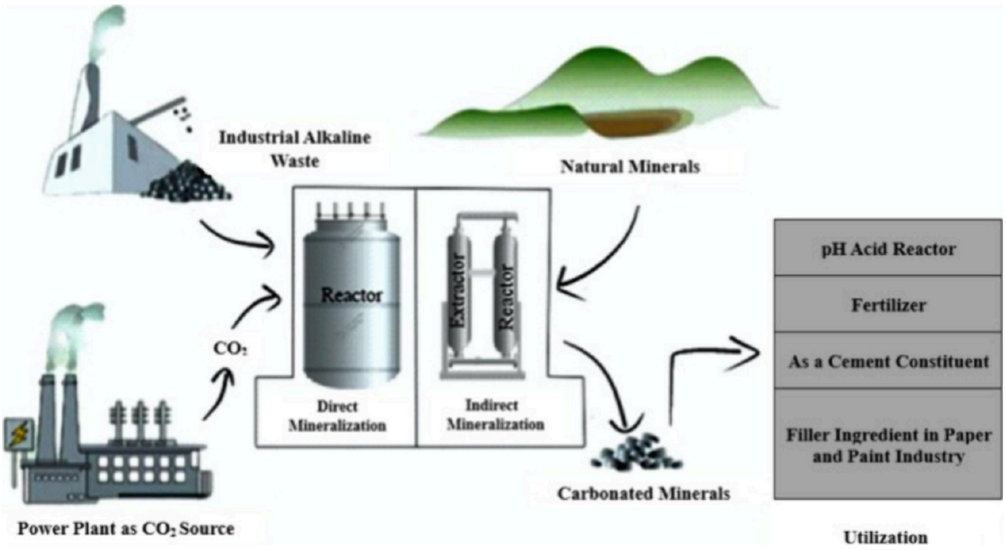


Figure 23. Ex situ mineralization processes. Reproduced with permission from ref 135. Copyright 2024 Springer.

Generally, ex-situ mineralization is categorized into direct and indirect methods (Figure 23).[135] Direct carbonation reacts crushed Ca/Mg-rich solids directly with pressurized CO_2 in a reactor to form carbonates. Indirect carbonation is a two-step process: first, Ca^{2+}/Mg^{2+} are leached into solution, then CO_2 is introduced to precipitate carbonates. This allows for milder CO_2 conditions and byproduct recovery but adds chemical handling.[136] A common limitation across all carbonation pathways is the dissolution rate of the silicate or oxide feedstock.[137] Consequently, process intensification strategies, including feedstock pretreatment, optimized reactor design, chemical additives, and pH control, are employed to accelerate ex-situ direct and indirect carbonation while enhancing product value and economic viability.[135,138−140]

5.2. $LCCO_2$ in Enhanced Critical Metal Recovery.

Recent research has focused attention on CO_2-enhanced critical metal recovery (ECMR), a promising approach to sustainable metal extraction that leverages the unique properties of CO_2 to improve efficiency, selectivity, and environmental performance of metal recovery processes. In enhanced metal recovery, CO_2 is employed as a versatile agent to improve efficiency, selectivity, and sustainability for extracting critical metals (CMs) from various sources, including ores, tailings, and industrial waste. Unlike enhanced mineralization, which aims at long-term sequestration and storage of CO_2,[130,136,137] the primary goal of ECMR is the recovery of critical metals from unconventional resources. ECMR leverages the unique properties of CO_2 to facilitate the dissolution, separation, and purification processes related to metal leaching and recovery. By utilizing $LCCO_2$, ECMR aims to reduce the reliance on harsh chemicals (e.g., cyanide, strong mineral acids) and energy-intensive methods traditionally used in the mining industry, thereby minimizing environmental impacts and promoting resource recovery efficiency.

Recent studies have placed great emphasis on the extraction of CMs using $LCCO_2$ in ex-situ direct mineralization processes.[141−143] $LCCO_2$ can be potentially integrated into existing mineral extraction or tailings management processes to generate value-added CM products,[144−146] as indicated in Figure 24. Ndlovu et al.[147] reviewed the applications and potential of $LCCO_2$ for ECMR through mineral leaching and flotation. Ultramafic mineral ores or mine wastes are suitable feedstocks for mineral carbonation, and aqueous mineral carbonation at ambient temperature is an attractive and energy-efficient approach. Hamilton et al. investigated accelerated mineral carbonization in ultramafic mine tailings via direct $LCCO_2$ reaction. $LCCO_2$ (10 vol % in flue gas) treatment doubled carbon mineralization within 4 weeks, while acid leaching using dilute H_2SO_4 achieved CO_2 sequestration rates of up to 21.2 kg/m^2/year. Additionally, trace critical metals, including Ni, Co, and Cr, were enriched in secondary Fe phases during the leaching process, indicating dual potential for carbon capture and CMs recovery.[148]

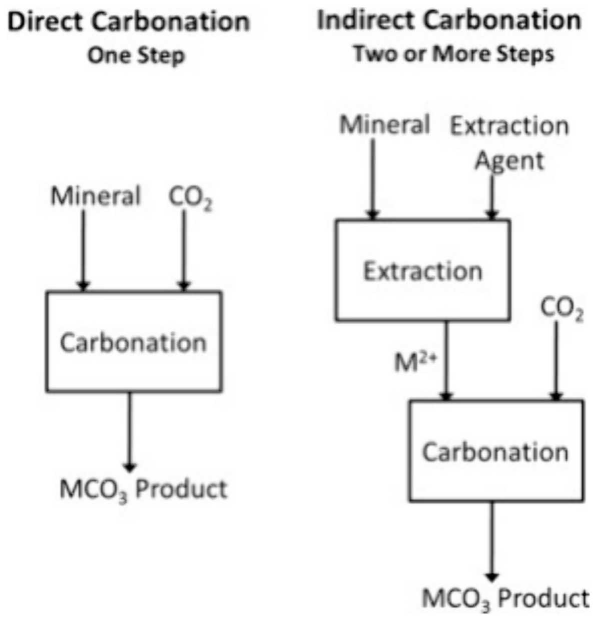


Figure 24. Overview of one step direct mineral carbonation and two or multistep indirect mineral carbonation. Note M refers to either calcium (Ca) or magnesium (Mg). Reproduced with permission from ref 145. Copyright 2012 Elsevier.

The use of $LCCO_2$ mineral carbonation with mine waste is a promising strategy for sustainable mine waste management. Hosseini et al.[149] examined the technical and economic feasibility of metal recovery from Victorian brown-coal fly ash. The overall process consists of two major processes: fly ash leaching using NH_4Cl and HCl, and carbonation with $LCCO_2$ (15 vol % in flue gas). Metal carbonates, primarily magnesium and calcium carbonates, are the major products, with the production cost for different scenarios ranging from \$61 to \$333 per ton of products. Further improvements to ECMR approaches might be considered to improve the overall profitability of the processes. Chen et al.[150] reviewed how integrating carbonation processes with existing mining operations and waste management practices can provide synergistic benefits, reducing costs and environmental impacts. Enhancing the economic viability of mineral carbonation involves recovering high-value products (e.g., critical metals) during the process, as shown in Figure 25. Other studies, according to combining $LCCO_2$ in ECMR, are summarized in Table 3.

Considering the low efficiency or slow kinetics of in situ dissolution of divalent metal containing silicates or oxides, ex-situ indirect CO_2 mineralization employs a two-step route: leaching to release Ca^{2+} and Mg^{2+}, followed by their precipitation as stable carbonates. Inorganic acids (e.g., HCl, HNO_3, etc.), organic acids (e.g., acetic acid, succinic acid, etc.), and salts (e.g., $(NH_4)_2SO_4$, NH_4HSO_4, NH_4HCO_3, CH_3COONH_4, NH_4NO_3, NH_4Cl, etc.) are extensively explored to facilitate the first step of metal extraction in ex-situ indirect CO_2 mineralization.

Kashefi et al.[154] conducted a process optimization for the integrated mineralization of $LCCO_2$ (10 vol %) and ECMR from bauxite residue (i.e., red mud). They found that a three-stage ex-situ leaching process could convert Jajarm red mud into high-purity (98%) $CaCO_3$. This was achieved via carbonation using Na_2CO_3, which was generated by scrubbing $LCCO_2$ with NaOH, precipitating 98%-purity calcite at a 32.7% efficiency.[154] Another $LCCO_2$ pH-swing aqueous process was applied to amorphous iron-slag using carboxylate ligands (acetate, propionate, butyrate), as shown in Figure 26. This approach achieved markedly higher metal extraction rates than using nitrate: at pH 3, acetate leached 63.6 wt % Ca and 97.3 wt % Mg, while all three ligands recovered >50 wt % total rare earth elements (REEs), including Y, La, Ce, Pr, and Nd, versus 43 wt % for nitrate.[155] Girdwood et al.[156] described a method of selectively extracting vanadium manganese, and

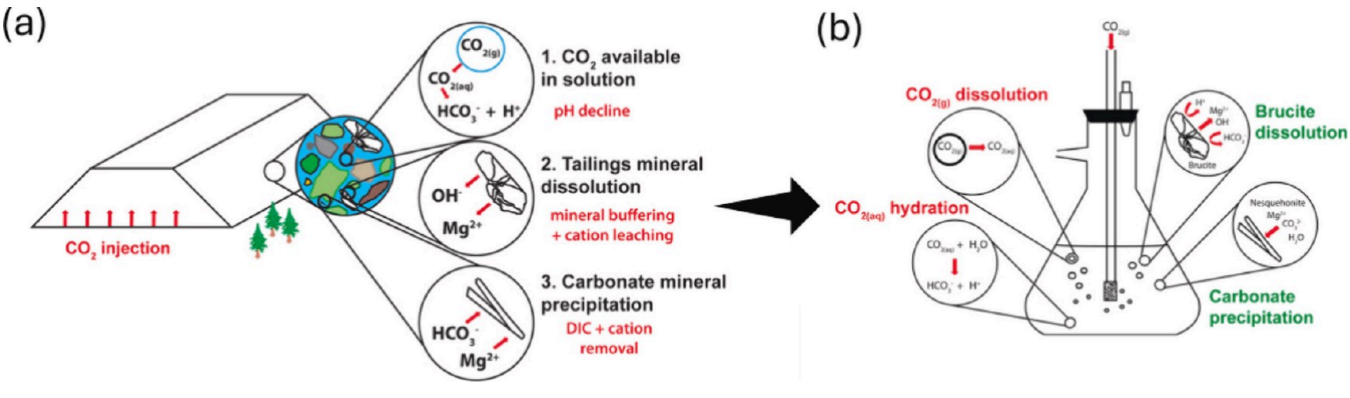


Figure 25. (a) Schematic of in situ carbon mineralization in situ in a tailings storage facility. (b) Laboratory experimental setup to mimic CO_2 mineral carbonation process with tailings. Reproduced with permission from ref 150. Copyright 2025 Elsevier.

Table 3. Ex Situ Direct CO_2 Mineralization and CM Extraction

carbonation pathway	method	feedstock	metal recovery	key results	ref
ex situ direct carbonation	accelerated mineral carbonation using tailings and $LCCO_2$ gas or solution	mafic/ultramafic tailings (serpentine, olivine, wollastonite)	up to 92% Mg conversion within 1 h (thermal activation method)	effective CO_2 sequestration with enhanced metal recovery	151
ex situ direct carbonation	CO_2 pressurized treatment (0–100 psi) on bastnaesite ore	bastnaesite ore	9.48% reduction in bond work index (grinding energy), improved grindability; no direct metal recovery	significant energy saving in grinding with minimal environmental impact	152
aqueous carbonation with sulfidation	sequential/concurrent carbonation and sulfidation in aqueous solution	Mg- and Ni-containing aqueous solutions	high Ni recovery through sulfidation; Mg carbonate formation up to magnesite	selective Ni recovery and Mg carbonate formation achieved under controlled pH and reaction sequences	153

calcium over iron from steel slag using the carbonated water, a saturated aqueous CO_2 solution.

Ex-situ direct CO_2 mineralization has become the leading laboratory-scale research focus due to its simplicity, rapid reaction kinetics, and controllability, particularly in aqueous systems. Recent advancements emphasize its dual potential for effective CO_2 sequestration and the enhanced recovery of critical battery metals such as Ni and Co. Complexation-assisted mineral carbonation was investigated by Wang et al. that converts silicate ores and laterites into stable carbonates while selectively extracting battery metals: per ton of CO_2 sequestered, the process recovers ≈99 kg Ni and ≈3 kg Co at >90% metal recovery and ∼60% carbonation efficiency. They found that combining weak $NaHCO_3$ with moderate-strength sodium NTA (nitrilotriacetic acid) sustains ligand-assisted dissolution of olivine, accelerating aqueous carbonation while selectively complexing Ni^{2+}/Co^{2+} and driving Mg^{2+}/Fe^{2+} into carbonate precipitation.[139] Wang and Dreisinger studied a highly effective process that integrates carbon mineralization with ECMR from olivine. By introducing a metal-complexing ligand, ethylenediaminetetraacetic acid (EDTA), as shown in Figure 27, the method achieves nearly 90% extraction of Ni and Co alongside efficient CO_2 mineral sequestration.[157] Wilson and Hamilton emphasized the dual potential of the fizzy ore direct aqueous carbon mineralization process to sequester CO_2 and enhance critical metal recovery from low-grade ultramafic ores and mine tailings. The process significantly improved Ni and Co recovery by utilizing pressurized CO_2 and H_2S gas mixtures combined with EDTA, which selectively extracted valuable metals while accelerating magnesite formation.[158] Katre et al.[159] investigated the corecovery of Ni and Fe via integrated carbon mineralization of serpentinized peridotite, using EDTA as an organic ligand to enhance metal recovery. The process involves reacting serpentinized peridotite with CO_2 in the presence of $NaHCO_3$ and $Na_2H_2EDTA{\cdot}2H_2O$ under high pressure and temperature. EDTA demonstrates greater stability with Ni complex ions compared to divalent metal carbonates. Furthermore, the buffered environment promotes simultaneous mineral dissolution and carbonate formation. These factors collectively contribute to the efficient corecovery of Ni alongside carbon mineralization and the production of magnesium carbonate.

6. PERSPECTIVES

This comprehensive review illuminates the evolving landscape of $LCCO_2$ enrichment technologies, highlighting both their transformative potential and the significant hurdles toward widespread adoption. The transition from traditional carbon capture and storage toward a circular carbon economy, powered by novel materials and processes, can be facilitated through effective harnessing of underutilized $LCCO_2$ resources from varied sources.

Technological progress in this area is contingent on addressing several key challenges. First, ensuring the long-term stability of sorbent materials, which are susceptible to degradation from moisture, contamination, and temperature fluctuations, is paramount. The design of robust and regenerable sorbents with high stability and efficiency must be achieved. Molecular dynamic simulations can be used in tandem with experimental work to assist in developing antioxidative sorbent formulations that withstand thousands of cycles. Machine learning (ML) can be utilized, for example,

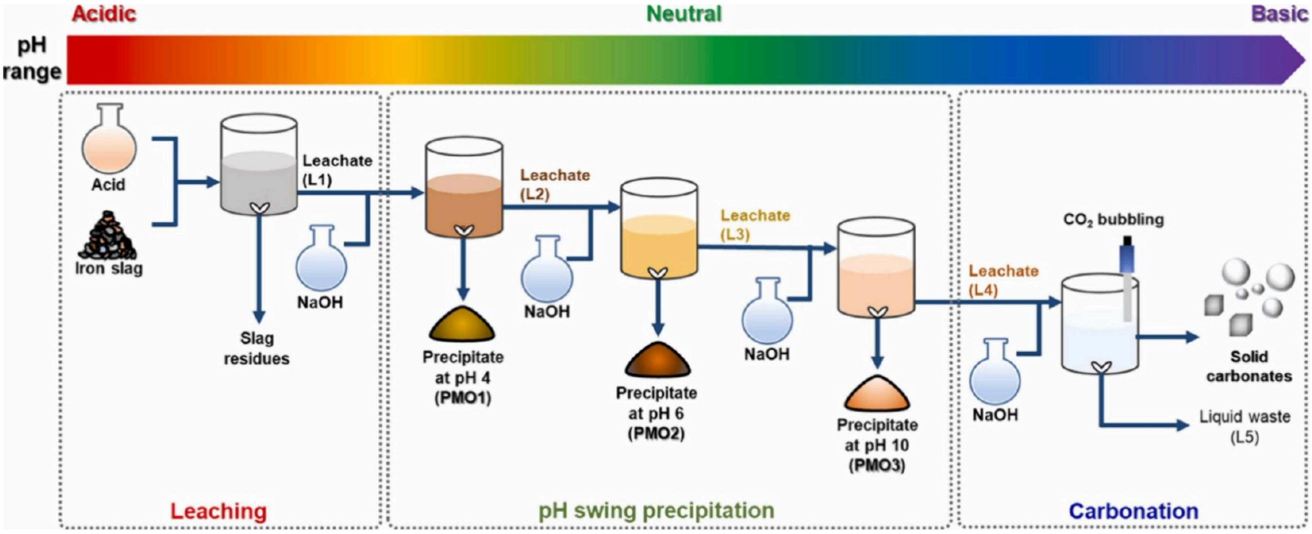


Figure 26. Scheme of aqueous pH swing-assisted carbon mineralization process for the coproduction of high-purity solid carbonates and rare-earth elements. Reproduced with permission from ref 155. Copyright 2023 Elsevier.

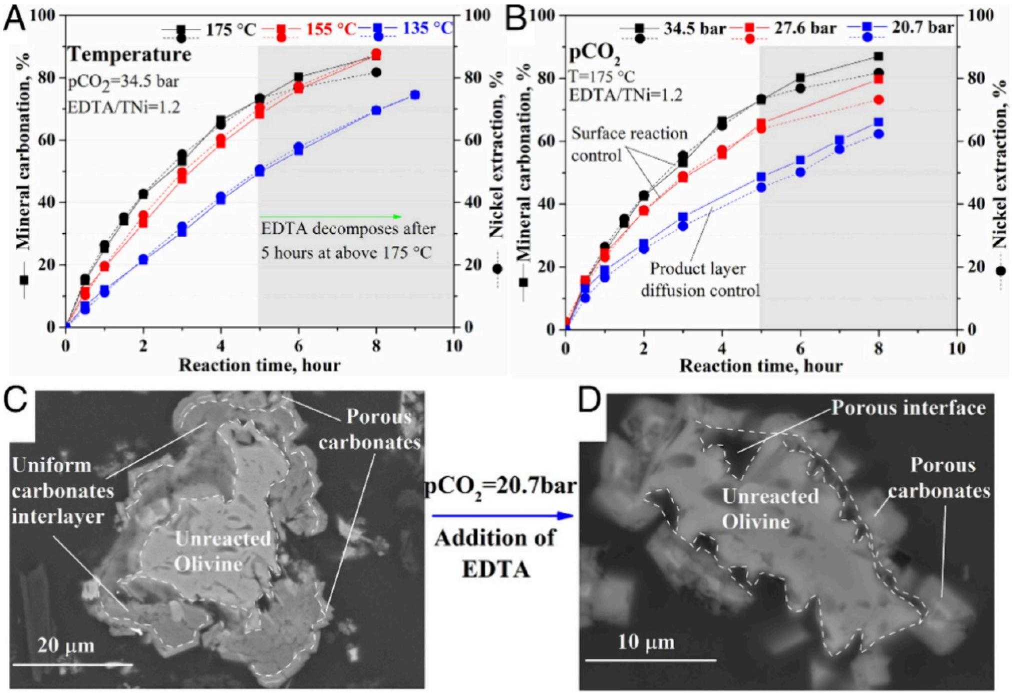


Figure 27. Suitability of mineral carbonation and concurrent metal recovery at (A) various temperatures and (B) various pCO_2 and the positive effects of EDTA on kinetic regime of mineral carbonation varying from (C) uniform carbonate layer diffusion control without addition of EDTA to (D) surface reaction control with the addition of EDTA. Reproduced with permission from ref 157. Copyright 2022 National Academy of Sciences.

to develop sophisticated degradation models, accounting for the effects of oxidation, leaching, and structural rearrangement of amines within the pores of BIAS. These techniques can be used for a wide variety of sorbent processes beyond those incorporating immobilized amines. Second, reactive capture systems must be designed to balance strong CO_2 binding and efficient catalytic turnover. The development of adaptive catalysts with dynamically responsive interfaces holds the key to optimizing both capture and conversion processes based on prevailing environmental conditions. Third, effective process integration is required to maximize the overall efficiency of $LCCO_2$ utilization, which calls for careful matching of the thermal and kinetic characteristics of the CO_2 capture and conversion steps and developing novel reactor designs and process control strategies. Moving forward also requires a focus on evaluating catalyst performance and establishing structure—function relationships, increasing the utilization of renewable energy sources, and working to make industrial translations more cost-effective to balance activity and selectivity, all improvements that will set the stage for innovation. Routine techno-economic analyses (TEAs) of different CO_2 enrichment processes need to be performed within the same research study every few years to ensure consistency among model parameters across the different technologies as they mature. These TEAs can be used to guide research into the most economical path – ex. economic viability of joule heating of

BIAS systems vs thermal heating of the calcium looping process. Furthermore, within each TEA analysis, results could help elucidate a new research direction to make the associated process more economical − ex. determining the optimal sorbent lifetime, which invites researchers to use ML to assist with sorbent refinement.

Advancing reactive CO_2 utilization will require improved experimental and modeling tools. On the experimental side, operando techniques—such as X-ray absorption, infrared, and Raman spectroscopy—will be essential for probing dynamic adsorption−conversion processes under realistic $LCCO_2$ conditions. These methods can uncover key intermediates and guide catalyst design. On the modeling front, multiscale simulations combining DFT, kinetic models, and reactor-scale CFD will help link atomic-level insights to system performance. Additionally, machine learning offers a powerful approach for screening sorbent−catalyst combinations and optimizing reaction networks based on experimental and simulation data. Integrating these tools will accelerate the rational design of next-generation $LCCO_2$ valorization systems.

Despite these challenges, the development and utilization of $LCCO_2$ demonstrates enormous potential for innovations. This extends to value-added chemical production, enhanced critical metal recovery and utilization of mine wastes through carbon mineralization, and agricultural practices such as improving crop yields through integrated greenhouse systems. Furthermore, a key area of future discovery lies in the efficient integration of renewable energy technologies.

Realizing this vision will require a concerted, multidisciplinary effort encompassing materials science, catalysis, chemical engineering, process design, and other fields. By overcoming the existing technological challenges and capitalizing on the abundant opportunities for innovation, $LCCO_2$ enrichment can play a critical role in shaping future industrial systems that are both carbon-conscious and resource-resilient, contributing to a more sustainable and environmentally sound future.

■ AUTHOR INFORMATION

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