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The Innovation Materials

Solar-powered integrated CO_2 capture and conversion-A potential paradigm shift for carbon neutrality

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Received: April 15, 2025; Accepted: May 23, 2025; Published Online: May 23, 2025; https://doi.org/10.59717/j.xinn-mater.2025.100134

Citation: Kim C.-A., Wu J., Sun Z., et al. (2025). Solar-powered integrated CO_2 capture and conversion-A potential paradigm shift for carbon neutrality. *The Innovation Materials* **3**:100134.

THE ESCALATING CARBON CRISIS AND TECHNOLOGICAL IMPERATIVES

The relentless combustion of fossil fuels since the Industrial Revolution has increased atmospheric CO_2 concentrations to unsustainable levels, exceeding 420 ppm - a 50% increase from pre-industrial values. This surge directly correlates with global warming, ocean acidification, and extreme weather events, threatening human civilization. Despite international pledges under the Paris Agreement, current decarbonization trajectories fall short of limiting warming to 1.5°C, necessitating negative emission technologies (NETs) capable of actively removing CO_2 from the atmosphere.[1]

Current carbon removal systems utilizing direct air capture (DAC) increasingly depend on geological sequestration strategies. Yet, critical questions persist regarding the century-scale integrity of subsurface CO_2 repositories and their potential to induce geochemical perturbations in Earth's crustal systems. Coupling DAC with renewable fuel and chemical conversion to achieve a carbon-neutral cycle shows great promise. Especially, the accelerated advancement of DAC technology has led to a continuous reduction in both the energy requirements and operational costs associated with carbon capture.[2] However, standalone CO_2 conversion technologies (e.g., thermocatalysis, electrocatalysis) often rely on purified CO_2 streams, requiring energy-intensive separation steps, undermining their sustainability. These fragmented approaches highlight an urgent need for coupling CO_2 removal from air with renewable energy-driven conversion into long-lived products, ensuring negative carbon emissions.

THE PROMISE OF SOLAR-POWERED INTEGRATED SYSTEMS

Solar-driven integrated DAC and conversion (SIDACC) systems demonstrate a transformative paradigm, leveraging sunlight as the primary energy input to power both the CO_2 capture step and its subsequent catalytic upgrading. By circumventing the need for external heat or electricity, SIDACC systems align with circular carbon economy principles, offering a pathway to decarbonize industries while producing sustainable feedstocks (e.g., methane, methanol, formic acid). While this research currently remains at the conceptual development stage for integrated carbon capture and solar-powered utilization systems, we maintain that substantial technical advancements could elevate this proof-of-concept technology. The proposed solar-driven CO_2 capture and conversion platform through systematic optimisation demonstrates the potential to enable decentralized, off-grid scalable systems supporting sustainable fuel generation and chemical synthesis applications.

SIDACC systems offer potential multifaceted advantages:

Low-carbon-footprint. Unlike conventional carbon capture approaches powered by non-renewable sources, solar-driven systems harness photonic energy directly to power CO_2 capture and catalytic transformation. Photothermal (or Photovoltaic) components enable continuous operation with near-zero operational emissions, and their modular scalability allows deployment in decentralized settings, further reducing transport-related carbon costs. The dual functionality of the in-situ CO_2 capture and conversion system eliminates the parasitic energy losses inherent in separated capture-storage and conversion processes while avoiding embedded emissions from external energy supplies. Crucially, life-cycle assessments demonstrate that the embodied carbon in solar infrastructure is offset within months of operation, yielding negative net emissions over the system's lifetime when paired with durable CO_2 utilization technique such as solar fuel synthesis. This integrated approach epitomizes the circular economy principles essential for achieving net-negative emissions targets by synergizing renewable energy harvesting, carbon cycling and durable utilization pathways.

Product Flexibility. Solar-driven CO_2 conversion with H_2O or H_2 produces various monocarbon (C_1) and multicarbon (C_{2+}) chemicals. Monocarbon C_1 chemicals play significant industrial roles, among which gaseous CO/CH_4 and liquid $HCOOH/CH_3OH$ stand out as the main products. Gaseous CO combined with H_2 (as syngas) is the raw material in Fischer-Tropsch synthesis for energy-dense hydrocarbon production. Many achievements in solar-driven C_1 production from CO_2 have been obtained through rational design of photocatalysts, photoreactors, and process engineering. C_{2+} production can be achieved via either direct or stepwise CO_2 conversion pathways (Figure 1).[3]

Scalability. SIDACC systems offer inherent scalability due to their modular design, reduced energy dependencies, and process intensification. By combining CO_2 capture and solar-driven conversion into a single photoelectrochemical or photothermal unit, these systems eliminate the need for energy-intensive CO_2 compression, transportation, and storage infrastructure required in conventional carbon management approaches. For example, modular photoelectrochemical reactors-demonstrated in systems that convert captured CO_2 into syngas using photocatalyst-based photocathodes-can be deployed in decentralized settings, leveraging sunlight as both an energy source and a driver for continuous operation. Solar-driven systems also avoid reliance on grid electricity or fossil fuel-derived energy, simplifying site selection and reducing lifecycle carbon costs. The development of continuous-flow photoreactors and photothermal catalysts further support scalability by enabling stable, long-term operation with minimal maintenance. These factors collectively position SIDACC as a scalable solution for distributed carbon management aligned with circular economy principles.

SYNERGISTIC DESIGN PRINCIPLES

The core innovation of SIDACC lies in the strategic coupling of two functional modules: the CO_2 capture module and the conversion module. The CO_2 capture module consists of advanced adsorbents (e.g., alkaline substance, metal-organic frameworks, covalent organic frameworks, or amine-functionalized mesoporous silica) with high selectivity and low-temperature regeneration (<100°C) capabilities. Photocatalytic or photoelectrochemical reactors that utilize solar energy to drive CO_2 reduction reactions (CO_2RR) using water or renewable H_2 as proton sources. For instance, Tian et al. have demonstrated integrating direct CO_2 capture material with a CO_2 reduction photocatalyst for atmospheric CO_2 capture and utilization. A C_2H_6 yield rate of 1.85 μmol g^{-1} h^{-1}, even though the optimal yield was attained under controlled laboratory-scale illumination conditions, has been achieved in the air through adsorbent ZSM-5 with NiV_2Se_4 (As illustrated in Figure 1A: direct CO_2 capture and conversion in a single reactor).[4]

Kar et al. present an innovative dual-bed flow reactor system for integrated DAC and solar-driven CO_2 conversion to syngas (CO + H_2).[5] The system employs a silica-polyamine adsorbent to capture atmospheric CO_2 at ambient conditions during nighttime, followed by photothermal CO_2 release and conversion using a TiO_2-cobalt bis(terpyridine) molecular-semiconductor hybrid under concentrated sunlight (As illustrated in Figure 1B: Stepwise capture and photothermal conversion). The composite achieved complete CO_2 saturation within 18 hours, demonstrating a DAC capacity of 87 ± 4 mg CO_2 per gram of adsorbent material under experimental conditions. Key

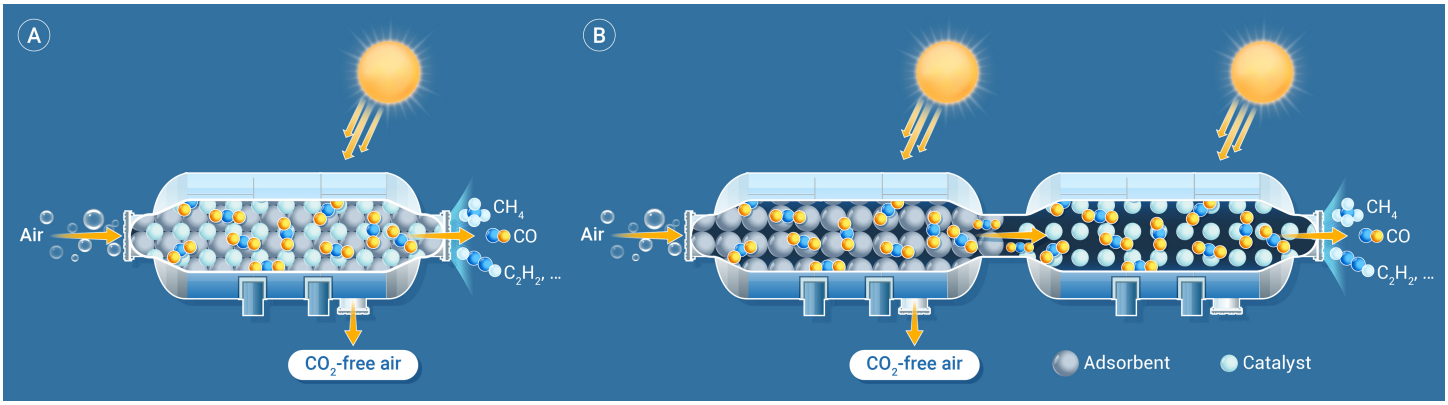

Figure 1. Schematic diagram of solar-driven integrated CO2 capture and conversion systems via (A) direct and (B) stepwise CO2 conversion pathways.

advancements include achieving 30-42% CO_2 concentration during desorption and producing syngas with a CO:H_2 ratio of 4:1, utilizing depolymerized PET-derived ethylene glycol as an electron donor. The reactor operates diurnally, separating CO_2 capture (dark phase) from conversion (light phase), thereby mitigating challenges of low atmospheric CO_2 concentration (400 ppm) and O_2 interference. The modular design demonstrates scalability, with a solar-to-CO_2 release efficiency of 0.6% and stable performance over multiple cycles, though catalyst deactivation at elevated temperatures (~56°C) and energy losses during intermittent operation remain limitations. This work circumvents the need for energy-intensive CO_2 compression or geological storage.

FUTURE DIRECTIONS AND CONCLUDING OUTLOOK

Integrating co-catalysts with semiconductor supports highlights a promising strategy to enhance selectivity and activity in gas-phase photoreduction. Future research could focus on developing dual-function materials that merge high-capacity CO_2 adsorption with photocatalytic activity, optimizing light-harvesting architectures, and integrating waste-derived reductants beyond PET. Additionally, it is flexible to coupling this system with downstream syngas-to-liquid fuel processes or hybrid renewable energy grids. This approach paves the way for decentralized, solar-powered carbon-neutral cycles, aligning with global net-zero goals by bridging materials innovation with system engineering.

Nevertheless, several critical challenges hinder practical deployment. First, the intermittent nature of sunlight imposes operational constraints on photo-driven carbon capture-conversion systems, particularly regarding sustained operation across diurnal cycles. Temporal decoupling strategies—as exemplified by Kar et al.'s dual-bed reactor architecture-have enabled phase-separated daytime CO_2 adsorption and high-intensity light-driven conversion. To mitigate energy intermittency, hybrid systems integrating multimodal storage demonstrate significant potential: (i) Li-ion batteries for short-term buffering of auxiliary power loads, and (ii) intermediate reservoirs storing surplus photoelectrochemical intermediates for nocturnal catalytic utilization. Second, catalyst deactivation under moderate thermal stress (≥56°C, as reported by Kar et al.) jeopardizes long-term stability, necessitating atomic-scale interface engineering to enhance thermal resilience. Third, selective adsorption of dilute CO_2 streams (<400 ppm) faces thermodynamic limitations that demand molecular-level tuning of adsorbent-gas interactions. While state-of-the-art sorbents achieve CO_2/N_2 selectivity >200 via chemisorption-physisorption hybridization, competitive adsorption by H_2O and O_2 remains unresolved in ambient conditions. Fourthly, energy-efficient regeneration is paramount for sustainable DAC scalability. Recent breakthroughs in mechanoactivated regeneration using 2D single-atom solution circumvent thermal processes, achieving unprecedented energy consumption of 0.56 GJ/tCO_2-a 78-86% reduction relative to conventional thermal systems (2.5-4.0 GJ/tCO_2).[2] Finally, in general, the solar-to-fuel conversion efficiency of 10% is suggested for practical industrial applications. Photoelectrochemical CO_2 reduction demonstrates enhanced charge separation kinetics through built-in electric fields, enabling low-bandgap photocatalysts to broaden photon harvesting into the near-infrared spectrum. This mechanism elevates solar-to-fuel conversion efficiencies to 1%-5%. In contrast, photocatalytic systems remain constrained by rapid carrier recombination, with efficiencies of around 1%. Critical breakthroughs in heterojunction engineering and cocatalyst integration must be realized to bridge the gap toward industrial viability.[3]

In conclusion, SIDACC technologies could redefine the greenhouse gas CO_2 - transforming a planetary threat into a renewable resource. Realizing this vision demands interdisciplinary collaboration, bridging gaps between materials science, chemical engineering, and energy policy.

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FUNDING AND ACKNOWLEDGMENTS

FUNDING AND ACKNOWLEDGMENTS The Chinese University of Hong Kong (Shenzhen) Startup Fund, Department of Science and Technology of Guangdong Province (No. 2023QN10L141), the Innovation and Entrepreneurship Team of Guangdong Provincial Pearl River Talents Program (2019ZT08L101), the Program for Shenzhen Science and Technology Innovation Committee (JSGGKQTD20221101115701006) and the University stability support program of Shenzhen (Grant No. 2023SC0027) for the financial support for this work. The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

AUTHOR CONTRIBUTIONS

C.K., J.W. and H.L. co-wrote the draft. C.K. helped draw the figure. Z.S. and H.L. supervised and revised the manuscript. All authors contributed to the manuscript and approved the final version.

DECLARATION OF INTERESTS

The authors declare no competing interests.