***Review Paper:***

# Recent Advances in Integrated CO2 Capture and *In situ* Conversion for Transitioning to a Lower Carbon Industry

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## Abstract

*Global warming from excessive carbon dioxide ($CO_2$) emissions has garnered international attention. The advancement of carbon-neutral technologies is a strategic imperative for a sustained human society. Integrating $CO_2$ capture and conversion (i-CCC) technologies enables the concurrent transformation of captured $CO_2$ from exhaust gases into value-added substances, thereby conserving significant energy and costs associated with the compressing and transportation processes of traditional Carbon Capture, Usage and Storing (CCUS) technologies. The study critically examines dual-function materials (DFMs), intermediate-temperature i-CCC techniques for methane generation and high-temperature applications for syngas manufacturing. The reactor's layout and the improvement of the operational parameters are highlighted from an industrial use standpoint. A comparative study was conducted on the dual-fixed-bed reactor method, which involves alternating flue gas and reaction substances and the dual-fluidized-bed reactor method, characterized by the circulating DM particulates.*

*The research anticipates that this analysis will encourage additional research, including the layout and fabrication of viable DFMs in lower-carbon industry, the exploration of practical catalytic processes for $CO_2$ conversion into high-value substances, the development of functional reactor models, the optimization of operational circumstances and the establishment of industrial demonstrations for the real-world application of i-CCC technologies in the future.*

**Keywords:** Carbon dioxide, $CO_2$ Capture, *In situ* Conversion, Lower-Carbon Industry.

## Introduction

The atmospheric carbon dioxide ($CO_2$) content has increased unprecedentedly, escalating from 270 ppm in 1750 to 410 ppm in 2022, hence becoming a focal point of climate change[8]. Carbon capture, use and storage (CCUS) is regarded as a viable approach for significantly mitigating anthropogenic $CO_2$ pollution. The carbon capture and storage (CCS) technique will eliminate roughly 25% of total $CO_2$ emissions from fossil fuel-dependent, energy-intensive industries including coal-fired power stations, cement production, iron and steel manufacturing and the petrochemical sector by 2040[14]. The substantial expenses associated with $CO_2$ enlargement, transportation and injections for storage, together with the ecology and environmental safety of later geological preservation, continue to pose significant obstacles for CCS in the lower-carbon industry.

In contrast, carbon capture and utilization (CCU) technologies offer a more viable option through usage rather than sequestration following $CO_2$ capture[6]. Numerous assessments have examined CCU methods including increased oil recovery, hydroformylation in $CO_2$ and mineral acidification. Moreover, utilizing captured $CO_2$ as a carbon source for synthesizing value-added compounds and fuels, CCU offers enhanced profitability in chemical engineering, thereby offsetting the substantial expenses associated with carbon capture[4]. Various CO-utilization techniques have garnered significant scientific interest including thermal catalysts, electronic catalysts, photocatalysis and plasma catalysts methods. A discernible trend in CCU involves the integration of carbon dioxide capture and conversion (C), which significantly reduces energy consumption, infrastructure requirements and costs associated with the compression and shipping of carbon dioxide in traditional CCU systems that utilize separate processing units in lower-carbon industry[19].

Integrating $CO_2$ capture and conversion (i-CCC)[6] technique can be realized by laying out and manufacturing dual-function materials (DFMs)[13] that possess adsorptive or catalytic elements. The adsorptive element typically comprises of alkali iron oxides or carbonates, which offer basic sites for $CO_2$ capture from flue gases in exhaust structures. In contrast, *in situ*, the catalytic element can transform the adsorbed $CO_2$ into value-added goods, including light hydrocarbons and commodities substances[5]. The desired attributes of DFMs include high $CO_2$ adsorption capability, exceptional catalytic efficiency, long-term stability and cost-effectiveness. It has significant obstacles in aligning the two processes, attributable to the disparate reactions of $CO_2$ adsorbed and conversion and the complexities in managing the temperature associated with endothermic $CO_2$ captivation and the endothermic converting in the lower-carbon industry[12].

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Significantly, a technical gap consistently exists between studies and practical implementation; thus, engineering procedures and reactor architectures necessitate a profound comprehension of the kinetics and thermodynamics of the i-CCC procedure[11]. This research critically presents the DFMs and the i-CCC procedures, emphasizing reactor construction and procedure-optimizing parameters from a manufacturing viewpoint. This review examines recent advancements in two categories of temporary and high-temperature i-CCCs, specifically between 250-450 °C and above 550 °C, with an emphasis on three significant products: restrictive hydrogenation to methane (i-CCC-Methane) and syn fuel (i-CCC-Syngas), along with the oxidative dehydrating of propane to ethylene (i-CCC-Ethylene).

## Comprehensive overview of i-CCC

Most chemical reactions are engineered to function in steady-state circumstances; for simplicity, the research will utilize the commonly employed fixed-bed columns to exemplify the QC procedures[15]. The DFM materials enable simultaneous $CO_2$ capture and $CO_2$ conversion procedures inside the same fixed-bed system in the lower-carbon industry. In the initial phase of $CO_2$ sorption, $CO_2$ is preferentially extracted from the exhaust gas and concentrated on the adsorptive element in DFM until saturated is reached, resulting in the exit of treated $CO_2$-free gas[1]. In the subsequent phase of $CO_2$ transformation, the gas is transitioned to a hydrogenation substance such as $H_2$, $CH_4$, or $C_2H_6$ found in organic or shale gases, accordingly facilitating the transformation of adsorbed $CO_2$ into multiple compounds including methane, syn gases and light-colored olefins at the outlet.

The DFM materials are reconstituted for the cyclic sorption procedure. Pure $N_2$ (the processed $CO_2$-free flue gas) is introduced during purging to eliminate the remaining product pollutants. Using two fixed-bed pillars, the i-CCC method can be constantly controlled through gas switching[18]. The beneficial relationship between the adsorptive and catalysis elements of the DFMs and the operating environment can significantly influence the efficiency of the i-CCC and value-added goods. The thermal energy from the high-temperature exhaust gases can be used as a heating source for the $CO_2$ transformation process, thereby diminishing the need for external energy sources[10]. The temperature of the exhaust gas is classified as medium (250-450 °C) as well as elevated (> 550 °C); thus, the advancements in i-CCC technologies are examined accordingly in the lower-carbon industry.

## Advancements in integrated $CO_2$ capture and reverse water-gas shift processes

The reversible water gas shifting (R-WGS) reactions are a significant process in C1 science, facilitating the generation of valuable compounds via Fischer-Tropsch synthesis with the resultant syngas. Equation (1) illustrates that the R-WGS response is an endothermic procedure with less Gibbs free energy than DRM. Despite the higher cost of $H_2$, the R-WGS procedure exhibits fewer adverse effects than DRM[3]. The combined $CO_2$ capturing and R-WGS (ICCU-R-WGS) (IRW) technology can directly generate pure syngas from the exhaust gas.

$$CO_2 + H_2 = CO + H_2O = 41.3kJ/mol \qquad (1)$$

**Impact of process factors:** Various essential factors for IRW, including temperatures, $CO_2$ attention and the amount of pollutants are reviewed as in figure 1. Elevated temperatures enhanced both $CO_2$ transformation and CO selection due to the exothermic characteristics of the R-WGS process[16]. A similar pattern regarding the impact of temperature was also observed with Ni/CaO DFMs. The research examined the temperature-programmed deactivation of IRW utilizing a Fe DFM and discovered that CO is produced at 550 °C, preceding the expulsion of $CO_2$ [2]. Within a specific temperature spectrum (<650 °C), using the IRW at elevated temperatures may produce increased CO due to the enhanced release of $CO_2$. An excessively rapid CO-releasing rate at elevated heat may diminish the transformation to CO in the lower-carbon industry.

$O_2$ and HO are constituents of flue gas that influence the efficacy of IRW. The study examined the effect of $O_2$ or $H_2O$ on R-WGS utilizing DFMs (Figure 1). The actual circumstances led to diminished R-WGS effectiveness due to the deactivation of active locations such as by surface oxidization or absorption[9]. The existence of $H_2O$ may inhibit IRW by aggressively adsorbing $CO_2$ and influencing the R-WGS balance. Compared to $H_2O$, excess $O_2$ exerts a more substantial detrimental effect on IRW. The addition of $O_2$ would markedly reduce CO transformation due to the oxidization of catalytic locations.

The amount of CO may influence both the efficacy of $CO_2$ adsorption and the subsequent R-WGS procedure[20]. As the amount of CO increased from 5.7% to 10.2%, the effectiveness of CO capturing and CO transformation marginally declined at heats below 460 °C, specifically from 100% to 97% and from 82% to 65% correspondingly. Conversely, CO sensitivity rose from 91% to 94%.

### Advancement of DFM performance for IRW

**Adsorbents in DFM for IRW:** Cao, an economical and plentiful adsorbent with superior CO capability, has been extensively utilized in IRW owing to its established usefulness in calcium chemical cycling. To mitigate the sintering of CaO during cyclic absorption and desorption, developing an open structure or enhanced dispersion improves the adsorbent's cyclic efficiency[17].

The sol-gel technique is a straightforward and efficient approach for synthesizing porous substances in the lower-carbon industry. Incorporating elements with elevated thermal conductivity aid in mitigating excessive sintered CaO. The introduction of CeO as a material obstacle inhibited the melting of CaO.

**Figure 1: $CO_2$ capture analysis**

Likewise, the research demonstrated remarkable cycle reliability (no decline after 12 cycles) by including MgO in the DFM. In addition to the often utilized CaO, hydrotalcite serves as a viable adsorbent owing to its stable structure at higher temperatures. DFMs utilizing hydrotalcite as an adsorbent demonstrated remarkable stability, remaining stable after 750 rounds.

**Catalysts in DFMs for IRW:** R-WGS is a prominent research subject owing to its prospective practical applications in C1 science. Precious metals are the predominant catalysts. R-WGS is favored at elevated reaction temperatures. Evident equilibrium constraints restrict the potential for producing high-purity synfuel from conventional R-WGS. IRW serves as a solution to enhance the implementation of R-WGS. The distinctive reaction process of ICCU enables R-WGS to occur at elevated hydrogen concentrations, facilitating significant CO transformation and the production of high-purity synfuel.

In the current IRW study, transitional metal-based catalysts have demonstrated exceptional catalytic efficiency and investigated the application of bimetallic $Fe_3$/Fe and $Co^3$/Co+ redox pairs inside hierarchical porosity CaO/MgO composites, resulting in extraordinary efficiency. The bimetallic couplings substantially reduced the electrical potential differences via the newly established Fermi threshold in FesCo, Mg10 CaO, facilitating electron spillover and enhancing the catalytic activity. It was proposed that the same concentrations of Fe and Co might attain optimal efficiency. The authors suggested that Fe was the catalytic location, while Co was the catalytic activator in DFMs. The catalytic procedure involves the reduction of $CO_2$ to CO using magnetite.

Hematite was reconstituted using $H_2$ with Co as the catalyst. The uniformly distributed Fe can guarantee the ongoing and effective execution of this catalytic activity. The study constructed DFMs for WGS under realistic circumstances, proposing the essential roles of Cu and K for effective CO reduction and capturing accordingly. Supporters can also play crucial functions in the catalytic process by enhancing and maintaining active catalytic areas. It attained nearly 100% selectivity for CO, a 51.8% transformation rate for CO and exceptional cycle reliability after 20 cycles of ICCU. The oxygen vacancy and the connection between Ni and CeO facilitate WGS. The proposed connection would substantially inhibit the aggregation of NiO.

### Integrating chemical looping $CO_2$ captivate and *in situ* transformation with manufacturing plants:

Linking chemical looping (LC) CL-I-CCC (CIC) with industrial facilities (ICL-I-CCC) (IIC) is a more successful method than combining it with successive CCU routes because it offers the benefits of (i) eliminating the post-treatment process for captivated CO and (ii) reducing energy and cost penalty. CIC can seamlessly integrate into industrial facilities as an independent subsystem utilizing CO-containing exhaust gas emissions. Combining the power and methane plants with CIC demonstrates higher thermodynamic and financial results than CCU. In terms of thermodynamic performance, IIC (Figure 2a) shows an energy utilization rate that is 1.7 percentage points superior, accompanied by increased emission of CO and reduced CaCO consumption (Figure 2b).

The $CaO/CO_2$ ratio and CaCO conversions (Figures 2c and 2d) exhibit a positive association with CO generation while demonstrating an adverse association with energy utilization. An additional CaL unit at the end of IIC can enhance energy efficiency by five percentage points despite increased energy penalties. Integrating a Fischer-Tropsch synthesizing step downstairs of IIC for synthesized fuel generation reduces heat need, enhancing converting energy and exergy efficiency.

Regarding the economic factors for the power plant scenario, IIC requires total yearly costs, CO production costs and $CO_2$ avoidance costs that are 14.21%, 26.12% and 75.15% lower than those of ISCCU correspondingly. The reduction in CO product cost would rise to 35.72% with the implementation of an additional Cal component to purify the syngas output from the CIC unit. The cost allocation indicates that $H_2$ expenses are the most significant component of the total annualized cost, necessitating more efforts to achieve the commercial display of IIC. Lowering $H_2$ prices (Figure 2e), prolonging plant lifespan (Figure 2f) and decreasing interest rates contribute positively to the economic viability of IIC and ISCCU. The operational cost of IIC is 121% less than that of ISCCUs when integrated into an ethanol plant, with helium cost being a significant influence.

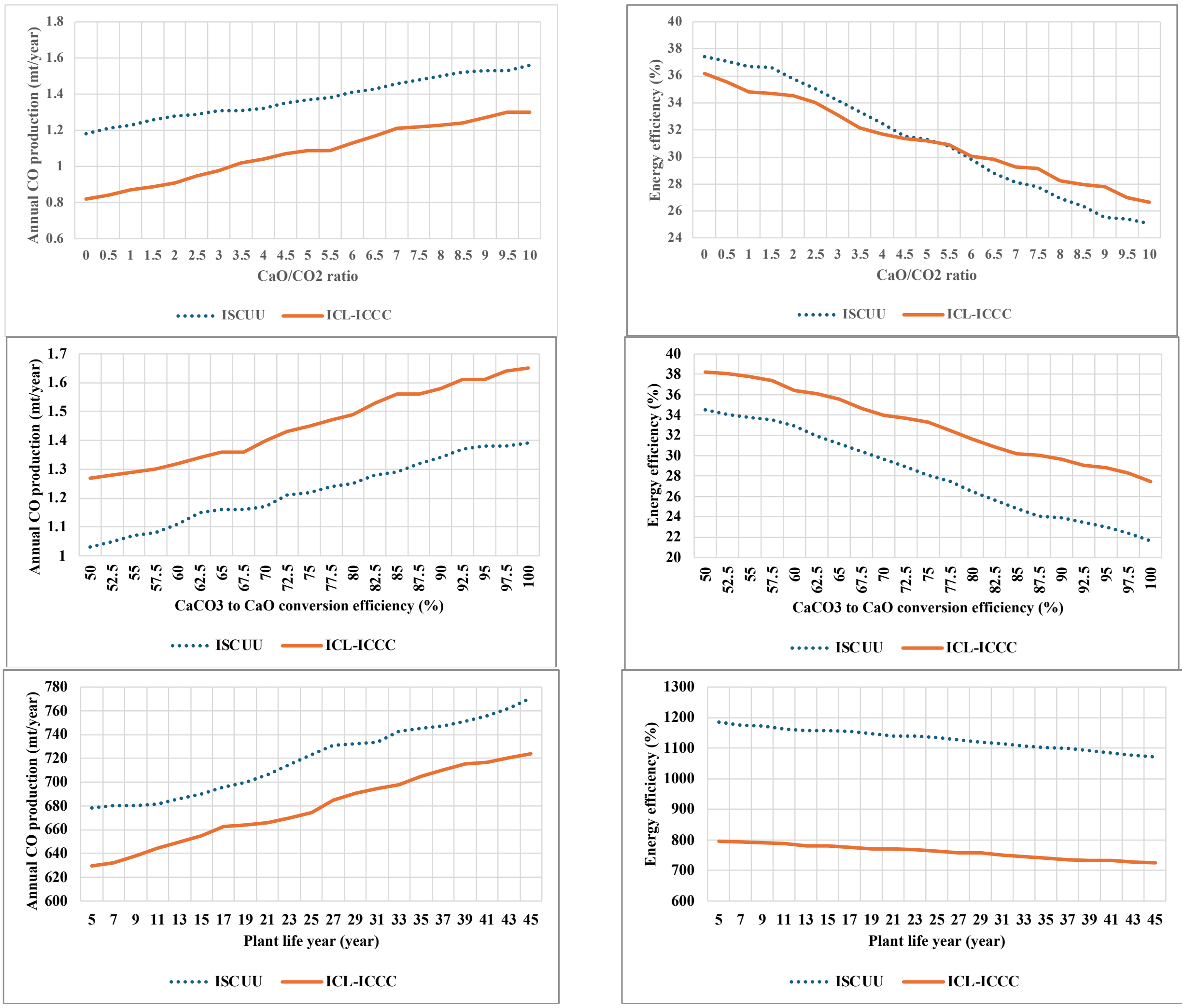

**Figure 2: CO production rate analysis**

These situations indicate a potential future for integrating CIC with industrial units but specific inherent challenges must be solved. The initial objective is the logical development of a dual-functional material that exhibits outstanding effectiveness, security and cheap cost to address the complex components of exhaust gases from diverse industries. The intrusion of oxygen, $SO_x$, $NO_x$ and other contaminants severely poisons bifunctional materials, leading to a deactivation of reactions. The prospective savings in thermal effectiveness and financial expenditure must be recognized. The thermodynamic immutability and cost creation of CIC and IIC processes can be examined by process modeling and sophisticated thermodynamic techniques. Moreover, the possible recovery of waste heat from industrial facilities can be re-purposed through heat and system cooperation methods.

## Chemical looping $CO_2$ captivate and *in situ* transformation for industrial transition

A perspective on attaining a green and lower-carbon industry transformation is provided through a process intensification method that integrates CIC, flue gas enhancement, solid waste recycling and industrial plants into a unified system (Figure 3). In this eco-friendly industrial structure, flue gas from the production line enters the CO-capturing reactor within the CIC component to isolate CO from inert contaminants through an organic adsorption reaction utilizing bifunctional materials. A "turning garbage into treasures" subsystem converts solid waste containing active elements from an identical industrial facility into a dual-functional material that acts as a catalytic site for converting $CO_2$ into valuable goods. These goods can be marketed to consumers, re-purposed for high-value applications, or returned to the industrial facility as fuels.

This approach achieves the zero-emission aim and the "one stone with two birds" impact for the industry by concurrently purifying commercial exhaust and solid waste greenhouse gases and integrating them into the CIC procedure with possible fuel supply to create a closed system. The viewpoint of transitioning to a green economy is not merely theoretical. Still, it represents a bright future, as its essential components, solid waste recycled into bifunctional materials and the capture and transformation of flue gas CO, have been established as possible, notwithstanding the need to address specific challenges before commercial implementation.

Renewable energy and method intensification technologies may be integrated to enhance the thermodynamic and economic performance of the suggested framework. Separation by membranes and reactor systems is proposed to mitigate contaminants' adverse effects on efficiency and to improve product purity accordingly. The conversing fluidized bed (CFB) is advised for operating the CIC method since it addresses the problems of inconsistent product density and temperature variations in stationary bed processors.

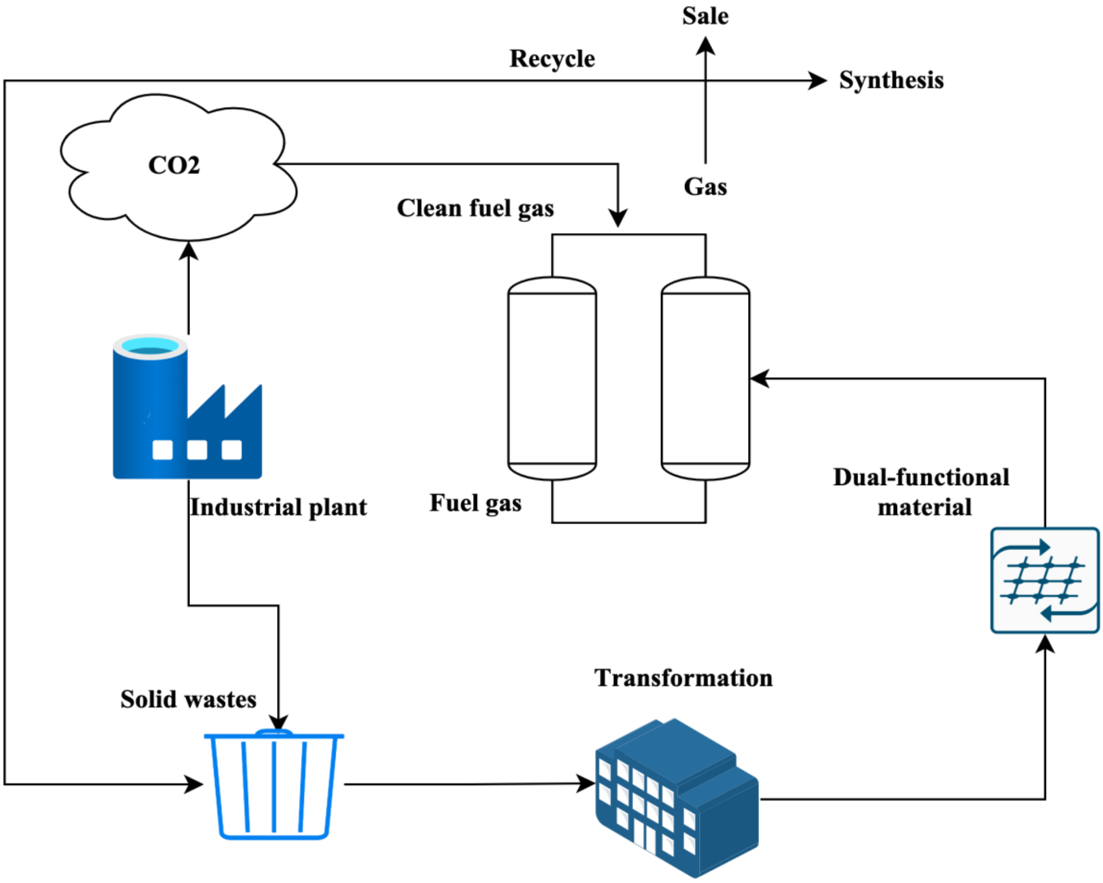

**Figure 3: Chemical looping $CO_2$ captivate and *in situ* transformation**

Implementing CFB may result in significant requirements for dual-functional materials and reactor architecture. A dual-functional composite particle must have enough rigidity to meet the demands of breaking and erosion during the fluidization process. This generally requires granulation and forming processes which leads to a specific reduction in CIC sensitivity. Choosing appropriate fluidization phases for capturing and converting processes in reactor architecture should be meticulously evaluated to enhance converting effectiveness for CIC. Solar power can be delivered to the converting half-cycle by solar reactors under extremely focused solar radiation. At the same time, thermochemical energy preservation is accomplished during the capture phase, significantly reducing energy requirements for the CIC scheme.

## Advancements in low-temperature research ICCU

The extensively utilized $CO_2$ collection methods (i.e. MEA adsorbed and calcium looping) and utilization techniques (i.e. $CO_2$ methanation and R-WGS) require significant energy for sorbent replenishment and $CO_2$ transformation. To address the limitations of the procedures above, many investigators focused on photo, electronic and plasma-assisted catalytic methods. Using photo, technological, or plasma energies, the combined process can attain commendable performance at lower temperatures (ambient temperatures and stress).

The conventional CO collection and utilization method can attain remarkable effectiveness. It requires either elevated temperature (>550 °C) or excessive pressure (>25 bar). Moreover, these procedures necessitated comparatively costly reducing agents (i.e. $H_2$ and $CH_4$) for the conversion of $CO_2$. The photo-catalytic elimination of $CO_2$ with $H_2O$ has surfaced as a viable alternative. Hybrid elements are utilized in combined $CO_2$ gathering and utilization, yielding promising outcomes.

Imidazolium fluids, imidazolylidene heterocyclic carbohydrates and zeolitic imidazolate structures are often used to achieve this process. Combined CO capturing and photocatalytic transformation offer a sustainable approach for lowering the production and usage of $CO_2$ emissions, although they encounter obstacles in enhancing catalytic performance. The electrocatalytic carbon dioxide reduction reaction for producing valuable compounds using alternative energy sources is appealing for converting intermittent green energy sources.

Carbon monoxide (CO) presents a significant challenge owing to several protons and electron transport pathways and the chemical inertness of $CO_2$ atoms. Numerous researchers have created a range of meticulously engineered materials to enhance the effectiveness of $CO_2$ conversion. The study employed a wet chemical and pyrolysis approach to synthesize Sb, resulting in efficient formate manufacturing. A study accomplished an electrochemical enhancement of CO from amine-capturing solutions, indicating significant advancement in the large-scale use of combined $CO_2$ capturing and electrocatalytic transformation. Electrocatalysis exhibits exceptional energy consumption effectiveness. The potential disadvantages include the stringent purity demands on the reactants to prevent the detrimental impact of contaminants on catalysts and apparatus.

Non-thermal plasma (NTP) accelerated CO conversion has emerged as a viable technique for substantially lowering reaction conditions, as plasma may activate CO at normal temperatures and under atmospheric stress. The membrane-based reactor is increasingly attracting the interest of researchers alongside DMs that incorporate adsorbents and enzymes. The study achieved integrated $CO_2$ membrane separating and $CO_2$ transformation, resulting in 91.8% carbon absorption effectiveness and 71.7% carbon incorporation efficacy. ICCU validated this integrated procedure with NTP to achieve exceptional reaction effectiveness and scalable CO output. The electrical usage associated with NTP-ICCU requires thorough examination.

Low-temperature ICCU has emerged as a viable avenue, utilizing meticulously engineered materials that exhibit exceptional catalytic efficiency in mild circumstances. Reducing $CO_2$ and neutralization must address concentrated $CO_2$ in lower-carbon industrial settings (e.g. power stations and cement manufacturers), necessitating high $CO_2$ discharge. Elevated material costs may render large-scale processing impractical, while the impurity resistance in $CO_2$ supplies will influence the accessibility of associated CCU capabilities.

## Conclusion

Due to the rapid advancement of DFMs, i-CCC technology has garnered considerable interest as it offers a novel approach to transform collected $CO_2$ from flue gases into value-added compounds. The reactions under certain circumstances, particularly the temperature alignment between the $CO_2$ capturing and conversion procedures, are essential for effective i-CCC, as both the mechanics and kinetics of adsorbent and catalytic reactions are temperature-dependent. The thermodynamic examination of reaction variations with temperature examines the i-CCC-Methane method at temperatures in between and the i-CCC-Syngas method at elevated temperatures in depth. Regarding i-CCC-methane, no supplementary heat is required because of the exothermic character of both the adsorbent and conversion phases; the ability to adsorb is insufficient for practical methane synthesis in the subsequent conversion step.

The i-CCC-Syngas method, which combines exothermic magnesium looping with endothermic DRM or R-WGS, enhances energy efficiency but presents issues in achieving compatibility. The primary challenge is the instability of DFM due to CaO sintered and catalytic deactivation triggered by carbon deposition at elevated temperatures. The i-CCC represents an innovative pathway for $CO_2$ consumption and the production of value-added methane in the lower-carbon industry.

To develop the i-CCC technique to a commercial level, employing dual-fixed-bed reactors that alternate between exhaust gases and reactants is a viable strategy. Challenges such as the uniform distribution of chemicals, the safety of common valve-switching, precise heating and cooling management and the feasibility of high-temperature compression must be resolved for further advancement. To mitigate the constraints of the fixed-bed system, the dual-fluidized-bed reactors model utilizing the circulation of DFM particles is suggested. The feasibility of complete energy consumption, uniform temperature transportation and comprehensive reactant interaction facilitates its operations. It imposes elevated demands on the mechanical strength of DFMs. It is imperative to produce DFMs with superior adsorption and catalytic efficacy alongside exceptional stability and mechanical resilience.

Current research mainly concentrates on the laboratory-scale creation of innovative DFMs. However, there is insufficient evidence regarding their performance under harsh circumstances including exposure to $O_2$, $SO_x$, $NO_x$ and $H_2O$ atmospheres and long-term testing. Further enhancement and shaping research are crucial for practical applications.

In conclusion, the i-CCC technologies represent an effective and environmentally friendly method for carbon capture and utilization, demonstrating superior $CO_2$ conversion effectiveness, lowering overall costs by circumventing $CO_2$ compressing and conveyance and attaining high energy effectiveness through full utilization of high-temperature flue gas temperature and response temperatures. Subsequent research, including the creation and production of viable DMs for practical applications, the discovery of innovative pathways for the transformation of high-value-added substances, the discovery of suitable reactor configurations and the setting up of lower-carbon manufacturing lateral line demonstrations, is crucial for enhancing the efficacy of i-CCC technologies in the future.

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