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Recent Advances in Multi-Pathway Catalytic Conversion Technologies for Carbon Dioxide under the Context of Carbon Neutrality

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Abstract

Carbon dioxide is the main greenhouse gas, and excessive carbon dioxide emissions have exacerbated global warming. Recycling and utilizing carbon dioxide is a key approach for China to achieve the "dual carbon" goals and achieve deep emission reduction in high-energy-consuming industries. This article outlines three carbon dioxide conversion products and their conversion processes, compares the performance and limitations of catalysts used for the production of monocarbon compounds and the dual-functional catalytic system of molecular sieves, summarizes the current engineering and material bottlenecks in the catalytic conversion process, and discusses future research directions.

Keywords

Carbon neutrality; CO_2 thermocatalytic hydrogenation; C_1 chemicals; Oxygen vacancy; Catalyst deactivation; CCUS.

1. INTRODUCTION

1.1. Research Background on CO_2 Resource Utilization

Excessive global greenhouse gas emissions are intensifying climate change. According to China's carbon neutrality development strategy, CCUS technology has become a key approach for reducing emissions in high-energy-consuming industries [1]. CO_2 thermal catalytic hydrogenation can recover CO_2 from exhaust gases of steel, thermal power, and coal chemical plants, converting it into high-value-added chemicals such as methanol, methane, and low-carbon olefins. This process simultaneously achieves emission reduction and carbon resource recycling, offering both environmental and economic benefits, and has now emerged as a major research focus in the field of energy catalysis[2].

1.2. Current Status and Existing Challenges in CO_2 Hydrogenation to C_1 Products

Currently, various catalytic systems—including copper, indium, nickel, iron, and cobalt—have been developed for the hydrogenation of CO_2 to produce C_1 compounds such as CO, methane, and methanol. However, competing reaction pathways and thermodynamic equilibrium constraints result in low single-pass conversion rates of CO_2. The three main types of catalysts—monometallic catalysts, oxide-molecular sieve composites, and methanation catalysts—commonly suffer from high-temperature sintering, carbon deposition, permanent poisoning by flue gas impurities, and high costs associated with precious metal feedstocks. Most existing research focuses on modifying catalyst interfaces through metal doping and support engineering to improve conversion efficiency, product selectivity, and catalyst lifetime[3].

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1.3. Research Approach and Writing Purpose of This Paper

To systematically clarify the reaction mechanisms of CO_2 hydrogenation, the structure-activity relationships of catalysts, and industrialization bottlenecks, this article classifies the processes based on product carbon chain length, focusing on the conversion pathways of three types of C_1 products. It summarizes methods for catalyst modification and deactivation mechanisms, providing a comprehensive theoretical reference for the engineering application of CO_2 resource utilization.

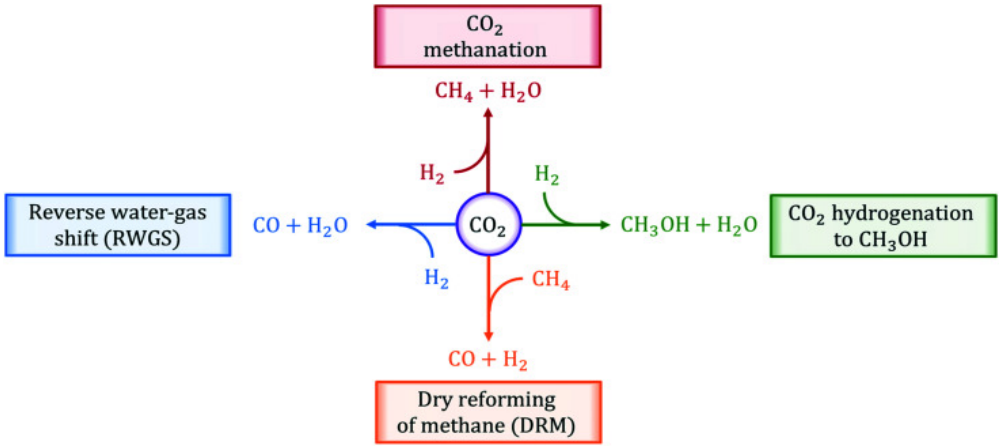


Figure 1. The three main conversion pathways of CO_2in thermal catalysis [4]

2. CARBON DIOXIDE TO CARBON MONOXIDE

2.1. Principle of Reverse Water-Gas Shift Reaction

The water-gas shift reaction (RWGS) is the core process of converting carbon dioxide into carbon monoxide through hydrogenation, and it is an endothermic reversible reaction. From a thermodynamic perspective, increasing the temperature is beneficial for the forward reaction. Although it significantly improves the conversion rate of CO_2 at high temperatures, it also greatly increases energy consumption [5]. At low temperatures, carbon monoxide is difficult to participate in the reaction, resulting in a low equilibrium conversion rate. This reaction lays the foundation for all CO_2 hydrogenation pathways. This reaction mainly consists of three parallel conversion pathways: redox dissociation, formate intermediate, and carbonate intermediate [6]. At low temperatures, CO_2 first reacts with dissociated hydrogen to form a formate intermediate, and then decomposes to release CO. When the temperature is above 300°C or on reducible oxides, CO_2 can directly dissociate at the oxygen vacancies to generate CO and adsorbed oxygen; the adsorbed oxygen combines with active hydrogen to form water and regenerate the oxygen vacancies. In addition, defect carriers such as cerium oxide and zirconium oxide can also promote the conversion through the carbonate intermediate. Oxygen vacancies are common active sites in all three pathways, and the dominant reaction path depends on temperature and the interface structure of the catalyst.

2.2. Typical Catalysts and Structure-Activity Relationships

Single-metal-based catalysts, oxides, and high-entropy alloys are the three main typical categories of RWGS catalytic materials. Iron-based and cobalt-based catalysts exhibit excellent RWGS activity and carbon chain growth ability under conditions of 280–400°C and 2.0–3.5 MPa. Promoting agents such as sodium, potassium, and rubidium can further enhance the reactivity by regulating the surface electron density [7]. The RWGS activity of nickel-based and copper-based catalysts is relatively weak; copper-based catalysts mainly produce methanol at low temperatures, and only significant RWGS side reactions occur at high temperatures. Indium

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oxide and zirconium oxide with a large number of inherent oxygen vacancies can capture and distort the CO_2 structure, thereby inhibiting the RWGS reaction. In contrast, reducible oxides such as iron(II) oxide and cerium dioxide can efficiently catalyze the conversion of CO_2 to CO through the redox cycle of lattice oxygen. The Fe-Co-Ni-Cu-Al multi-element high-entropy alloy can simultaneously optimize the activation of CO_2 and the reaction rate of RWGS, but its complex preparation process and high raw material cost limit its application to laboratory-scale research [8]. Density functional theory calculations indicate that oxygen vacancies at the metal-oxide interface can lower the activation energy barrier of CO_2, which is a key structural factor for enhancing catalytic activity. Additionally, there are significant differences in the adsorption strength of CO_2 and its intermediate form of formic acid on different metal crystal surfaces, making it possible to precisely control CO selectivity through grain size regulation and metal doping [9].

2.3. Major Technical Challenges

At low temperatures, the single-pass conversion rate of carbon dioxide is relatively low. Although increasing the temperature can enhance the conversion rate, it will significantly increase energy consumption, making it difficult to achieve a balance between energy efficiency and conversion performance. The RWGS reaction competes strongly with alcoholization reaction and carbon chain growth reaction, making it challenging to simultaneously achieve high carbon dioxide conversion rate and high selectivity for the target product under a single operation condition [10]. Long-term operation of catalysts often leads to deactivation: iron-based materials are prone to carbon deposition and carbon accumulation at high temperatures, covering the active sites and limiting the continuous stable operation time to less than 1000 hours [8]; nickel-based catalysts suffer irreversible loss of specific surface area due to particle sintering at high temperatures. Trace amounts of sulfur dioxide, halogens, and alkali metal ions in industrial flue gas can combine with the active sites to form stable salts, causing permanent poisoning of the catalyst. This requires additional complex flue gas pretreatment equipment, thereby increasing the overall operating cost.

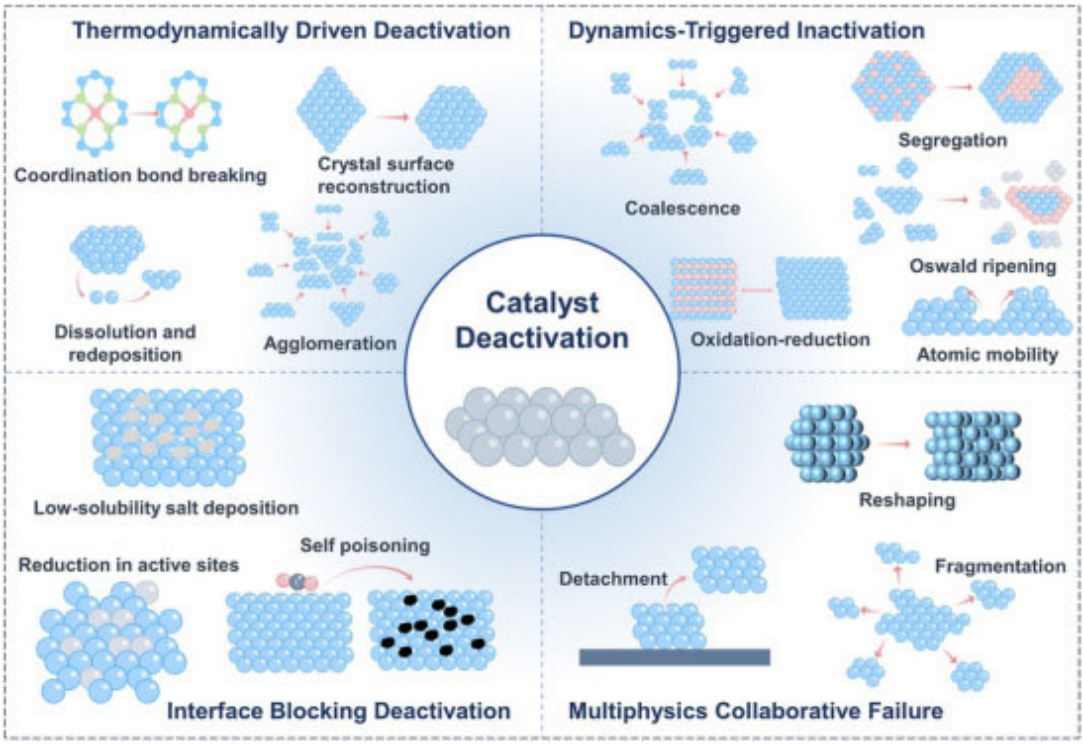


Figure 2. Schematic diagram of catalyst deactivation mechanism [11]

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3. METHANE PRODUCTION FROM CO_2 HYDROGENATION

3.1. Sabatier Reaction Mechanism and Thermodynamic Characteristics

The conversion of CO_2 to methane via hydrogenation, known as the Sabatier reaction, follows the overall reaction: $CO_2 + 4H_2 <=> CH_4 + 2H_2O$, with a standard enthalpy change of $\Delta H_{298K} = -165 kJ \cdot mol^{-1}$, making it a strongly exothermic and reversible reaction[12].Thermodynamic calculations indicate that low temperature and high pressure facilitate the movement of the forward equilibrium towards the positive direction, enabling a theoretical CO_2 conversion rate approaching 100% [13]. However, as the temperature rises, the equilibrium shifts in the opposite direction, resulting in a rapid decrease in methane selectivity and an exacerbation of the reverse water-gas shift side reaction, thereby generating a large amount of CO as a by-product. The reaction proceeds through multiple consecutive elementary steps: H_2 dissociates on metal active sites to form adsorbed atomic hydrogen $H\cdot$; CO_2 adsorbs at oxygen vacancies on the support to generate formate intermediates $\cdot HCOO$. These intermediates undergo successive hydrogenation, breaking the C–O bond and releasing oxygen-containing groups to form adsorbed CO. The CO then undergoes stepwise hydrogenation to produce methyl intermediates such as $CHO\cdot$, $CH2\cdot$, and $CH3\cdot$, which ultimately combine with hydrogen and desorb as methane[3]. The metal sites are responsible for the dissociation of hydrogen and the hydrogenation of carbon intermediates, while the oxygen vacancies activate CO_2; their synergistic effect can lower the reaction energy barrier. In situ infrared spectroscopy can observe the characteristic peaks of each intermediate, and density functional theory (DFT) calculations further confirm the interface synergy mechanism, providing strong support for the proposed reaction pathway. This technology can convert blast furnace gas and industrial flue gas into pipeline-grade natural gas, and is one of the key methods for carbon resource recovery [14, 15].

3.2. Nickel-Based and Noble Metal Catalysts

3.2.1 Nickel-Based Catalysts (Industrial Mainstream Catalysts)

Pure nickel catalysts offer the best cost-effectiveness and are widely used in the steel plants for the synthesis of methane from mixed CO/CO_2 gas streams. However, they have obvious drawbacks: they are prone to carbon deposition at high temperatures, and trace hydrogen sulfide in the flue gas reacts with nickel to form stable nickel sulfide, permanently deactivating the active sites and causing catalyst poisoning [3].

By doping rare earth elements such as lanthanum, cerium, or gadolinium to modify the nickel catalysts, the metal-support interaction can be enhanced, the nickel crystal size can be refined, the surface oxygen vacancies can be increased, and carbon precursors can be eliminated, significantly improving the sulfur resistance. The layered double hydroxide-derived catalyst composed of Al-Ce-La-Ni exhibits excellent low-temperature activity, achieving a total conversion rate of CO and CO_2 over 98% at 250°C, suitable for industrial low-temperature flue gas treatment. Moreover, this catalyst also has excellent thermal stability, capable of continuous operation at 400°C. However, it has the problems of high metal loading and relatively high raw material costs. Magnesium and calcium promoters can adsorb acidic sulfides, thereby alleviating the sulfur poisoning of the catalyst to a certain extent [3, 16, 17].

3.2.2 Noble Metal Catalysts

Based on noble metal catalysts of ruthenium and rhodium, they exhibit excellent activity in the synthesis of methane at low temperatures, and have outstanding tolerance to sulfur and carbon deposition [18, 19]. However, due to the scarcity and high cost of these noble metals, such catalysts are limited to small-scale, high-end applications and are difficult to achieve large-scale industrial applications [20, 21].

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3.3. Heat Management, Carbon Deposition, and Poisoning by Impurities

The Sabatier methanation reaction has extremely strong exothermic properties, causing local overheating in the fixed-bed reactor. This not only damages the catalyst structure but also shifts the equilibrium towards the production of CO, thereby increasing the generation of by-products. Industrial systems require segmented heat exchange equipment, which increases capital investment. Under high-temperature conditions, methane cracking and CO isomerization reactions will produce elemental carbon, which deposits on the active sites of the nickel and blocks the pores, shortening the service life of the catalyst. The trace hydrogen sulfide remaining in industrial flue gas can easily react with nickel to form nickel sulfide, causing irreversible poisoning of the catalyst [22]. Adding a deep desulfurization device will increase energy consumption and construction costs, thereby limiting the application of this process in steel mills. At the same time, impurities such as potassium, sodium, and chloride ions in the flue gas may damage the metal-carrier interface, weaken the interaction between the metal and the carrier, accelerate the sintering and agglomeration of nickel grains, and lead to catalyst deactivation [3,15].

4. METHANOL SYNTHESIS FROM CARBON DIOXIDE

4.1. Reaction Thermodynamics and Main Conversion Pathway

The primary reaction for methanol synthesis via CO_2 hydrogenation is $CO_2 + 3H_2 <=> CH3OH + H_2O$, an exothermic reversible process. The thermodynamically optimal conditions are temperatures between 200–260 °C, high pressure, and a high H_2/CO ratio. The reaction proceeds through the formate pathway: H_2 dissociates on the catalyst surface to generate active hydrogen species H ·; gaseous CO_2 adsorbs at oxygen vacancies at the metal-support interface and becomes activated, then combines with H · to form the formate intermediate ·HCOO. This intermediate undergoes successive hydrogenation steps to produce ·H2COO and ·H3CO, ultimately releasing a methanol molecule upon hydrogen desorption. Throughout this process, oxygen vacancies stabilize intermediates and lower activation energy barriers. In situ DRIFTS spectroscopy can detect the characteristic peak of ·HCOO, while DFT calculations confirm that this pathway has a lower energy barrier, establishing it as the dominant mechanism in methanol synthesis [23,24].

4.2. Reverse Water-Gas Shift Competing Side Reaction and Its Control Strategies

A competing reverse water-gas shift (RWGS) pathway coexists within the system, where CO_2 directly cleaves the C=O bond to form adsorbed CO ·, which subsequently desorbs as CO, consuming reactant hydrogen and directly reducing methanol yield. Above 300 °C, this side reaction gains significant kinetic advantage, becoming dominant and sharply decreasing methanol selectivity. Additionally, water molecules generated during the reaction occupy active sites and block oxygen vacancies, further suppressing CO_2 activation. Thermodynamic equilibrium calculations and comparative catalytic experiments demonstrate that stabilizing the formate intermediate and limiting CO desorption are key strategies for enhancing methanol selectivity [23,24].

4.3. Copper-Based Catalysts, Indium Oxide, and Other Oxide Catalysts

4.3.1 Copper-Based Catalytic Systems (Conventional Industrial Systems)

The traditional $Cu/ZnO/Al_2O_3$ catalyst is only suitable for the production of methanol using pure carbon monoxide as the raw material. When applied to a mixed gas stream containing CO_2, the reverse water-gas shift reaction side reaction becomes intense, resulting in a methanol selectivity lower than 60%. Introducing binary dopants such as zirconium, gallium, yttrium, or indium can regulate the copper grain size, construct the metal-support interface oxygen

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vacancies, stabilize the formic acid intermediate, and inhibit CO desorption, thereby increasing the methanol selectivity to 70%–78%. The ternary catalysts such as $Cu/ZnO/ZrO_2$ and $Cu/Ga/TiO_2$ exhibit excellent catalytic activity and thermal stability, making them current research hotspots. There are significant differences in the CO_2 activation ability on the Cu(111), Cu(100) crystal faces, and Cu-Zr alloys; reducing the interface reaction energy barrier is the core mechanism for improving catalytic performance [9, 25].

4.3.2 High-Selectivity Indium Oxide-Based Catalytic Systems

The inorganic oxygen vacancies are abundant in the indium oxide lattice, which can effectively capture and distort CO_2 molecules, thereby significantly inhibiting the reverse reaction of the water-gas shift. Therefore, it becomes an ideal benchmark material for converting CO_2 into methanol. By incorporating rare earth elements such as lanthanum and cerium, or main group metals such as zirconium and yttrium, the defects in the indium oxide lattice can be regulated, increasing the H_2 dissociation rate by 2 to 3 times. Using inert carriers such as TiO_2, SiO_2, or ZrO_2 helps to disperse the active particles, inhibit high-temperature sintering, and extend the catalyst's lifespan. However, this system has obvious limitations: indium is a rare metal with limited reserves and high costs, resulting in extremely high preparation costs for high-load indium oxide catalysts. Although copper-indium or iron-indium composite catalysts can reduce costs, phase separation often occurs at the interface, seriously affecting the long-term stability of the catalysts [26, 27].

Table 1. Comparison of operating data of the two catalysts (Cu/Zn/Al/Zr catalyst and $PdIn/SiO_2$ catalyst)

catalyst system	Reaction conditions (temperature / pressure / H_2:CO_2)	CO_2 conversion rate	Stable operation duration
Cu/Zn/Al/Zr catalyst (CZAZ-HF) [28]	240 ℃ / 5 MPa / 2.5:1	72.6%	1000 h
$PdIn/SiO_2$ catalyst [29]	280 ℃ / 2.5 MPa / 4:1	16.0%	900 h

4.4. Deactivation Mechanisms and Industrialization Bottlenecks

Catalysts for CO_2 hydrogenation to methanol face multiple industrialization bottlenecks: at temperatures above 300 °C, copper nanoparticles migrate and agglomerate, indium oxide crystals grow, and a significant number of oxygen vacancies are lost, leading to high-temperature sintering and deactivation; sulfur dioxide, alkali metals, and halide ions present in industrial flue gas can react with active copper sites to form sulfates and halides, causing irreversible poisoning, while the required flue gas pretreatment processes significantly increase operating costs; the reaction is thermodynamically limited, resulting in low single-pass conversion efficiency of CO_2, necessitating additional product separation and tail gas recycling units in industrial plants, which increases system energy consumption; furthermore, various catalytic materials have inherent limitations—indium oxide-based catalysts suffer from high raw material costs and scalability issues, copper-based catalysts exhibit poor thermal stability and resistance to poisoning, and bimetallic composite catalysts tend to undergo interfacial phase separation during prolonged operation, making their service life incompatible with the long-term demands of industrial production [30,31].

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5. CHALLENGES

Currently, the technology of thermocatalytic hydrogenation of CO_2 to produce C_1 compounds still faces numerous limitations. Catalytic materials commonly suffer from high-temperature sintering, surface carbon deposition, and permanent poisoning by industrial flue gas impurities such as sulfur, halogens, and alkali metals. Long-term operation of bifunctional composite catalysts often leads to interface delamination and degradation of synergistic catalytic performance[32]. Thermodynamically, low-temperature conversion efficiency is poor, while high-temperature operations increase side reactions producing CO[34]. The synthesis reaction of methanol and methane is highly exothermic, which easily leads to uncontrolled bed temperature. Moreover, the water produced during the reaction continuously inhibits the activation of CO_2 [33]. The precious metal catalysts such as indium and ruthenium are costly, and the preparation process of new composite catalysts is complex [34]. In addition, the industrial-scale pre-treatment of flue gas and product recovery and separation systems require a large amount of capital investment and energy consumption. Currently, the stable operation time of most catalysts is less than 1000 hours. Combined with the immature green hydrogen supply system, these factors seriously hinder the large-scale application of this technology [35]. In summary, all three pathways of converting CO_2 to C_1 are restricted by four common bottlenecks: thermodynamic side reactions, the tendency of catalysts to sinter and be poisoned and deactivated, difficulty in precisely controlling oxygen vacancies, and high energy consumption and high costs in the industrial process. Therefore, it is necessary to develop catalysts that are resistant to deactivation and low-cost.

6. CONCLUSION

The thermal catalytic hydrogenation process of CO_2 is regulated by controlling temperature, pressure, H_2/CO_2 ratio, and catalyst design to control the product distribution. Monometallic catalysts can selectively produce C_1 products such as CO, methane, and methanol, while oxide molecular sieve bifunctional systems are conducive to the synthesis of high-value multi-carbon chemicals. The main strategies for optimizing catalytic performance include metal doping, rare earth modification, and carrier confinement. Future research should focus on developing low-cost, poison-resistant non-precious metal catalysts, precisely designing stable bifunctional interfaces, integrating advanced processes such as fluidized beds and membrane separation to alleviate thermal runaway and thermodynamic limitations, and coupling with renewable green hydrogen to build an integrated carbon conversion technology. These efforts will help break through current technical barriers, promote large-scale application in high-energy-consuming industries such as coal-fired power generation and steel production, and contribute to achieving the "dual carbon" goals.

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