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ELECTRODE MATERIALS AND CELL CONFIGURATIONS FOR EFFICIENT CO_2 REDUCTION

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ABSTRACT

The electrochemical reduction of carbon dioxide (CO_2RR) offers an attractive potential solution to climate change by converting CO_2 into useful fuels and chemicals (e.g. CO, formic acid, methanol, hydrocarbons) using renewable electricity. This paper comprehensively reviews the recent progress in electrode materials (metal-based, oxide-derived, carbon-based, single-atom catalysts (SACs)), and their contributions to improved selectivity, lowered overpotentials, and increased durability. Copper is still a promising catalyst for a wide range of C_1 and C_2 products, while metals such as Au, Ag, Sn, and Bi exhibit excellent selectivity for either CO or formate. Advances in electrochemical cell designs (e.g. H-type, flow, membrane electrode assembly systems) can substantially impact CO_2RR performance through improved mass transport and current density. Performance indicators, such as Faradaic efficiency, current density, overpotential, and durability, are evaluated to assess the viability of the system under realistic conditions. Optimized catalysts, with cell designs suitable for large-scale manufacturing, significantly contribute to the development of CO_2 conversion technologies that can be more efficient and relevant to industrial standards.

Keywords: *CO_2 Reduction, Electrocatalysis, Copper Catalyst, Flow Cell, Membrane Electrode Assembly, Faradaic Efficiency, Single-Atom Catalyst, Renewable Energy.*

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1. INTRODUCTION

The anthropogenic rise in atmospheric CO_2 has exacerbated the global concern around climate change, creating necessary action to develop carbon mitigation strategies. One approach is electrocatalytic CO_2 reduction reaction (CO_2RR), which is the electrochemical conversion of CO_2 into carbon neutral fuels or value-added chemicals such as carbon monoxide (CO), formic acid (HCOOH), methanol (CH_3OH), or hydrocarbons. This method presents the possibility of mitigating climate change (by decreasing greenhouse gas emissions) but also of storing renewable electricity's cohesively as chemical energy. Yet, CO_2RR faces significant challenges such as low product selectivity, high overpotentials, and poor long-term durability of the electrocatalyst. Evolving electrode materials and optimizing electrochemical cell architecture are essential to addressing some of the shortcomings of CO_2RR.

The efficiency and selectivity of CO_2RR can be controlled through the properties of the electrocatalyst and design of the reaction environment. Ideal catalysts should be able to activate CO_2 at low overpotentials, access a preferential pathway, and suppress competing pathways such as hydrogen evolution. Adequate tuning of the electronic structural properties of materials that consist of transition metals, metal oxides, carbonaceous materials, and single-atom catalysts has been extensively studied for their capability to stabilize important intermediates through CO_2 reduction, while conversely tuning the electronic structure. Likewise, the configuration of the electrochemical cell (H-type, flow cell, and membrane-electrode assembly (MEA)) has a significant effect on factors such as mass transport, current density, and up-scaling.

Additionally, performance parameter points such as Faradaic efficiency, energy efficiency, current density, and operational stability are critical performance metrics for assessing the potential of CO2RR systems for practical applications. Research efforts are increasingly focusing on bridging catalyst development and advances in reactor designs to improve the overall performance of CO2RR systems in conditions relevant to permitting industrial uptake. This review summarizes recent developments in terms of electrode materials and electrochemical cells for efficient CO2 reduction, and identifies avenues for overcoming limitations and increasing the practicality of CO2RR technology.

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2. LITERATURE REVIEW

Ge et al. (2022) carried out a thorough study into the electrochemical CO_2 reduction reaction (CO_2RR) in membrane-electrode assembly (MEA) systems. They specifically set out to test how the performance, stability, and selectivity of CO_2 conversion was affected by different MEA systems and materials. The authors emphasized the important contribution of MEAs to improving CO_2RR efficiency through decreased ohmic resistance, improved gas diffusion, and very high current density achievable under relevant conditions for industrial applications. They discussed some of the challenges of MEAs such as membrane degradation, Catalyst layer adhesion, and ionic transport limits, along with engineering approaches to mitigate these issues for improved long-term operation and scale-up capacity.

Han et al. (2020) examined the application of nanostructured materials based on main group metals - bismuth, tin, and indium - towards the selective electrochemical reduction of CO_2 to formate, and the review highlighted the benefits of these materials including - low cost, low toxicity, and the thermodynamic binding energies for the formate producing thermodynamic reactions. Their review looked at recent advances in the synthesis of metal nanostructure, surface tuning, and heterostructures that improved Faradaic efficiency and reactor activity. Han et al. also reviewed the limitations of these systems, including stability and low current densities, while suggesting some approaches to audience like interface engineering and preference for electrolyte optimization.

Kibria et al. (2019) offered a wide-ranging overview of CO_2 electroreduction, linking fundamental electrocatalytic mechanisms to system level design considerations. They evaluated different types of catalysts including metal, alloy, and molecular catalysts, and related their structure-activity relationship to selectivity toward desirable chemical feedstocks as CO, hydrocarbons, and alcohols. The authors highlighted the important relationship between kinetics, mass transport, and reactor design when attempting to go from laboratory-scale experiments to usable CO_2 conversion systems. Their review emphasized that a significant understanding of mechanistic pathway and cell engineering is crucial for new technologies to develop commercially viable electrochemical CO_2 reduction technologies.

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Lai et al. (2022) concentrated on design strategies to substantially improve the energy efficiency of the electrocatalytic CO_2 reduction reaction (CO_2RR). They stressed the importance of maximizing selectivity and conversion rates, while reducing energy input. They reviewed several avenues of investigation design strategies for electrocatalytic CO_2RR, which included catalyst structure modification, interface engineering, and modifications to the electrolyte. They also discussed tandem and cascade reaction systems to decrease the overpotential and improve the energy utilization efficiency as alternatives to the separate transformations needed to convert CO_2 into useful chemicals. Their research successfully balanced thermodynamic efficiency against kinetic activity to facilitate consideration of design next generation CO_2RR systems with "reasonable" energy consumption limitations.

Lei et al. (2023) offered a thorough overview of recent advancements in electric catalysis (>99% CO2 converting) CO_2RR as a method for producing valuable intermediates (<1% CO2 to product). They systematically organized target products (i.e., carbon monoxide, formats, hydrocarbons, and alcohols) based on their reaction pathways. They included detailed discussions on the essential catalysts (metals, metal oxides, and molecular complexes). In addition, the authors emphasized that in order to improve catalyst design, the understanding of reaction mechanisms, intermediate species, and structure-activity was of critical importance. They concluded that marrying fundamental insights with materials design would be paramount for CO2RR technology to move toward applied industry.

3. ELECTRODE MATERIALS FOR CO_2 REDUCTION

Metal-based, oxide-derived, carbon-based and MOF/SAC catalysts all have unique advantages for CO_2 reduction, with a difference in selectivity, stability and efficiency. Advancements are made by tuning the active sites, enhancing conductivity, and improving CO_2 adsorption, to enhance the total catalytic efficiency.

3.1 Metal-Based Catalysts

Metal-based catalysts are some of the best studied systems for electrochemical CO_2 reduction due to their tunable electronic structures and catalytic flexibility. Different metals promote distinct reaction pathways and products, enabling targeted CO_2RR outcomes.

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Metal	Primary Product(s)	Mechanism	Challenges
Cu	CH_4, C_2H_4, C_2H_5OH	CO dimerization, PCET, hydrogenation of *CO intermediates	Poor product selectivity, catalyst restructuring
Au, Ag	CO	Weak CO adsorption and facile CO desorption	High cost, low scalability
Sn, Bi	HCOOH	Formate pathway via HCOO* intermediate	Stability issues, electrolyte sensitivity

Copper (Cu) is the only metal capable of producing hydrocarbons and alcohols, due to its moderately strong binding to *CO, a key intermediate. However, it is difficult to control product distribution (e.g. CH_4 vs. C_2H_4) because of competing pathways and catalyst surface restructuring during the reaction.

For gold (Au) and silver (Ag), the primary product is carbon monoxide (CO), following the reaction:

$$CO_2 + 2H^+ + 2e^- \rightarrow CO + H_2O$$

Their weak CO binding enables easier desorption, but high costs and scalability concerns limit broader application.

Tin (Sn) and bismuth (Bi) favor the production of formic acid (HCOOH) through the pathway:

$$CO_2 + 2H^+ + 2e^- \rightarrow HCOOH$$

These catalysts stabilize the HCOO* intermediate effectively, though they may degrade in acidic conditions.

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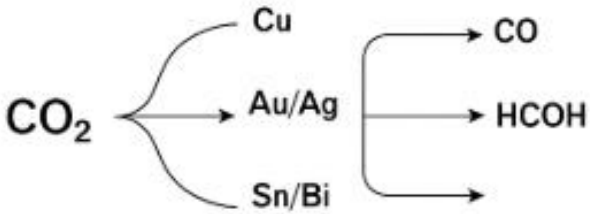

Figure 1: Schematic of CO_2RR Pathways by Catalyst Type.

The diagram illustrates the major product pathways associated with each catalyst group—Copper (Cu) leads to hydrocarbon products like CH_4 and C_2H_4, Gold (Au) and Silver (Ag) to carbon monoxide (CO), and Tin (Sn) and Bismuth (Bi) to formic acid (HCOOH).

3.2 Metal Oxides and Derivatives

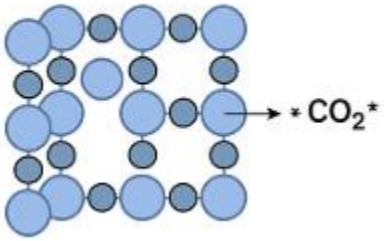

Figure 2: Role of Oxygen Vacancies in Metal Oxides

Metal oxides and their derivatives (including perovskites, layered double hydroxides (LDHs), and spinels) have variable redox properties and can interact strongly with CO_2. One important aspect of these materials is the presence of oxygen vacancies (V_o), which improve CO_2 adsorption and decrease the activation energy barrier.

$$M_xO_y + e^- \rightarrow M_xO_y .^- \ (V_o \text{ generation})$$

These oxygen vacancies are active sites that stabilize important intermediates ($*CO_2^-$) increasing catalytic turnover. TiO_2 based catalysts with designed vacancies displayed increased selectivity for the production of CO, while other catalysts made from In_2O_3 had much higher selectivity towards methanol. The stability and reusability of these materials can generally be good, but conductivity often requires hybridization with conductive supports e.g. carbon.

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3.3 Carbon-Based Materials

Carbon-based materials, especially carbon materials doped with heteroatoms, like N, B, or S, have been of considerable interest due to their low-cost, high surface area, and modifiable electronic characteristics. Nitrogen-doped graphene and carbon nanotubes (CNTs) have both pyridinic and graphitic nitrogen sites to serve as catalytic sites.

These dopants improve CO_2 affinity through π–π interactions and localize the charge density around the dopants, enhancing the stabilization of intermediates like *COOH or *HCOO. The porous architecture of the carbon material also enables gas diffusion and electron transfer. Though they are usually less active than metal-based catalysts for multicarbon products, they have a great promise for producing CO or formate.

3.4 Metal-Organic Frameworks (MOFs) and Single-Atom Catalysts (SACs)

Metal-organic frameworks (MOFs) offer an unparalleled amount of structural tuning which enables spatial confinement of active sites, and tuning of electronic environments. MOFs feature tunable pore structures and functionalized organic linkers that are optimal for implementing catalytic architectures compatible with accessible CO_2.

Single-atom catalysts (SACs) such as Fe-N_4 or Co-N_4 integrated into carbon matrices provide maximum access to metal atoms and facilitate discrete active sites. These isolated metal centers have high turnover frequencies and high selectivity through their unique coordination configurations and electronic environments.

SACs also tend to be resistant to aggregation and exhibit improved stability under catalytic reaction conditions. Advanced synthetic strategies, including pyrolysis of metal-organic frameworks (MOFs) and atomic layer deposition, have enabled the scalable preparation of SACs with high catalyst performance.

4. CELL CONFIGURATIONS FOR CO_2RR

Electrochemical CO_2 reduction (CO_2RR) performance is not fully determined by the catalyst; cell configuration is also important for efficiency, scalability and selectivity towards products. There

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are various different types of cells that could suit various research and industrial settings, with each presenting their own strengths and weaknesses.

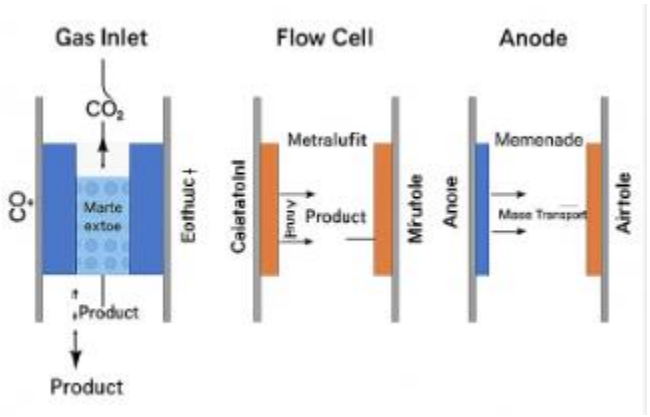

Figure 3: Comparative Illustration of Electrochemical Cell Configurations

To help clarify the structural, functional distinctions among the major electrochemical cell designs for CO_2 reduction, Figure 3 offers a schematic comparison of the three basic systems that are commonly used: the H-Type Cell, Flow Cell, and Membrane Electrode Assembly (MEA) system.

Table 2: Comparison of Electrochemical Cell Configurations for CO_2 Reduction

Configuration	Advantages	Limitations
H-Type Cell	Simple setup; ideal for fundamental studies and catalyst screening; well-controlled environment for mechanistic understanding	Low CO_2 solubility in aqueous electrolytes (~33 mM); diffusion-limited mass transport; not scalable for industrial use
Flow Cell	Enables continuous CO_2 supply; higher mass transport; achieves high current densities (>100 mA/cm²); suitable for long-duration testing	Requires precise engineering; gas diffusion electrode (GDE) may suffer from flooding or dry-out; pressure and flow control increase operational complexity
MEA System	Compact, zero-gap architecture minimizes ionic resistance; avoids	High cost of membranes (e.g., Nafion, Sustainion); complex water and heat

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	electrolyte crossover; supports industrially relevant current densities	management; requires humidification to maintain conductivity and prevent dry-out

1) H-Type Cell

This dual-chamber format—divided by a proton exchange membrane—is commonplace in laboratory studies. This design is ideal for mechanistic studies but is not applicable to actual CO_2RR because of poor mass transport of CO_2 in aqueous media, resulting in low current densities and poor efficiency.

2) Flow Cell

Flow cells utilize gas diffusion electrodes (GDEs), which allow CO_2 gas to be delivered more effectively to the catalyst layer. In this design, high productivity and product formation rates can be achieved, particularly for gaseous products such as CO and ethylene. However, GDEs are susceptible to flooding (due to excess electrolyte flow in) or drying, which vary performance over time and can limit performance stability long term.

3) Membrane Electrode Assembly (MEA) System

MEA systems use a zero-gap configuration consisting of pressing the anode, membrane, and cathode together, minimizing ohmic losses. MEAs also have the greatest promise for CO 2 RR for larger scaled industrial projects. It also allows a high current operational period while minimizing parasitic losses, but also requires continual water management strategies. There are anion exchange membranes (AEMs) and cation exchange membranes (CEMs), as well, but both can significantly raise capital costs depending on the product desired and the relevant electrolyte chemistry.

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4.1 Efficiency Metrics

A fundamental indicator for assessing the performance of CO_2RR system is the Faradaic Efficiency (FE), which evaluates the efficiency of the electrical charge used for producing a particular target product:

$$Faradaic\ Efficiency\ (FE) = \left(\frac{n\ \times F\ \times\ moles\ of\ product}{total\ charge\ passed}\right)$$

Where:

- **n =** n represents the number of electrons transferred per molecule of product (e.g., n=2 for CO or formate, n=8 for CH_4)
- **F =** Faraday's constant (96485 C/mol)
- **total charge passed =** measure of current integrated over time

Faradaic efficiency gives information about selectivity, while current density (mA/cm^2) tells us about rates of reaction. For our purposes, we want a FE > 80% for a single product, and a current density <200 mA/cm^2, preferably with long-term stability (> 100 hours).

Also, other metrics of importance are:

- **Energy Efficiency (EE):** the ratio of the Gibbs free energy of product formation to electrical energy input.
- **Turnover Frequency (TOF):** moles of product produced per mole of active site per unit time.
- Stability and durability will be the degradation of the catalyst or membrane over time during continuous operation.

5. PRFORMANCE METRICS

The performance of CO_2 electroreduction systems typically necessitates many parameters as descriptors of the interdependent metrics of efficiency, selectivity, activity, and stability. These

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descriptors will allow the viability of catalysts and cell configurations for both laboratory experimentation and industrial use to be assessed.

To provide a visual representation of how Faradaic Efficiency (FE) relates to current density and overpotential, figure 4 present generic behaviour found in CO_2RR systems:

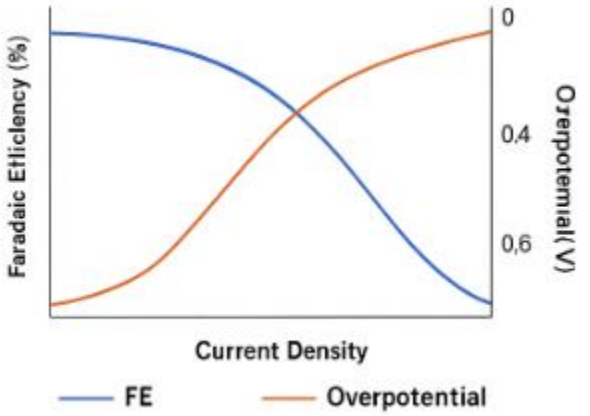

Figure 4: Faradaic Efficiency and Overpotential Illustration

- **Blue Curve (FE):** Indicates that Faradaic Efficiency is typically high at low current densities where there are few side reactions, specifically the hydrogen evolution reaction (HER). As current density increases, we often see FE decrease due to HER competition and limits in mass transport.
- **Orange Curve (Overpotential):** Indicates overpotential increases as current density increases because of the greater energy input needed to overcome kinetic and transport barriers. Inputting greater current density is critical for productivity, however, overpotential limits energy efficiency and shortens the catalyst lifetime.

This graph reveals the engineering trade-offs we face: we can achieve a high current density, which is important for industrial relevance, but we are always balancing FE losses and rising overpotentials. Good catalyst and cell design will optimize and balance as many of the elements above as possible, especially for common configurations and increasing current densities that are of industrial importance (>200 mA/cm^2).

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These images are also helpful to explain the numbers provided in Table 3 and further illustrates the challenges with developing effective, scalable CO_2 electroreduction systems.

Table 3: Key Performance Metrics for CO_2 Reduction Systems

Metric	Description	Ideal Target
Faradaic Efficiency (%)	Indicates the percentage of total charge that is effectively used to produce the desired product. High FE reflects excellent product selectivity and minimal side reactions such as hydrogen evolution.	>90%
Overpotential (V)	Represents the extra voltage required beyond the thermodynamic equilibrium potential to drive the reaction. Lower overpotentials improve energy efficiency and reduce power losses.	As low as possible (<0.5 V preferred)
Current Density (mA/cm²)	Denotes the rate of electrochemical conversion per unit electrode area. High current density is essential for achieving commercially viable production rates.	>200 mA/cm² (industrial relevance)
Stability	Measures the long-term durability of the catalyst and cell components under continuous operation. Stability ensures consistent performance and reduced maintenance.	>100 hours (continuous operation)

- **Faradaic Efficiency (FE)**

FE is a direct measure of selectivity. An efficient catalyst suppresses competing reactions (especially the hydrogen evolution reaction, HER) and directs electrons for the production of CO, formate, hydrocarbons, or alcohols. For multi-carbon (C_2^+) products, FE values greater than 60% are excellent when it comes to C–C coupling complexity.

- **Overpotential**

Reducing the overpotential is important for increasing energy efficiency and decreasing operational costs of CO_2RR. Overpotentials result in superfluous energy used, and contribute to

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catalyst degradation (lower activity and stability). Catalyst design aims to decrease the kinetic barriers hence minimizing the overpotential while preserving activity.

- **Current Density**

High current density is a benchmark for scalability because high current density yields high product formation throughput. Laboratory systems typically run in the range of 10-50 mA/cm², while industrial scale targets are usually over 200 mA/cm². Maintaining high FE at these high current densities is not trivial and is a major area of research.

- **Stability**

The stability of a catalyst under realistic conditions (e.g. fluctuating potentials, potential electrolyte degradation, CO_2 impurities in the feed) is necessary for commercial viability. Instabilities may stem from catalyst poisoning, agglomeration of the catalyst, or membrane failure. Long-term tests over hundreds to thousands of hours only losing a small amount of performance will be crucial for practical adoption.

6. CONCLUSION

The efficacy of electrochemical reduction of CO_2 is determined by a combination of the choice of advanced electrode materials and the specific configuration of the electrochemical cell itself. Metal-based catalysts, including copper or copper derivatives, offer potential for hydrocarbon and alcohol production. Other metals like gold, silver, tin, and bismuth exhibit high selectivity for CO and formate at certain conditions. New classes of electrochemical materials such as metal oxides, doped carbon materials, metal-organic frameworks (MOFs), or single-atom catalysts show advanced reaction kinetics and stability, boasting unique product specificity. However, consideration of the electrochemical cell configuration is key, since H-type cells are typically used for fundamental research, flow cells represent systems that can operate at higher rates of CO_2 conversion (e.g., enhanced electrochemical performance due to radiative losses), and membrane electrode assemblies (MEAs) can be scaled up for industrial purposes while minimizing resistive losses. Assessment metrics such as Faradaic efficiency, current density, over potential, and longevity are important for assessing and comparing systems; therefore, a combination of the best

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catalyst designs and the best reactor engineering is imperative for the commercial development of CO_2RR, which would offer an environmentally friendly alternative to fossil fuels.

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