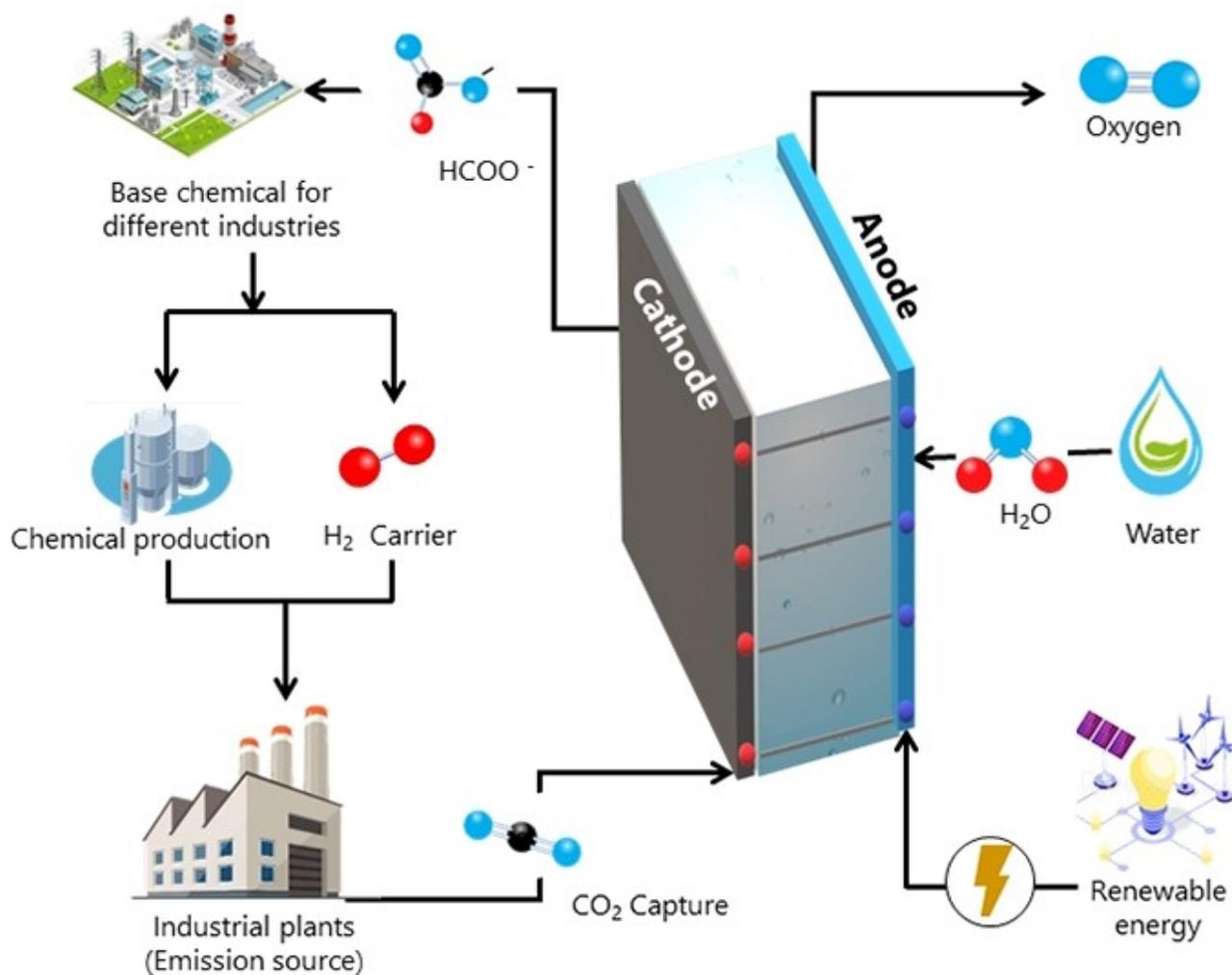


Electroreduction of Carbon Dioxide into Formate: A Comprehensive Review

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Carbon dioxide conversion into useful products has been gaining considerable attention as a global-warming-mitigation technique. The electrochemical conversion of CO₂ into high-value chemicals involves the utilization of electrical energy in the presence of an effective catalyst. The process products depend on the number of transferred electrons during the reaction and the characteristics of the electrode. Recently, electrodes coupled with active catalysts have been used to convert CO₂ into valuable products including formic acid,

hydrocarbons, and syngas. This review offers an overview of the recent literature on the electrochemical conversion of CO₂ to valuable products, with an emphasis on the production of formate/formic acid. In addition, it compares the main features of electrochemical conversion to other techniques and summarizes their key advantages. It also provides future perspective for research and development, such as the need for novel and selective catalysts to obtain high conversion and product yield with low energy consumption.

1. Introduction

Carbon dioxide (CO₂) is one of the main greenhouse gases (GHGs) that contributes to climate change, and it can absorb and emit infrared radiation.^[1] Consequently, it leads to global warming which causes a significant rise in the temperature globally. In addition, CO₂ emissions can cause acidification of water bodies due to the formation of carbonic acid from excess dissolved CO₂. The expected rise in energy demand in the next few years will eventually lead to more CO₂ emission which is estimated to reach 39.1 billion metric tons in 2030.^[2] The impact of CO₂ can be reduced by applying CO₂ capture techniques that include adsorption,^[3] membrane separation,^[4,5] chemical and physical absorption^[6] and mineral carbonation.^[7,8] CO₂ capture strategies are only considered as a transitory solution, on the long-term further CO₂ conversion techniques are required to re-functionalize the carbon dioxide by converting the gas to value-added products. Hence, several techniques such as electroconversion^[9,10,19,20,11–18] bioconversion,^[21,22] photoconversion,^[22,23] bio-electroreduction^[24–26] and photo-electroreduction^[27] are being investigated in the literature. Electroconversion has potential to convert CO₂ to methanol,^[28] syngas,^[29] ethanol,^[30] methane^[31] and formic acid^[32], depending on which boundary conditions are set for the industrial context.

One of the main advantages of producing formic acid is its versatile use in multiple applications. Particularly, formic acid has a great capacity to store hydrogen, which can reach up to 4.35 % of its weight;^[33,34] hence, it can be considered as an effective hydrogen carrier. At standard pressure and temperature, the capacity of hydrogen gas storage in formic acid is 580 times higher than the capacity of hydrogen gas that has the same volume.^[35] Moreover, formic acid is used to produce acetic acid and methanol.^[36] In addition, 2000 tons of formate solutions (50 wt/vol%) are used in US airports as deicing salts

annually.^[36] In spite of the importance of formic acid as a base material for several industries, the number of recent comprehensive literature reviews on CO₂ conversion into formate/formic acid is rather limited and mostly focus on specific aspects of electrochemical reduction. For example, Lu et al.^[34] presented a general discussion of electrochemical reduction of CO₂ to formic acid, while Du et al.^[37] reviewed inorganic cathode materials. Recently, Philips et al.^[38] compared several types of electrodes and cell configurations in terms of current efficiency toward the formation of formate. Nonetheless, the formate production literature is still deficient in providing detailed analysis of catalysts, cell configuration, membrane types and electrode materials. Thus, this review addresses these aspects, focusing on the electro-catalytic reduction of CO₂ to formate/formic acid and comparing it with other possible products, such as carbon monoxide, methanol, ethylene and ethanol. In addition, the review proposes further steps towards the enhancement of CO₂ reduction and suggests synthesis of new efficient catalysts with high Faradaic efficiency (FE) that can have industrial applications.

2. Types of CO₂ Conversion

CO₂ can be utilized through two routes: chemical and physical pathways.^[39] For the physical route, CO₂ can be used as pure or in a mixture. Physical utilization of CO₂ can be achieved by either direct or by indirect utilization. However, direct physical utilization has minor reduction of CO₂ and it has limited in applications. On the other hand, the chemical pathway can convert CO₂ molecules to fuels or chemicals.^[40] This can be achieved by several methods, such as photoconversion, bioconversion, thermal conversion and electroconversion.

Photoconversion may involve photocatalytic conversion^[41,6,42–44] or photo-electrocatalytic conversion.^[45] The main difference between these two methods is that photoconversion has a semiconductor electrode with photocatalyst that is exposed to sunlight to absorb photons and convert CO₂ into other chemical products,^[46] while photo-electrocatalytic consists of photoanode, photocathode or both in electrolysis cell separated by membrane.^[47] The combination of photoconversion with electrochemistry enhances the efficiency of the conversion.^[47]

Bioconversion can be achieved through artificial biosystems,^[48] microalgae^[49] or fermentation.^[50] Microbial bio-electrochemical systems are one of the artificial biosystems that

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utilize microorganisms or enzymes as biocatalysts with an electrical power source. The product selectivity can reach up to 100% using enzymes, however, this process has a high overall cost that needs to be reduced to become economically feasible.^[51] Microalgae are unicellular photoautotrophic species that are driven by sunlight^[52,53] and can be used in CO₂ utilization. The microalgae growth depends on several factors including nutrients, CO₂ as carbon source and light^[52] to produce biofuel.^[53,54] Furthermore, fermentation may also convert CO₂ into methane as a biogas. Besides fermentation, methanation is one of the other types of CO₂ conversion into methane; however, it is a natural mechanism that requires the use of catalysts due to the limitation in mass transfer.^[55]

Thermal conversion is achieved through plasma splitting^[56,57] and catalytic reforming.^[58,59] The process mainly depends on a catalytic reaction to produce carbon monoxide and hydrogen.^[59,60] This reaction suffers from carbon deposition that causes catalyst deactivation.^[61] Plasma splitting converts CO₂ to carbon monoxide and a free oxygen atom.^[60,62] It is

worth mentioning that conversion and capacity of the process are still low to be efficiently used in industrial applications.^[63]

A comparison of the core advantages of the above-mentioned methods for CO₂ conversion, including electrochemical conversion, is shown in Table 1. Among these, electrocatalysis has been able to achieve the highest efficiency.

3. CO₂ Electroreduction

3.1. Mechanisms

Electroreduction is a multistep process that depends on the number of electrons transferred, and it is divided into four main categories as shown in Figure 1.^[71] 1) two-electron transferred, which mainly produces carbon monoxide and formate. 2) Six-electron transferred, resulting in methanol production. 3) Eight-electron transferred to obtain methane as the main product. 4) twelve-electron transferred to synthesize ethylene and



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Table 1. Main advantages and disadvantages of CO ₂ conversion types.		
CO ₂ Conversion types	Advantages	Disadvantages
Bio-conversion	Sustainable conversion method. ^[48]	low efficiency and suffers from difficulties while operating in continuous mode on large scale ^[48,64] Dependence on sunlight limits its tractability and scalability ^[65] Methanation process is considered to be kinetically slow and suffers from poor mass transfer. ^[55]
Photo-conversion	Using sunlight as an energy source. ^[46] Sensible production cost ^[41,66,67]	The efficiency can be limited by: ^[42,68] Charge carrier separation. low CO ₂ solubility.
Electroconversion	Operates at ambient condition Ability to use renewable electricity including solar, wind, and hydro energy sources. ^[46] Can be carried out at low temperatures in liquid electrolytes. ^[46]	High overpotentials are required to overcome a competitive hydrogen evolution reaction. ^[69] Poor product selectivity and slow kinetics are required to be solved by developed materials. ^[69,70] Low operation temperature can cause low reaction rate and low selectivity in liquid electrolytes. ^[46]
Thermal-conversion	Plasma splitting can be operated at moderate conditions. ^[63] long term decarbonization. ^[58]	Mass transfer limitations. ^[55] High methane production cost by thermal conversion method compared to other conventional methods. ^[24] Economic perspective challenges in converting CO ₂ into CO. ^[60] High energy consumption since it operates at high temperature and pressure. ^[48]

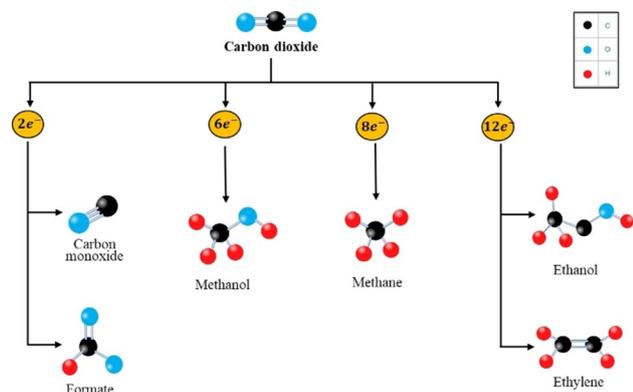


Figure 1. Reaction pathways of CO₂ electroconversion into different products based on the number of transferred electrons.

ethanol. According to the CO₂ reduction routes in Figure 2,^[72,73] the critical step in the process is the initial reaction step of producing the radical anion of carbon dioxide ($\text{CO}_2^{\cdot-}$), which is required to control the high overpotential that is competing with hydrogen evolution.^[72] The overpotential value can be significantly reduced by selecting a suitable catalyst. This initial step can be divided into two pathways: formate production pathway and carbon monoxide pathway. For carbon monoxide pathway, the produced CO gains four electrons to produce $\text{CH}_2(\text{ads})$, which is used for the production of ethylene, ethane and methane. For the formate pathway, it has three mechanisms including radical anion (Pathway 1), protonation step (Pathway 2) and electrode surface bonding (Pathway 3).^[74] Radical anion mechanism depends on transferring protons to a radical carbon atom ($\text{CO}_2^{\cdot-}$) since the oxygens are connected to the electrode surface. Thus, the protonation occurs on the carbon atom to produce HCOO^{\cdot} intermediate. This intermediate

further receives another transferred electron and a proton for the formation of formic acid. This indicates that less energy and electricity are needed to produce formate and formic acid, which makes this pathway more favorable in terms of product selection. Mainly, after the formation of the product on the catalyst surface, it is released to the electrolyte. Therefore, the product selectivity can be determined according to the used catalyst and electrode potential.^[11,13–18,75] It is worth mentioning that there are other proposed pathways for formate production such as the formation of OCHO^{\cdot} intermediates (pathway 2) and electrode surface bonding (pathway 3). This pathway, unlike radical anion pathway, has OCHO^{\cdot} as an intermediate after transferring an electron to the HCOO^{\cdot} intermediate. After the protonation step takes place at the produced OCHO^{\cdot} to obtain formic acid. The last possible mechanism pathway depends on the bonding site of the electrode surface with carbon atom of radical anion. When this bonding takes place, the protonation of oxygen atom of the radical anion occurs to form COOH^{\cdot} intermediate. This intermediate is further reduced to formic acid by transferring protons and electrons.

3.2. Electroreduction Products

3.2.1. Carbon Monoxide

Carbon monoxide is one of the possible products that results from the electroconversion of CO₂. Lee et al.^[76] described a modified technology for CO₂ conversion that focused on applying a membrane electrode rather than a conventional reduction of CO₂ in liquid phase. The membrane electrode involves anion exchange membrane (AEM) and gas diffusion electrode (GDE). The catalyst materials of the cathode are

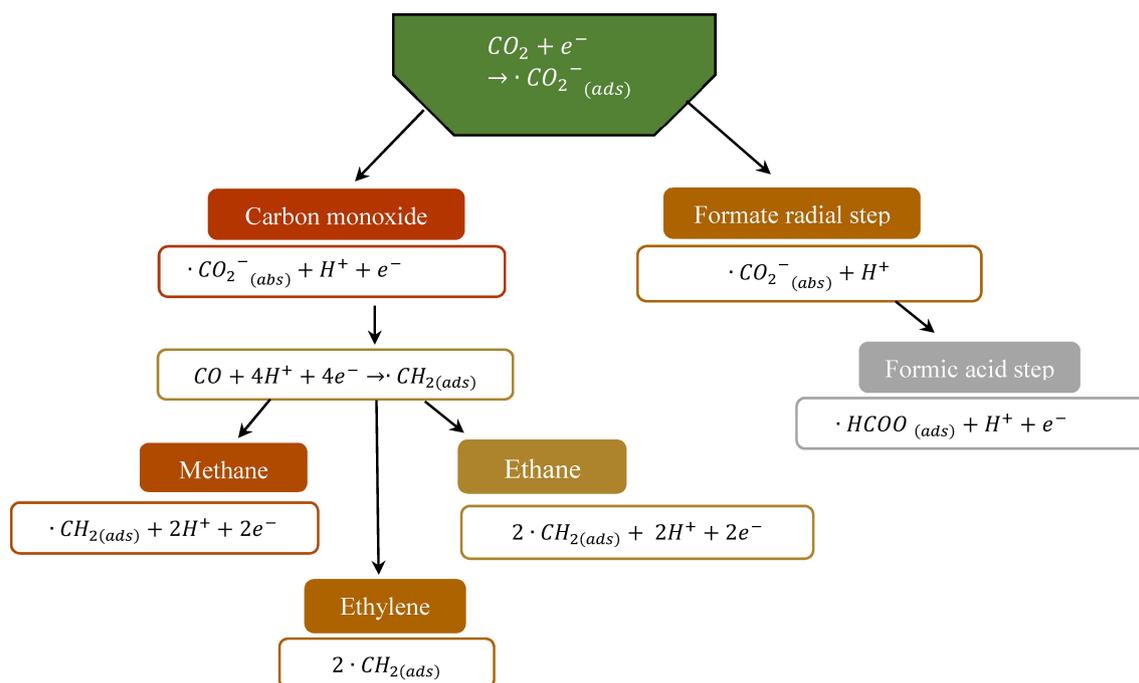


Figure 2. Flowchart of the reduction reaction steps of CO₂ electroconversion into carbon monoxide, formate/formic acid, methane, ethylene and ethane.

applied as powders including Ni, Pd, Zn, Ag, Sn, Ru, Cu, Pt. CO₂ is reduced to CO with a Faradic efficiency (FE) higher than 95%. The formation of carbon monoxide in the KHCO₃ solution saturated with the CO₂ resulted in a substantial FE of about 84% with Cu₂Cd/Cd/Cu bimetallic catalyst.^[77] Furthermore, the performance of the electroreduction with the Ag/Co₃O₄ catalyst reported excellent results with high selectivity of CO in the used solution based on KHCO₃.^[78] The FE reached a value greater than 95% with the Ag foam electrocatalyst.^[79] This electrocatalyst was produced by the combination of 1-butyl-3-methylimidazolium tetrafluoroborate ([Bmim][BF₄]) ionic liquid. However, another highly active catalyst, pyrrolidinonyl-nickel phthalocyanine (PyNiPc), achieved close to 100% FE.^[69]

3.2.2. Methanol

The other product options by electro-conversion is methanol. Marepally et al.^[80] studied the production of methanol using Cu-based catalysts in a continuous reactor with a pure flow of carbon dioxide and electrolyte consisting of an aqueous solution of KHCO₃. The main results showed that at average current density, the formation of methanol could be highly achieved with Cu nanoparticles, which were prepared as nanowires, compared to the classical preparation of the commercial carbon tube (Cu/CNT). In another study, Zarandi et al.^[81] evaluated the electroreduction of CO₂ to methanol with platinum nanoparticles catalyst on histamine-reduced graphene oxide plates (Pt@His-rGO) supported by a glassy carbon (GC). The optimization study was obtained by examining several parameters, such as the applied potential, pH and CO₂ purging time. The results showed that throughout the production of

methanol, lower applied potential is required for the nano-composite electrocatalysts. Table 2 summarizes numerous literature work on the electrochemical conversion of CO₂ to syngas or methanol in terms of different experimental parameters.

3.2.3. Formic Acid/Formate

The formic acid or formate is one of the major products from electroconversion, and it is the main focus of this review. The production of formic acid or formate depends on the pH value during the experiment; formic acid can be produced at low pH (less than 3), while at higher pH values (weak alkaline) the production of formate can take place.^[34,37] Figure 3 shows reaction of formate production. Mainly, the carbon dioxide gains an electron and gets into an excited state (intermediate step), where the formate is produced by combination with a

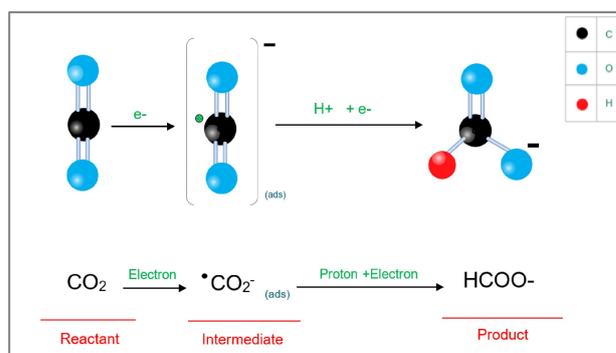


Figure 3. CO₂ reduction reaction into formate by electroconversion method

Type of products	Current density	Applied potential*	Type of catalyst	Faraday efficiency	Electrolyte	Ref.
Syngas with water	6–100 mA.cm ⁻²	–1.1 to –0.6 V vs. RHE	CuZnO/CNT	15–85 %	0.1 M KHCO ₃	[82]
Carbon monoxide	20.4 mA.cm ⁻²	–1.28 vs. NHE	ZnO/Cu	47.2 %	0.5 M KHCO ₃	[83]
Carbon monoxide	0.04–0.08 mA.cm ⁻²	–1.8 V vs.SCE	Ag/Co ₃ O ₄	21.3 %	0.1 M KHCO ₃	[78]
Carbon monoxide	6.72 mA.cm ⁻²	–2.3 V vs. Fc/Fc ⁺	Zn	83 %	PC/TBAP catholyte 0.1 M H ₂ SO ₄ anolyte	[84]
Carbon monoxide	–11.4 mA.cm ⁻²	–1.2 V vs RHE	CdS-CNT	95 %	0.1 M KHCO ₃	[85]
Carbon monoxide	8 mA.cm ⁻²	–1.0 V vs. RHE	Cu ₂ Cd/Cd/Cu	84 %	0.1 M KHCO ₃	[77]
Carbon monoxide	19.1 mA.cm ⁻²	–0.93 V vs. RHE	PyNiPc/CNT	≈100 %	0.5 M KHCO ₃	[69]
Carbon monoxide	~2.6 mA.cm ⁻²	–0.4 V vs. RHE	ZrO ₂ /N–C	64 %	0.5 M KHCO ₃	[86]
Methanol	250 μA/0.044 cm ⁻²	–0.3 V vs. Ag/AgCl (KCl sat)	Pt@His-rGO	37 %	0.1 mol/L KNO ₃	[81]
Ethanol	2.3 mA.cm ⁻²	–0.8 V vs RHE	HMMP Cu ₅ Zn ₈	46.6 %	0.1 M KHCO ₃	[87]
Methane	100 mA.cm ⁻²	–1 V vs.RHE	Cu(II)	42 %	1 M KOH	[88]

* Reference electrode not normalized against a single one due to non-uniformity in reporting in different studies.

proton and another electron. This explains the reason why formate production by CO₂ electroreduction requires two-electron transferred, which is due to the intermediate step. The involvement of proton (H⁺) in the intermediate step is essential in formate production. During the reduction reaction, the oxygen evolution potential (OEP) of the anode can affect the required voltage. Higher values of OEP lead to higher applied voltages, which corresponds to more H⁺ transfer across the proton membrane resulting from strengthening of the electric field.^[89] It is noteworthy that hydrogen evolution reaction is more thermodynamically favorable than the reaction of formate or formic acid,^[37] therefore, more effort is needed to overcome the production of hydrogen by selecting a suitable electrocatalyst and reactor configuration. For more details, Figure 4 illustrates an electrolysis cell for the CO₂ electroreduction into formate using tin-based (Sn) electrocatalyst as cathode and platinum (Pt) as anode with an ion exchange membrane.

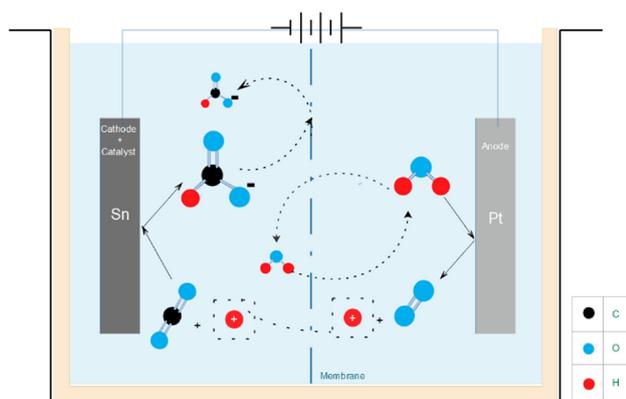


Figure 4. A schematic diagram of the CO₂ electrocatalytic reduction into formate on Sn-based catalyst.

4. Types of Electrolytic Cell Reactor for Formate Production

The configuration of the electrolytic cell plays a critical role in CO₂ electroreduction since the arrangement of the electrodes,^[90] types of membrane^[91] and types of electrolyte^[92] affect the performance of electroreduction and the desired product yield. The most common reactors types are: (i) H-type cell, (ii) solid-oxide electrolysis cell (SOEC), (iii) differential electrochemical mass spectrometry cell (DEMS), (iv) Microfluidic flow cell (MFC), (v) polymer electrolyte membrane (PEM) and (vi) GDE.^[93] To produce formate/formic acid, the main used configuration in the literature is based on H-type cell (lab-scale), filter-press cell and gas diffusion electrode, as mentioned in Table 4. The simplicity of H-type allows it to be tested at lab-scale to examine the performance of electrocatalyst selectivity; however, for large and long-time scales, further development are required to reduce the mass transfer resistance.^[93] The main challenges of CO₂ electrolyzers are electrode structure, reactor configuration, reaction conditions (temperature pH and pressure), and electrolyte selection.^[94] The local pH, near the cathode surface, is often different from the bulk pH due to diffusion limitation and proton consumption or OH⁻ generation.

4.1. Cell Configuration

The cell configuration depends on the selected reactor type that is efficiently used in producing formate or formic acid. According to the literature,^[95,96] the majority of the studies focused on applying 3 electrodes and H-type cell, while others applied GDE^[14,97] to improve the FE of formate production. Only a limited number of studies focused on using the filter press cell for a continuous reaction.^[98,99] Thus, this section describes the three commonly used types, as shown in Figure 5, including H-type cell, GDE and filter press cell.

H-type cell is considered as the most common applied reactor for electroreduction of CO₂,^[93] which consists of two separated compartments having three electrodes.^[95] The anodic

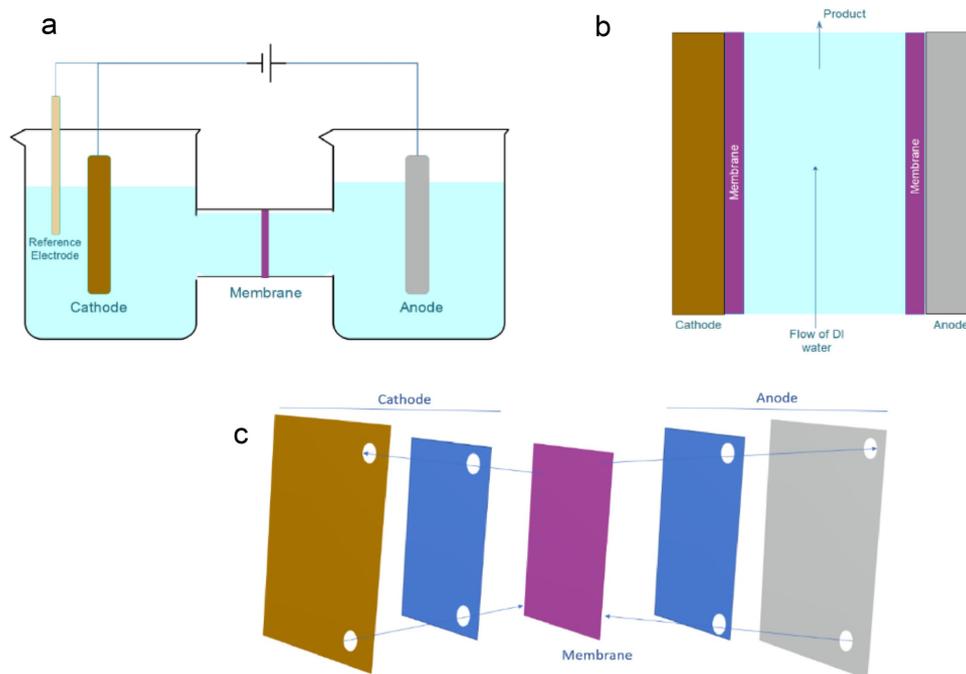


Figure 5. Sketch of reactors type a) H-type cell b) gas diffusion electrode cell and c) filter press cell

compartment contains one electrode, while the cathodic compartment contains two electrodes, a reference electrode and a working electrode. These two compartments are separated by a membrane, which controls the ions movement to prevent the further oxidation of the desired product.

GDE is one of the reactor types with highly improved mass transfer efficiency in comparison to H-type cell.^[93] The configuration of GDE allows CO₂ gas to be directly contacted by the inner electrode, which has a porous membrane that allows the electrolyte to be contacted with CO₂ gas at the outer side of the electrode. As a result, having a boundary consisting of three phases including gas phase (CO₂), liquid phase (electrolyte bulk) and solid phase (electrode) can lead to the production of formate due to the higher mass transfer at the boundaries.

Filter press cell is investigated frequently as parallel plates of selected electrodes (mainly different types depending on the case study) that are separated by a membrane.^[99] Usually, this type of reactors can run in a continuous mode, consisting of inlet and outlet for electrolyte path and inlet for CO₂ (feed) with another path for outlet with the formate or formic acid product.

4.2. Types of Membranes and Electrolytes

The selection of the membrane can significantly affect the results since most of the reactors depend on the membrane type between the chambers or even between the plates (in the case of filter press cell) to control the movement of ions. Thus, the types of membrane and electrolyte need to be carefully selected in order to obtain high performance electroreduction of CO₂ into formate. Proton exchange membrane, cation exchange membrane and bipolar membrane are used in

electroconversion of CO₂ into formate. Yang et al.^[100] showed that the membrane thickness can affect the FE of the formate production. The study was based on the performance of different types of membrane including Nafion 115, 212 and 324 on the anodic chamber at applied cell voltage of 3.5 V obtaining a high FE around 94%. The results showed that the thicker membrane, which is Nafion 324 with a thickness of 150 μm , obtained less crossover of formate ions from cathodic compartment to anodic compartment compared to the thinner membrane. Ramdin et al.^[101] compared the performance of cation exchange membrane and bipolar membrane. They concluded that bipolar membrane can be applied in large-scale process compared to cation exchange membrane due to its capability to have less amount of liquid crossover and its ability to maintain the pH gradient. Besides the significant role of membranes, the electrolytes also affect the performance significantly. As mentioned before, the competitive reaction (hydrogen evolution reaction) is required to be minimized, since it affects the formate production. According to Wang et al.,^[34] high overpotential electrolyte could increase the hydrogen evolution; however, the electrolyte could be considered as a support for the catalysts and system stabilization.

Electrolytes could be divided into two types: aqueous and non-aqueous electrolyte. Aqueous electrolytes have high electrical conductivity properties and they have different CO₂ saturation affinity. On the other hand, nonaqueous electrolytes are less conductive compared to aqueous electrolyte. Potassium carbonate (KHCO₃) is one of the most common aqueous electrolytes and it can be saturated by CO₂ within a concentration range of 0.1–0.5 M. Ju et al.^[102] investigated a solid polymer electrolyte membrane-based continuous-flow reactor

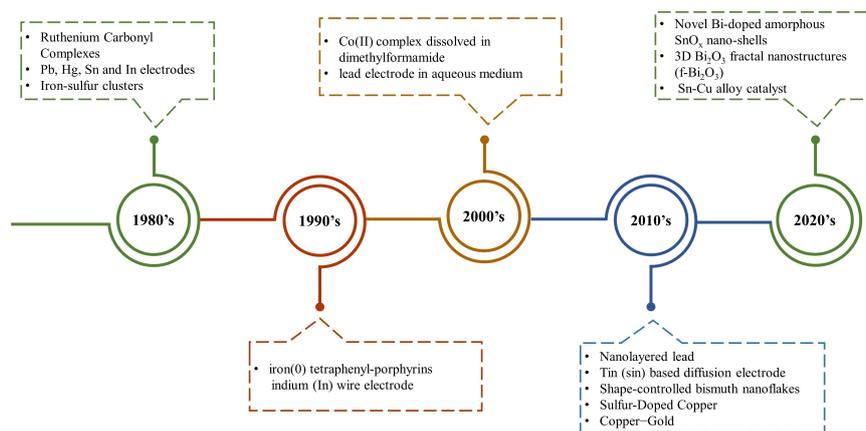


Figure 6. Timeline for the application of most common electrocatalyst of CO₂ conversion into formate

which is believed to offer an attractive pathway for CO₂ conversion into value products at low temperature.

5. Types of Catalysts Used for Formate/Formic Acid Production

The type of electrocatalysts plays a significant role in the performance of the electrochemical conversion, as well as it affects the type of final product from the CO₂ reduction.^[34,71,95] Effective electrocatalysts are expected to give high performance at low overpotential with high selectivity, activity and stability.^[71,72] The electroreduction of CO₂ to produce formate was first reported in 1985.^[103] In 1987, Ikeda et al.^[104] studied the formation of formic acid using several metallic electrodes such as lead, mercury and titanium. At the end of 1990's, iron was developed as an electrocatalyst using iron(0) tetraphenylporphyrins.^[105] At the early of 2000's, Co(II) complex was investigated as an electrocatalyst while dissolved in aqueous solution.^[106] Between 2008 and 2010, lead (Pb) electrodes were examined in aqueous medium.^[107,108] While between 2011 and 2014, tin electrodes were mostly studied.^[109,110] In 2017, Kim et al.^[111] used shape-controlled bismuth nanoflakes, which helped in increasing the process efficiency. In 2018 and 2019, copper-based catalyst was doped in sulfur and gold to enhance the selectivity towards formate.^[112,113] In 2020, Yang et al.^[114] synthesized a homogenous nanoparticles alloy using bismuth -doped amorphous SnO_x nanoshells. Figure 6 illustrates the electrocatalyst application timeline of CO₂ into formate. Through the decades, the electrocatalysts were developed via using main set of strategies, as shown in Figure 7, to enhance the CO₂ conversion into formate. For a detailed discussion about catalyst design and fundamental principles, the readers are directed to Zhang et al.^[115]

Electrocatalysts can be divided into homogenous and heterogenous catalysts. Homogenous catalysts are made of organic compounds or organometallic materials that have an intrinsic molecular structure, and they can be dissolved in

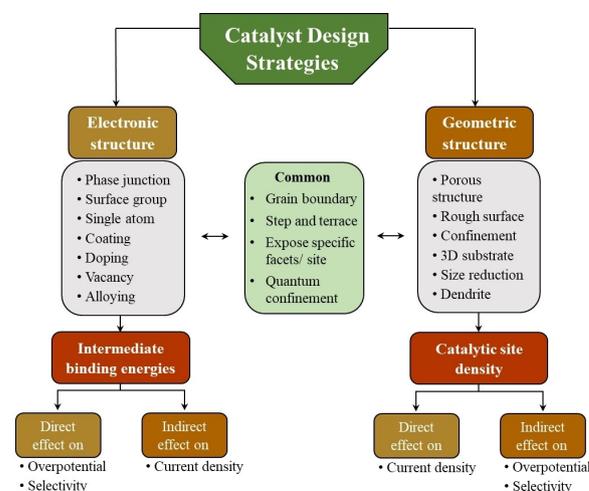


Figure 7. Proposed catalyst design strategies for CO₂ electroconversion.^[115]

electrolytes to enhance the exchange of protons and electrons.^[16] It has been reported that the reduction of CO₂ on homogeneous catalysts exhibits favorable kinetics with low overpotentials.^[16] However, they are considered to be more expensive than heterogenous electrocatalysts. Hence, utilizing heterogenous catalysts is more economically and environmentally viable at large scales. The heterogenous electrocatalysts fall under different categories including molecular catalysts,^[71] metallic catalyst^[34,71,116] carbon-based catalysts and alloy electrocatalysts.

Alloys catalysts, such as palladium-tin alloy, show more efficient performance compared to pure metallic catalysts (tin catalyst).^[117,118] In addition, using alloys helps in reducing the manufacturing cost of the electrocatalysts^[82] and controls the binding energies for intermediates in electrochemical CO₂ reduction.^[82,119] To improve the catalyst performance, catalyst supports are used to provide more active sites^[120] which will consequently promote high surface area.^[58] Carbon is usually the main support material and it can exist in different forms including carbon nanotube, carbon black and reduced graphene oxide (rGO) (as shown in Figure 8). A study conducted by

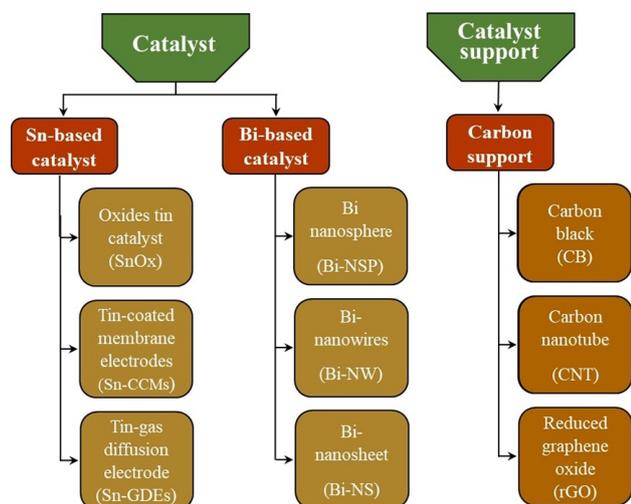


Figure 8. Common catalysts and catalyst support for CO₂ electroconversion into formate.

Jiménez et al.^[121] examined the effect of using different carbon supports on the catalytic activity of copper; the rGO support showed tendency toward formic acid production. In this section, the following catalysts: tin (Sn),^[96,117,122–125] bismuth (Bi),^[126,127,136–142,128–135] indium (In)^[143–146] and copper (Cu)^[121,147–150] are discussed in more details.

5.1. MOF Catalyst

Metal–organic framework (MOF) catalysts can be synthesized through bridging cation metals with organic linkers.^[151] MOFs show high stability when having different pore sizes and topology at wide range of temperature and pH. Table 3 shows the reported MOFs catalysts for electroconversion, photocatalytic and hydrogenation of CO₂.

5.2. Tin (Sn)-Based Electrocatalyst

Sn-based catalysts are considered to be non-toxic with high selective properties that can tolerate aqueous electrolyte at low overpotentials.^[95] According to Liu et al.,^[72] Sn-based catalyst has one of the highest selectivity toward formic acid/formate, which can be applied efficiently for CO₂ electroconversion. This efficiency is affected by the reaction conditions and electrode

properties, which can significantly change the Sn reactivity. Its FE, which is a function of structure and morphology of catalyst, varies between 18–95%.^[116] FE, which is shown in Equation 1, is defined as the number of transferred electrons to produce a certain number of formate moles at a certain electric charge which is provided during the reaction.^[157]

$$f = nFn_{\text{formate}}/Q \quad (1)$$

Where the term n represents the number of transferred electrons, F shows the Faraday constant, n_{formate} indicates the produced moles of formate and Q refers to the total passing charge through the reaction.

The performance of different forms of Sn-based catalyst, such as pure, oxide or alloy catalysts, were investigated in the literature at different experimental conditions to obtain formate.^[96,122–125,158–161] According to Zhao et al.^[95] Sn-based catalyst was prepared by electron-deposition method on Cu film. The film had a series of a deposited Sn catalyst, which achieved an optimum current density of 15 mA·cm⁻² and FE around 91%. Yang et al.^[100] further improved the Sn catalyst by using a novel cell composed of three compartments. The cell configuration achieved higher FE about 94% at a current density of 140 mA·cm⁻². This was obtained by applying ionomer (imidazole-based) to nanoparticle Sn catalyst at GDE to improve the performance of CO₂ reduction into formic acid. In addition, the efficient use of ion membranes helped in the transfer of ions between the chamber cells. It is worth noting that the authors did not use any salt electrolytes and only used deionized water. It is also important to note that authors recommended further development for GDEs as they observed liquid flooding during the reaction time. Although GDEs showed good performance by allowing the direct contact between the catalyst and CO₂ which increases the mass transfer, they still require more improvement. According to Castillo et al.,^[162] the implementation of GDEs with Sn-carbon support needs more research to improve the FE at high current densities. The authors work showed low efficiency of 70% compared to Yang et al.^[100] at relatively high current density. Consequently, Guillermo et al.^[158] described more advanced steps to improve formate selectivity by applying Sn-Catalyst Coated Membrane electrode (Sn-CCMs). This configuration is based on the utilization of solid polymer electrolyte, which reduces the effect of CO₂ solubility on the reaction rate. Although Castillo et al.^[162] and Guillermo et al.^[158] used the same Sn-based electrode (Sn/C-NPs and Sn-CCMEs), the Sn-CCMEs data did not show any improvement with respect to FE and

Table 3. Illustrated metal-organic framework used in CO₂ conversion.

Conversion type	Organic framework	Additions	Product	Ref.
Electroconversion	Benzne-1,3,5-tricarboxylic acid Cu ₃ (BTC) ₂	Graphene oxide	Formic acid	[152]
Photocatalytic	Cu/Cu ₂ O	Nanoparticles	Carbon monoxide	[153]
	manganese bipyridine complex Mn(bpydc)- (CO) ₃ Br	robust Zr(IV)-based metal	Formic acid	[154]
Hydrogenation	Copper- Alkoxide-Functionalized	–	Formic Acid	[155]
	chromium dicarboxylate MOF MIL-53(Cr)	cyclopentadienyl iron tricarbonyl complex	Formate	[156]

Table 4. Tabulated data of current density, applied potential and catalysts for Formate production by CO₂ ER.

Current density	Applied potential	Type of catalyst	Faraday efficiency	Electrolyte	Membrane	Cell-type	Ref.
15 mA.cm ⁻²	-1.4 V vs.SCE	Sn catalyst	91 %	0.1 M KHCO ₃	Nafion 211 proton exchange membrane	H-type	[95]
8.3 mA.cm ⁻²	-1.4 V vs.RHE	Oxide-derived -Sn-Pb-Sb-5 V	91 %	0.1 M KHCO ₃	Cation ion exchange membrane (CEM)	Conventional type	[161]
500 mA.cm ⁻²	0.5 V vs.RHE	SnO ₂	90 %	Aqueous	Bipolar membrane	GDE	[14]
11.2 mA.cm ⁻²	-1.2 V vs.RHE	Sn/SnOx	89.6 %	0.3 M NaOH	Nafion 117 cation exchange membrane	H-type	[96]
45 mA.cm ⁻²	-	Sn-CCME	49.4 %	Solid polymer electrolyte, SPE	Nafion 117 membrane	Continuous filter-press cell	[158]
150 mA.cm ⁻²	-1.5 V vs. NHE	Sn/C-GDEs	70 %	Anolyte: 1 M KOH Catholyte: 0.5 M KCl + 0.45 M KHCO ₃	Nafion 117 membrane	Continuous and single pass filter-press cell	[162]
5.4 mA.cm ⁻²	-1.8 V vs SCE for	Nano-SnO ₂ /black carbon	93 %	0.1 M NaHCO ₃	-	Flow cell or GDEs.	[163]
10.2 mA.cm ⁻²	-1.8 V vs SCE for	Nano-SnO ₂ /graphene	93.6 %	0.1 M NaHCO ₃	-	Flow cell or GDEs.	[163]
140 mA.cm ⁻²	3.5 V	Sn/GDE	94 %	Deionized water	Nafion® 115, 212, and 324 membranes	Novel 3-compartment formic acid cell design	[100]*
90 mA.cm ⁻²	3.1 V	Bi nanoparticles (Bi/C-NPs)	89.5 %	Anolyte: 1 M KOH Catholyte: a solution of 0.5 M KCl + 0.45 M KHCO ₃	Cation exchange membrane, Nafion 117	Filter press cell with single pass electrolyte	[99]
200 mA.cm ⁻²	-0.28 V vs. RHE	Bi ₂ O ₃	93 %	0.5 M KHCO ₃	-	flow cell	[128]
15.2 mA.cm ⁻²	-1.8 V vs. SCE	Bi nanodendrites-TCP	96.4 %	0.5 M NaHCO ₃	Proton exchange membrane Nafion 115	Conventional type	[126]
95 mA.cm ⁻²	-0.82 V vs. RHE	Biden/Pbporous	92 %	0.5 M KHCO ₃ (CO ₂ super-saturated)	Nafion 117	High-pressure flow cell	[129]
14.4 mA.cm ⁻²	-1.5 V vs.SCE	Bi@NPC	92.0 %	0.1 M KHCO ₃	Nafion	-	[170]
37.8 mA.cm ⁻²	-1.7 V vs.SCE	BiO _x	89.3 %	1 M KHCO ₃ , saturated CO ₂	Nafion 117	Gas-tight H-type cell	[174]
-3.54 mA.cm ⁻²	-1.9 V vs. Ag/AgCl	In	72.5 %	0.5 M KHCO ₃	Nafion N117	GDE	[97]
-12 to -1	-0.82 V vs. RHE	In/In ₂ O ₃ -x	89.2 %	Catholyte: 0.5 mol/L NaHCO ₃ Anolyte: 0.5 mol/L NaOH with 0.2 mmol octylamine	-	-	[144]
5.2 mA.cm ⁻²	-0.64 V vs.RHE	SW-Cu ₂ O/Cu	97.7 %	0.5 M KHCO ₃	Nafion Membrane	Full cell (high pressure cell)	[11]

* formic acid production by CO₂ ER

formate rate at similar current density. For example, the rate of formate production has decreased by 75% when using Sn-CCMEs compared to Sn-GDEs. This can be explained by the low current density that Sn-CCMEs operate at compared to Sn/C-NPs. Nonetheless, Sn-CCMEs can produce formate of high purity at low current densities (45 mA.cm⁻²) while consuming significantly lower energy. In addition, further research work is needed to improve FE and the formate production rate at low energy requirement.

There are other forms of tin-based catalysts that can also be applied for electro-conversion of CO₂ as illustrated in Figure 8 < xfigr8. Sn oxide electrocatalyst can be synthesized to nanoparticles by hydrothermal synthesis methods.^[163] This type of catalyst can be supported by carbon support to increase the surface area due to their 3D porous structure. The main advantage of using tin oxide is in its capacity of producing formate at low overpotential (0.34 V).^[163] The results showed that the graphene support with nano tin oxide had higher FE and current density than black carbon support. Rasul et al.^[161]

reported the first inexpensive electrocatalyst alloy for CO₂ electroreduction with low coats as Sn-Pb-Sb alloy.

The Sn-oxide derived alloys outperform the Sn-metal alone as it exhibits a reduction in hydrogen production at high overpotential. The oxidized alloy is also more favorable towards formate production since the oxidation is facilitated by the easy transfer of electrons within the metal alloy. Proietto et al.^[98] investigated for the first time the formation of formic acid by a pressured filter cell. The cell consisted of Sn cathode and 0.9 liters of electrolytic solution as a continuous recirculation. The results verified that high production of formic acid can be achieved at high current density and high CO₂ pressure. Pavithra et al.^[164] reported a Sn-based catalyst synthesized by microwave polyol method. SnO₂-CNT composites catalyst has been utilized to enhance the electroconversion of CO₂ into formate. This catalyst has been examined by varying the particle size and exhibited the ability to reduce the activation barrier for the formation of CO₂*- due to the availability of oxygen vacancy. The results have indicated that SnO₂-CNT catalyst with

5.26 nm particle size at a pH of 11, had a higher FE (76%) compared to others with different particle sizes; it also exhibited high stability for a 10-hour run with 60% of FE.

5.3. Bismuth (Bi)-Based Electrocatalyst

Another promising alternative catalyst for CO₂ conversion is bismuth (Bi), which is a low cost and nontoxic material with high selectivity.^[165] However, Bi-based catalyst has several limitations including lengthy preparation times using energy intensive processes which hinders its applicability for large-scale applications.^[165] Thus, further work is required to facilitate the application of large-scale of Bi-electrocatalyst. Another limitations of the Bi-based electrodes is the limitation in mass transfer between the reactants.^[166,167] This challenge can be solved by utilizing GDE.^[101,166] In general, bismuth-based catalyst can be prepared in three different shapes, as shown in Figure 8.^[168] Several experimental work by Piao et al.^[169] reported that Bi electrodes are highly porous and possess high electrocatalytic activity. Furthermore, Zhang et al.^[170] obtained formate by using Bi nanoparticles fixed in nitrogen-doped porous carbon (Bi@NPC). The study showed that the formate was detected at low potential and high current density, which improved the reaction compared to Bi nanoparticles (Bi-NP). The preparation of Bi-based catalysts might require large amount of energy, which makes it less attractive as it is considered to be energy- and time-consuming. However, Zhu et al.^[165] investigated networked Bi-based catalyst by using non-additional energy supply process, which can save time and energy. The results showed that at a current density of $68.51 \pm 4.04 \text{ mA}\cdot\text{cm}^{-2}$ the networked Bi-based electrode (N-Bi, Sn/Cu mesh) achieved FE of approximately 100%. Furthermore, a catalyst performance can be enhanced by applying two-dimensional engineering of materials through reducing the catalyst dimensionality which provides high activity. Zhang et al.^[171] reported the enhancement of nanosheet catalyst of few-layer bismuth through reducing the catalyst dimensionality by using electrochemical sub-carbonate exfoliation method. The developed nanosheet catalyst showed high activity and selectivity in reducing CO₂ into formate with 85% of FE. The catalyst stability was maintained for at least 12 hours.

5.4. Indium (In)-Based Electrocatalyst

Indium electrocatalyst can be used more efficiently in the oxide form (Indium(III) oxide) with 21.7% FE.^[144] Bohlen et al.^[97] reported the first electrodeposition process of deep eutectic solvents for the preparation of electrocatalyst using three materials including: copper, carbon and platinum. The results showed using indium with the copper sheet had higher FE (72.48%) than GDE (FE: 48.0%).

5.5. Copper (Cu)-Based Electrocatalyst

Copper-based catalyst can be used at high pressure with efficient performance during the electroconversion of CO₂ into formate.^[172,173] Li et al. discussed that the FE reached approximately 98% at 45 atm using SW-Cu₂O/Cu as a cathode.^[11] The results indicated that phase change of SW-Cu₂O/Cu cathode was slow, which affected the performance in terms of FE. Operation for a prolonged period caused decrease in the FE due to the decrease in the amount of the remaining Cu₂O on the surface.

6. Economic Study

The economic feasibility of CO₂ electroreduction into formate/formic acid is based on the selected configuration of electrolysis cell including the electrodes (anode and cathode materials) and membrane type. The lifetime of these features plays a significant role in estimating the plant lifetime for industrial applications.^[175] Rumayor et al.^[175] reported a techno-economic study for CO₂ electroconversion into formate, by comparing the conventional formate production process (by hydrolysis of methyl formate) and CO₂ electroconversion. The results showed that a cathode lifetime of about 4.45 years can save the consumable cost related to total cost around 10%. Durst et al.^[176] showed that formate production costs were lower than the market price by 2 to 4 factors. The estimated production costs around \$0.34, while the market price close to \$0.8–1.2.

7. Summary and Future Perspectives

In summary, carbon dioxide can be considered as a valuable byproduct that can be converted to useful products (i.e. formate, carbon monoxide and methanol) by several techniques including photoconversion, electrocatalytic conversion and bioconversion. Electroreduction of CO₂ into chemical products received considerable attention in recent years and can produce several products depending on the number of electrons transferred, while using a suitable catalyst that can reduce the cell overpotential. Cell configuration, current density and the types of catalyst play a significant role in the production of formic acid, carbon monoxide or hydrocarbons. Although CO₂ electroreduction into formate is gaining research momentum, more attention is needed at key research areas such as pilot-scale studies of CO₂ electroconversion into formate. In addition, there is an immense need to implement efficient and high performance electrocatalysts to achieve high formate production at high FE and current density. Among the studied catalysts, tin-oxide showed high catalytic activity toward CO₂ electroconversion reduction into formate, however, certain aspects require further investigations to get optimal conversion. Similarly, Sn-CCME performance can be improved by tuning FE. Finally, there is a need for detailed cost analysis studies of CO₂ electroreduction in literature especially at pilot scale.

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Conflict of Interest

The authors declare no conflict of interest.

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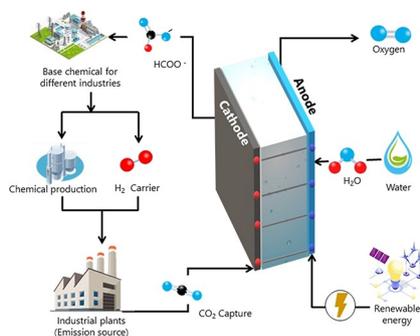
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REVIEWS

Carbon dioxide conversion is an essential step in transitioning to carbon neutrality, as it can convert the captured CO_2 into useful commodities. The electrochemical conversion of CO_2 to formate/formic acid has the potential to contribute to CO_2 mitigation and serve as a backbone chemical for several essential products including methanol. The conversion and product yield depend heavily on factors such as cell design and catalyst material.



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**Electroreduction of Carbon Dioxide
into Formate: A Comprehensive
Review**