

Cite this: *RSC Sustainability*, 2025, 3, 4426Received 11th May 2025  
Accepted 24th July 2025

DOI: 10.1039/d5su00334b

rsc.li/rscsus

## Perspective on electrochemical CO<sub>2</sub> reduction in CO<sub>2</sub>/O<sub>2</sub> mixed gas

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Electrochemical CO<sub>2</sub> reduction reaction (CO<sub>2</sub>RR) is a promising avenue to realize carbon neutrality. As the high-purity CO<sub>2</sub> in CO<sub>2</sub>RR may diminish the feasibility and economic viability, the direct conversion of CO<sub>2</sub> in O<sub>2</sub>-containing feed gas (CO<sub>2</sub>/O<sub>2</sub>) presents an attractive option. However, high CO<sub>2</sub>RR kinetic barriers and the challenges associated with O<sub>2</sub> reduction significantly hamper the effectiveness of CO<sub>2</sub>RR. Therefore, enhancing the selective CO<sub>2</sub>RR in CO<sub>2</sub>-O<sub>2</sub> mixed gas is critical. In this perspective, we first discuss factors of selective CO<sub>2</sub>RR in CO<sub>2</sub>/O<sub>2</sub>. Then, state-of-the-art interface design strategies for the selective CO<sub>2</sub>RR, including O<sub>2</sub> passivation, selective CO<sub>2</sub> adsorption and direct selective CO<sub>2</sub>RR, are highlighted. Finally, a brief discussion on the current challenges and outlook for future directions to achieve highly efficient and O<sub>2</sub>-tolerant CO<sub>2</sub>RR systems are presented.

### Sustainability spotlight

To realize the goal of a carbon-neutral society, converting excess CO<sub>2</sub> into valuable chemicals/fuels by biological, photochemical and electrochemical approaches has been extensively investigated. Electrochemical CO<sub>2</sub> reduction reaction (CO<sub>2</sub>RR) driven by renewable electricity is a promising avenue to catalyze CO<sub>2</sub> into high value-added products. As the high-purity CO<sub>2</sub> in CO<sub>2</sub>RR may diminish the feasibility and economic viability, the direct conversion of CO<sub>2</sub> in O<sub>2</sub>-containing feed gas (CO<sub>2</sub>/O<sub>2</sub>) presents an attractive option, which can reduce the costs of CO<sub>2</sub> purification greatly. However, high CO<sub>2</sub>RR kinetic barriers and the challenges associated with O<sub>2</sub> reduction significantly hamper the effectiveness of CO<sub>2</sub>RR. Therefore, it is essential to enhance the O<sub>2</sub> tolerance of CO<sub>2</sub>RR systems.

## 1 Introduction

Global economic growth and human activities significantly increase the CO<sub>2</sub> concentration in the atmosphere, leading to serious environmental problems, such as global warming, sea level rising and extreme climate.<sup>1–6</sup> Converting the excess CO<sub>2</sub> into valuable chemicals/fuels by biological,<sup>7,8</sup> photochemical<sup>9,10</sup> and electrochemical approaches<sup>11–15</sup> has been extensively investigated.<sup>16,17</sup> Among them, electrochemical CO<sub>2</sub> reduction reaction (CO<sub>2</sub>RR) driven by renewable electricity has been emerging as a promising way toward carbon neutrality through catalyzing CO<sub>2</sub> into high value-added products.<sup>18–24</sup> At present, most CO<sub>2</sub>RR systems utilize pure CO<sub>2</sub> as feedstock. However, the costs associated with CO<sub>2</sub> purification are significant, diminishing the economic benefits.<sup>25,26</sup>

The majority of human-induced CO<sub>2</sub> comes from flue gas (CO<sub>2</sub>/O<sub>2</sub>), therefore, the direct use of flue gas for feedstock of CO<sub>2</sub>RR can reduce the costs of CO<sub>2</sub> purification and represent a potential strategy for CO<sub>2</sub>RR applications. However, CO<sub>2</sub>/O<sub>2</sub> reduction is facing a few challenging issues: (i) dilute CO<sub>2</sub>

concentrations in CO<sub>2</sub>/O<sub>2</sub>, making the CO<sub>2</sub>RR kinetics sluggish, and (ii) energetically favorable O<sub>2</sub> reduction reaction (ORR).<sup>27</sup> It has been clarified that only 5% O<sub>2</sub> in CO<sub>2</sub> inhibits CO<sub>2</sub>RR completely.<sup>28–30</sup> Therefore, it is essential to enhance the O<sub>2</sub> tolerance of CO<sub>2</sub>RR systems.<sup>31</sup> To date, challenges of flue gas reduction such as low transformation efficiency and unavoidable ORR persist although the coupling between direct flue gas utilization and electrochemical conversion has been investigated.<sup>32</sup> Thus, improving conversion efficiency is still the largest obstacle for the selective CO<sub>2</sub>RR by using flue gas.

In recent years, numerous reviews have reported the electrochemical reduction of low-concentration CO<sub>2</sub> or CO<sub>2</sub>-containing gas mixtures with impurities. For instance, Wang *et al.* systematically analyzed the scientific challenges and innovative strategies for the direct electrochemical conversion of CO<sub>2</sub> derived from industrial flue gases.<sup>33</sup> Similarly, Li *et al.* reviewed key design strategies for CO<sub>2</sub>RR under dilute CO<sub>2</sub> conditions and in the presence of common gas impurities.<sup>34</sup> Despite O<sub>2</sub> being the most abundant impurity in industrial flue gas, with the O<sub>2</sub>/CO<sub>2</sub> ratio being even greater than 20%, they just paid less attention to the selective CO<sub>2</sub>/O<sub>2</sub> reduction in mixed gas systems. Therefore, the possible promotion effects of O<sub>2</sub> on CO<sub>2</sub>RR and potential strategies should be carefully considered in the design of catalysts, electrode structures and electrolyte compositions, and this is an important area of research that should be focused on in the recent future.

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In this perspective, we discuss the selective CO<sub>2</sub>RR on different systems using flue gas as feedstock, which is expected to provide insight into O<sub>2</sub>-tolerant CO<sub>2</sub>RR. Firstly, the factors of CO<sub>2</sub>/O<sub>2</sub> selective reduction performance are discussed. Then, strategies for designing the electrocatalytic reaction interface, such as surface modification that hampers O<sub>2</sub> transportation and enhances selective CO<sub>2</sub> adsorption or direct selective CO<sub>2</sub>RR, are discussed. Finally, the current challenges in achieving high O<sub>2</sub>-tolerant CO<sub>2</sub>RR performance and insights into realizing large-scale applications in the future are summarized. We hope that this perspective shall illuminate the pathways toward developing excellent O<sub>2</sub>-tolerant CO<sub>2</sub> electrocatalytic systems through the exploration of recent advances.

## 2 Factors affecting CO<sub>2</sub>/O<sub>2</sub> selective reduction performance

The volume concentration of CO<sub>2</sub> in flue gas emission varies between 5% and 35%, typically around 15%.<sup>35–38</sup> Furthermore, flue gas contains impurities, such as O<sub>2</sub> and balancing inert N<sub>2</sub>.<sup>39</sup> Considering that N<sub>2</sub> is hardly involved in the cathode reaction, the reduction of flue gas could be considered as CO<sub>2</sub>/O<sub>2</sub> selective reduction. The solubility of CO<sub>2</sub> in water is only 1.45 g L<sup>-1</sup> (273 K, 1 atm), indicating the limited concentration of CO<sub>2</sub> for CO<sub>2</sub>RR. At the same time, CO<sub>2</sub>RR is a complex process because of the multistep proton-electron transfer reactions as well as a variety of reaction paths. During the electrochemical reduction process, non-spontaneous electron transfer reactions are driven by an external power supply. The categories of CO<sub>2</sub>RR products mainly depend on the externally applied potentials and catalysts, as well as electrolyte composition (eqn (1)–(11) (*E* vs. SHE)). Most CO<sub>2</sub>RR systems still suffer from low Faraday Efficiency (FE) due to the competitive hydrogen evolution reaction (HER) (eqn (12) (*E* vs. SHE)) and multiple products (Fig. 1a). Importantly, ORR may also occur in flue gas reduction because it is thermodynamically more favorable than that of CO<sub>2</sub>RR (eqn (13) and (14) (*E* vs. SHE)), suppressing CO<sub>2</sub>RR simultaneously (Fig. 1b).

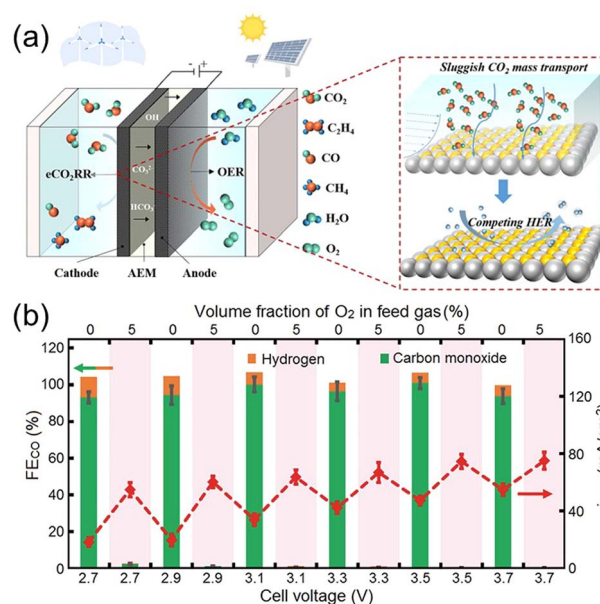
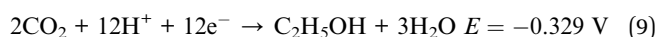
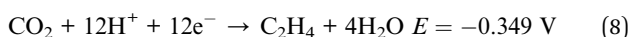
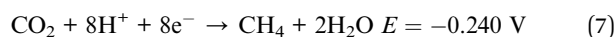
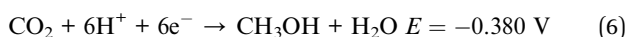
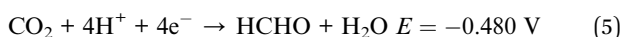
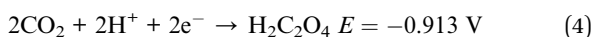
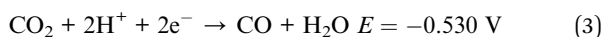
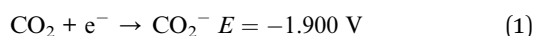
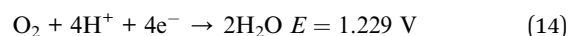
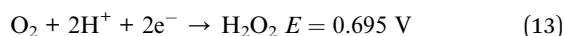
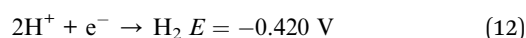
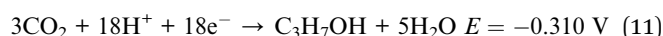
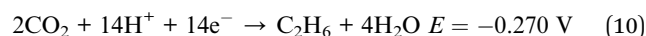


Fig. 1 (a) A typical design concept of a CO<sub>2</sub>RR system. Sluggish CO<sub>2</sub> mass transport and competitive HER constitute two fundamental challenges in CO<sub>2</sub>RR, especially for dilute CO<sub>2</sub> concentrations. (b) Faraday efficiency for CO production (FE<sub>CO</sub>), FE<sub>H<sub>2</sub></sub> and total geometric current density (*j*<sub>total</sub>) vs. cell voltage with and without O<sub>2</sub> in the feed gas. Compared to pure CO<sub>2</sub> feed (white background), the O<sub>2</sub>-incorporated system (5 vol% O<sub>2</sub>/CO<sub>2</sub>, pink background) shows near-zero FE<sub>CO</sub> and FE<sub>H<sub>2</sub></sub>, indicating complete dominance of O<sub>2</sub> reduction at this low ratio. The higher current density under O<sub>2</sub> is attributed to faster O<sub>2</sub> reduction kinetics versus CO<sub>2</sub>RR. (a) Reproduced with permission.<sup>40</sup> Copyright 2024, Elsevier. (b) Reproduced with permission.<sup>41</sup> Copyright 2019, Elsevier.



The electrocatalytic reaction happens at the solid–liquid–gas interface. Therefore, the design of the electrocatalyst–electrolyte interface is critical for selective CO<sub>2</sub>RR. By rationally designing the catalyst and optimizing electrocatalytic conditions, the following factors can be effectively leveraged to manipulate the reaction process and improve the activity and selectivity of CO<sub>2</sub>RR: (i) catalyst structure: surface active sites with specific element composition, crystallinity, defects, *etc.* can efficiently capture and enrich CO<sub>2</sub> to achieve high catalytic activity, selectivity and stability, by creating a low-O<sub>2</sub> environment through filtering out the O<sub>2</sub> molecules, or facilitating the conversion of key intermediates in CO<sub>2</sub>RR in the presence of O<sub>2</sub>. (ii) Electrolyte: electrolyte is another key component of CO<sub>2</sub>RR systems, which includes the main electrolyte and additives,



including cations, anions and small organic molecules. Cations and anions in the electrolyte that affect the pH value within the solid-liquid structure have a significant influence on the electrochemical reactivity by tuning the structure of the electrical double layer. Additionally, the reactants and intermediates may also dynamically interact with the solvent molecules and additives in the electrolyte at the interface, altering the electrochemical reactivity and selectivity.

### 3 Strategies for CO<sub>2</sub>/O<sub>2</sub> selective reduction

There have been a lot of strategies for improving the selectivity and activity of CO<sub>2</sub>RR by adjusting the catalyst structure with O<sub>2</sub><sup>-</sup> containing feed gas. Optimizing the CO<sub>2</sub>/O<sub>2</sub> ratio on the catalyst shall be an effective strategy for CO<sub>2</sub>/O<sub>2</sub> selective reduction, which could be achieved by O<sub>2</sub> passivation, selective CO<sub>2</sub> adsorption, and direct selective CO<sub>2</sub>RR.

#### 3.1 O<sub>2</sub> passivation strategy

The O<sub>2</sub> passivation strategy can enhance the selective CO<sub>2</sub>RR by directly or indirectly slowing down O<sub>2</sub> transport. Introducing hydrophilic nanopores with sluggish O<sub>2</sub> transport and constructing a selective penetration layer with the O<sub>2</sub> prohibition ability can effectively slow down O<sub>2</sub> transport for efficient CO<sub>2</sub>RR using flue gas.

**3.1.1 Hydrophilic nanopores.** CO<sub>2</sub> exhibits Lewis acidity due to the electrophilic carbon atom capable of accepting electron pairs, while O<sub>2</sub> is a non-polar molecule with minimal interaction with hydrophilic environments. Therefore, hydrophilic nanopore networks can selectively reduce O<sub>2</sub> mass flux to electrocatalytic centers by leveraging polarity-driven adsorption and size exclusion effects, thereby enhancing the efficiency and selectivity of CO<sub>2</sub> conversion.<sup>42,43</sup> Adding TiO<sub>2</sub> with hydrophilic nanopores on Cu catalysts can separate the O<sub>2</sub> and achieve good CO<sub>2</sub>RR selectivity. Xu and co-workers<sup>44</sup> developed a catalyst composed of an ionomer with hydrophilic nanopores and TiO<sub>2</sub> nanoparticles as support particles and Cu as the main electrocatalyst (Fig. 2). The ionomer layer slowed down the O<sub>2</sub> transport rate to the catalyst and enabled a more efficient conversion of CO<sub>2</sub> to C<sub>2</sub> products with a FE<sub>C<sub>2</sub></sub> of 68% and a non-*iR*-corrected full cell energetic efficiency of 26%.

**3.1.2 Selective penetration layer.** Constructing an O<sub>2</sub> selective penetration layer could also improve CO<sub>2</sub>RR selectivity by limiting O<sub>2</sub> penetration. Efficient O<sub>2</sub> selective penetration layers include: (1) specific frameworks with reversible photo-switching built to modulate the electron transfer rate and oxygen activation ability, and (2) microporous polymers with size-selective pores to filter O<sub>2</sub> and permeate CO<sub>2</sub> selectively. Zhu *et al.*<sup>45</sup> presented an O<sub>2</sub> passivation strategy to realize efficient CO<sub>2</sub>RR performance by feeding CO<sub>2</sub>/O<sub>2</sub> (a high FE<sub>CO</sub> of 90.5% with a *j*<sub>CO</sub> of -20.1 mA cm<sup>-2</sup> at -1.0 V vs. RHE) under UV/Vis irradiation, and using the photoswitching built block 1,2-bis(5'-formyl-2'-methylthien-3'-yl) cyclopentene (DAE) in the material (Fig. 3a-e). DAE reversibly modulates the electrical conductivity and O<sub>2</sub> activation capacity by the framework ring-closing/opening reactions. Specifically, upon irradiation with UV, the close-DAE-BPy-CoPor exhibits higher electronic conductivity than open-DAE-BPy-CoPor (under Vis irradiation) because of the strong charge delocalization in close-DAE moieties. Furthermore, density functional theory (DFT) calculations and *operando* ATR-FTIR experiments demonstrated that the excellent CO<sub>2</sub>RR performance of close-DAE-BPy-CoPor in co-feeding CO<sub>2</sub> and O<sub>2</sub> is attributed to the weak O<sub>2</sub> activation ability and high O<sub>2</sub> into \*OOH (the ORR limiting step) free energy, thus resulting in the excellent selective CO<sub>2</sub>RR performance in the presence of O<sub>2</sub>.

CO<sub>2</sub> enrichment by physical pore confinement can also achieve highly selective electrochemical CO<sub>2</sub> reduction under an aerobic environment. For example, inspired by the natural photosynthesis unit, Lu's group<sup>41</sup> designed a PIM-CoPc/CNT hybrid electrode as an "artificial leaf" to enrich CO<sub>2</sub> in the presence of O<sub>2</sub> (Fig. 3f-h), where the PIM layer played a pivotal role in realizing CO<sub>2</sub>/O<sub>2</sub> selective reduction. Serving as a molecular sieve with high gas permeability, the PIM layer effectively filters O<sub>2</sub> based on the molecular size and enriches CO<sub>2</sub> from the feed gas, and thus creates a low-O<sub>2</sub> local environment for the catalyst to achieve effective electrochemical CO<sub>2</sub>-to-CO conversion. With 5% O<sub>2</sub> in the CO<sub>2</sub> feed gas, a FE<sub>CO</sub> of 75.9% with a *j*<sub>total</sub> of 27.3 mA cm<sup>-2</sup> was achieved at a cell voltage of 3.1 V. Notably, an average CO<sub>2</sub>/O<sub>2</sub> selectivity of ~20 suggested that 95% O<sub>2</sub> in the feed gas was rejected by the PIM layer.

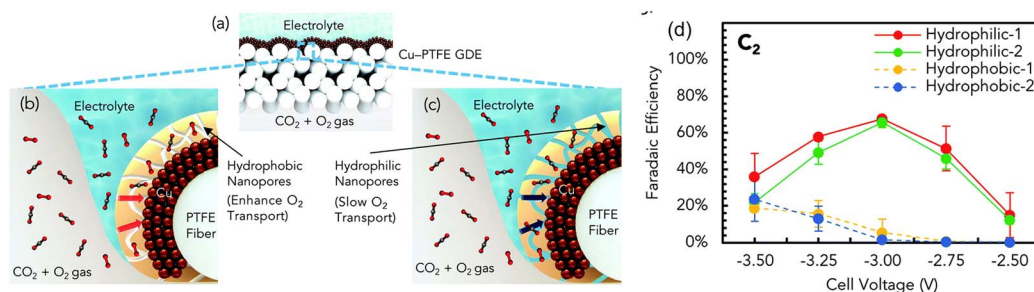
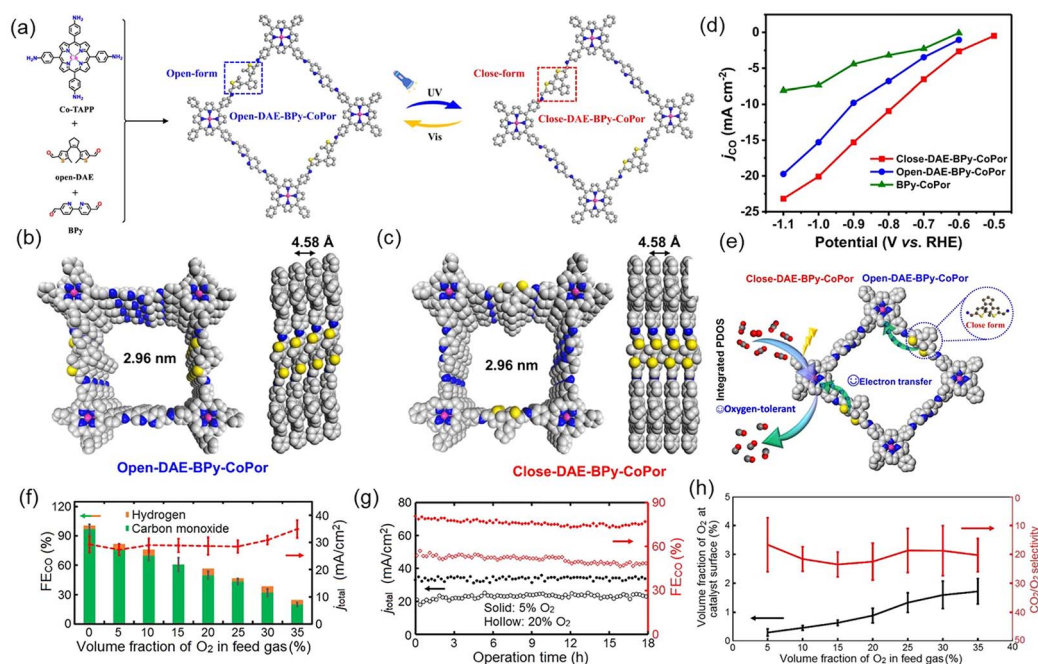


Fig. 2 (a) Schematic illustration of the Cu-PTFE GDE. Schematic of the GDE coated with the (b) hydrophobic and (c) hydrophilic nanoporous ionomer. (d) The FE toward C<sub>2</sub> products for different ionomers. Reproduced with permission.<sup>44</sup> Copyright 2020, The Royal Society of Chemistry.





**Fig. 3** (a) Synthetic route to open-DAE-BPy-CoPor and close-DAE-BPy-CoPor. Top and side views of (b) open-DAE-BPy-CoPor and (c) close-DAE-BPy-CoPor. (d) The  $j_{\text{CO}}$  of the BPy-CoPor, close-DAE-BPy-CoPor and open-DAE-BPy-CoPor under aerobic conditions. (e) Proposed schematic mechanism for the  $\text{CO}_2\text{RR}$  on close-DAE-BPy-CoPor under aerobic conditions. (f)  $\text{FE}_{\text{CO}}$ ,  $\text{FE}_{\text{H}_2}$  and  $j_{\text{total}}$  vs. volume fraction of  $\text{O}_2$  in the  $\text{CO}_2$  feed gas. (g)  $\text{FE}_{\text{CO}}$  and  $j_{\text{total}}$  during an 18 h electrolysis at  $\text{O}_2$  volume fractions of 5% (solid markers) and 20% (hollow markers). (h) Volume fraction of  $\text{O}_2$  at the catalyst surface vs. that in the feed gas, and  $\text{CO}_2/\text{O}_2$  selectivity of the PIM gas selection layer in the  $\text{O}_2$ -tolerant hybrid electrodes. (a–e) Reproduced with permission.<sup>45</sup> Copyright 2024, Springer. (f–h) Reproduced with permission.<sup>41</sup> Copyright 2019, Elsevier.

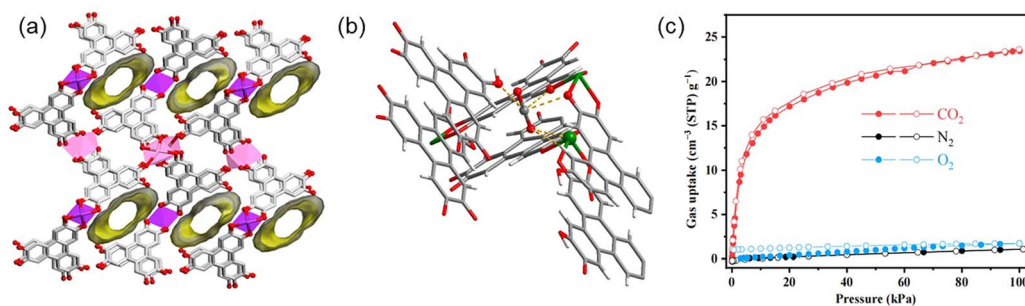
### 3.2 Selective $\text{CO}_2$ adsorption

Enhancing the selective  $\text{CO}_2$  adsorption on the catalysts by the design of specific morphological structures and surface coating/modification is another effective strategy to gain high-efficiency  $\text{CO}_2\text{RR}$  products in flue gas.

**3.2.1 Specific morphological structure.** Designing catalysts with specific morphological structures, like microporous architectures with the ability to capture  $\text{CO}_2$ , is able to promote the  $\text{CO}_2$  selective adsorption and reduction. Zhao *et al.*<sup>46</sup> prepared Bi-HHTP with a microporous conductive Bi-based metal-organic framework (Fig. 4), which only showed slightly lower  $\text{FE}_{\text{HCOOH}}$  values in a dilute  $\text{CO}_2$  (15 vol%,  $\text{CO}_2/\text{N}_2/\text{O}_2 =$

15 : 80 : 5, v/v/v) as the feedstock. Specifically, the  $\text{FE}_{\text{HCOOH}}$  still approached 90% with a current density of  $71 \text{ mA cm}^{-2}$  at a cell voltage of 2.6 V. It means that the oxygen concentration has a minor effect on the  $\text{CO}_2\text{RR}$  process. The open Bi sites and hydroxyl groups are exposed on the pore surface, playing a role as  $\text{CO}_2$  capture and conversion sites. DFT calculations showed that the relatively moderate binding strength of  $^*\text{OCHO}$  on Bi-HHTP made it favorable for further hydrogenation, thus achieving higher  $\text{CO}_2$ -to- $\text{HCOOH}$  selectivity.

**3.2.2 Surface coating/modification.** Surface coating or modification with alkalic groups can introduce strong chemical affinity to  $\text{CO}_2$ , a Lewis acid, that realizes selective  $\text{CO}_2$  adsorption. For example, Cao *et al.*<sup>47</sup> proposed a polyaniline



**Fig. 4** (a) 3D  $\pi$ - $\pi$  stacking structure of Bi-HHTP with 1D pores along the  $b$ -axis direction. (b) The adsorption site for the  $\text{CO}_2$  molecule in Bi-HHTP. (c)  $\text{CO}_2$ ,  $\text{N}_2$  and  $\text{O}_2$  adsorption (solid) and desorption (open) isotherms of Bi-HHTP measured at 298 K, respectively. Reproduced with permission.<sup>46</sup> Copyright 2024, American Chemical Society.



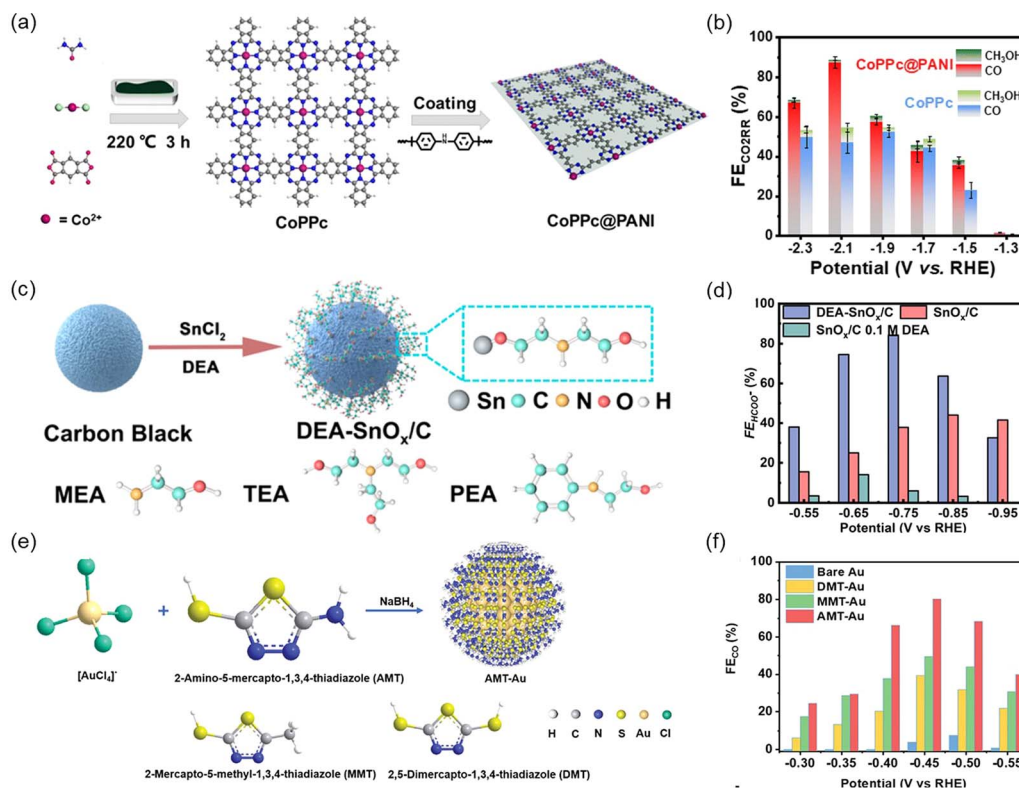


Fig. 5 (a) Schematic illustration of the CoPPc and CoPPc@PANI synthesis. (b) CO<sub>2</sub>RR performance between CoPPc@PANI and CoPPc in 5% O<sub>2</sub> and 95% CO<sub>2</sub> feed gas. (c) Fabrication of DEA-SnO<sub>x</sub>/C and structures of alkanolamines. (d) FE<sub>HCOO<sup>-</sup></sub> in 0.5 M KHCO<sub>3</sub> electrolyte under simulated flue gas. (e) Scheme of organic ligand modified Au NPs and the structure of organic ligands. (f) FE<sub>CO</sub> of AMT-Au with simulated flue gas saturated electrolyte in the cathode. (a and b) Reproduced with permission.<sup>47</sup> Copyright 2024, The Royal Society of Chemistry. (c and d) Reproduced with permission.<sup>48</sup> Copyright 2021, American Chemical Society. (e and f) Reproduced with permission.<sup>49</sup> Copyright 2024, Wiley-VCH.

(PANI) coating strategy to achieve highly efficient CO<sub>2</sub>RR performance using flue gas in acidic media (Fig. 5a and b). The unique imine groups on PANI can selectively adsorb CO<sub>2</sub> molecules and filter out O<sub>2</sub> molecules near the active Co-N<sub>4</sub> site in the conjugated cobalt polyphthalocyanine framework (CoPPc). Specifically, the acidic CO<sub>2</sub> molecules chemically interact with PANI, allowing faster CO<sub>2</sub> transfer during electrocatalysis compared with CoPPc. Therefore, CoPPc@PANI exhibits a high FE<sub>CO</sub> of up to 87.4% and an industry-level  $j_{CO}$  of  $-270 \text{ mA cm}^{-2}$  at  $-2.1 \text{ V vs. RHE}$  under 95% CO<sub>2</sub> + 5% O<sub>2</sub> feed gas in an acidic electrolyte. Similarly, Cheng *et al.*<sup>48</sup> grafted alkanolamines on a tin oxide surface and the surface grafted alkanolamines could selectively enrich CO<sub>2</sub>. Therefore, the ORR was inhibited and the reaction intermediates under an aerobic environment were stabilized. A diethanolamine (DEA) modified tin oxide catalyst (DEA-SnO<sub>x</sub>/C) (Fig. 5c and d) showed a maximum FE<sub>HCOO<sup>-</sup></sub> of 84.2% at  $-0.75 \text{ V vs. RHE}$  with  $a_j_{HCOO<sup>-}}</sup>$  of  $6.7 \text{ mA cm}^{-2}$  in 0.5 M KHCO<sub>3</sub> under simulated flue gas. Another redox-active molecule, 2-amino-5-mercapto-1,3,4-thiadiazole (AMT), was used to functionalize gold nanoparticles (Fig. 5e and f) for CO<sub>2</sub> enrichment by Kang's group recently.<sup>49</sup> The AMT ligand captured CO<sub>2</sub> with strong interaction in the reduced state, but could not capture O<sub>2</sub>. Therefore, the ORR was suppressed. The AMT-Au achieved a maximum FE<sub>CO</sub> of 80.2% at  $-0.45 \text{ V vs. RHE}$  in an H-type cell, and 66.0% at

a voltage of 2.7 V in a full cell, respectively, with simulated flue gas (15% CO<sub>2</sub>, 4% O<sub>2</sub>, balanced with N<sub>2</sub>). Recently, Sun *et al.*<sup>50</sup> achieved fast, selective CO electrocatalysis directly fed with simulated oxygen-containing flue gas (95% CO<sub>2</sub> + 5% O<sub>2</sub>) over the amine-confined Ag catalysts in a flow cell configuration. A FE<sub>CO</sub> of 84.2% with  $a_j_{CO}$  of  $333.7 \text{ mA cm}^{-2}$  was realized by using dimethylamine-modified Ag because amine modification could not only mediate CO<sub>2</sub> adsorption and \*COOH intermediate formation but also block the \*OOH intermediate pathway in the side reaction of oxygen reduction.

Modification of the pores with strong CO<sub>2</sub> affinity molecules, like amines, can effectively enrich CO<sub>2</sub> and eliminate the influence of O<sub>2</sub>. Li *et al.*<sup>51</sup> reported O<sub>2</sub>-tolerant catalytic electrodes for CO<sub>2</sub>RR by introducing guest aniline molecules into the pores of a PIM layer (Fig. 6). The chemical interaction between the acidic CO<sub>2</sub> molecule and the basic amino group of aniline could selectively capture CO<sub>2</sub>, which enhanced CO<sub>2</sub> separation and improved CO<sub>2</sub>RR selectivity. The PIM/aniline hybrid electrode achieved a FE<sub>CO</sub> of 71% with 10% O<sub>2</sub> in the CO<sub>2</sub> feed gas. Infrared spectroscopy measurements validly indicated that CO<sub>2</sub> was likely to be adsorbed by aniline *via* the chemical interaction between the acidic CO<sub>2</sub> and the basic amino group of aniline.

Based on these discussions, local CO<sub>2</sub> enrichment could be realized by introducing functional groups with alkali that can



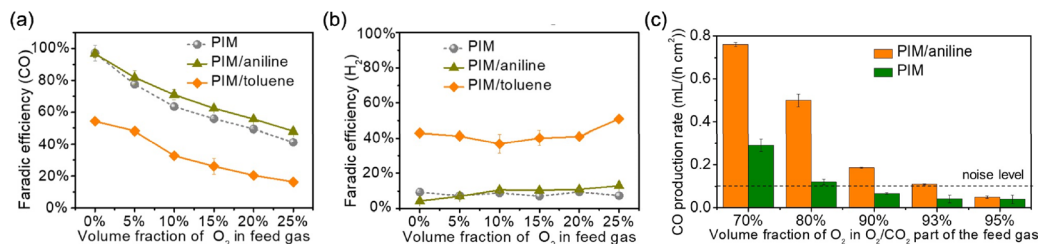


Fig. 6 (a) FE<sub>CO</sub> and (b) FE<sub>H<sub>2</sub></sub> for PIM, PIM/aniline and PIM/toluene cathodes operating with CO<sub>2</sub>/O<sub>2</sub> feed gas containing different O<sub>2</sub> percentages. (c) CO production rate vs. volume fraction of O<sub>2</sub> in the O<sub>2</sub>/CO<sub>2</sub> part of the feed gas with PIM or PIM/aniline as the CO<sub>2</sub>/O<sub>2</sub> selection layer. Reproduced with permission.<sup>51</sup> Copyright 2020, Wiley-VCH.

selectively adsorb CO<sub>2</sub> molecules, or filter the O<sub>2</sub> based on the molecular size, which can enhance the selective reduction of CO<sub>2</sub>RR in flue gas.

### 3.3 Direct selective CO<sub>2</sub>RR

Selective CO<sub>2</sub>RR in flue gas by designing electrocatalysts with specific electronic structures or optimized electrolyte compositions shall obtain highly selective O<sub>2</sub>-tolerant CO<sub>2</sub>RR.

**3.3.1 Special electronic structure construction.** Designing surface active sites with special electronic structures contributes to high CO<sub>2</sub>RR performances in the presence of O<sub>2</sub>. Recently, an O<sub>2</sub>-containing-species coordination strategy to boost CO<sub>2</sub>RR in the presence of O<sub>2</sub> was proposed by Cao *et al.*<sup>52</sup> The 2D conjugated COF catalyst (NiPc-Salen(Co)<sub>2</sub>-COF), which is composed of the Ni-phthalocyanine (NiPc) unit with Ni-N<sub>4</sub>-O and the salen(Co)<sub>2</sub> moiety with binuclear Co-N<sub>2</sub>O<sub>2</sub> sites, exhibited excellent high O<sub>2</sub>-tolerant CO<sub>2</sub>RR performance and achieved an outstanding FE<sub>CO</sub> of 97.2% at -1.0 V vs. RHE and

a high  $j_{CO}$  of 40.3 mA cm<sup>-2</sup> at -1.1 V vs. RHE in the presence of 0.5% O<sub>2</sub>. The combined ATR-IR and DFT calculations demonstrated that the \*OOH of ORR played a significant role in activating CO<sub>2</sub> by enhancing the charge polarization effect, which decreased the free energy of CO<sub>2</sub> activation and boosted the CO<sub>2</sub>RR.

**3.3.2 Electrolyte optimization.** Regulating the electrode/electrolyte interface by choosing an appropriate electrolyte is another key to enhance CO<sub>2</sub>RR directly. Acidic media for CO<sub>2</sub>RR achieve high carbon utilization efficiency, high overall energy utilization rate, and low carbonate formation, making them a compelling choice for industrial applications.<sup>53</sup> Recently, Wang *et al.* reported that acidic electrolytes have been found to significantly suppress ORR on Cu, enabling generation of multicarbon products from simulated flue gas. By using a Cu composite and carbon supported single-atom Ni as tandem electrocatalysts (Cu PTEE/Ni-N<sub>4</sub>), the Cu PTEE/Ni-N<sub>4</sub> achieved a multicarbon FE of 46.5% at 200 mA cm<sup>-2</sup> in acidic electrolyte,

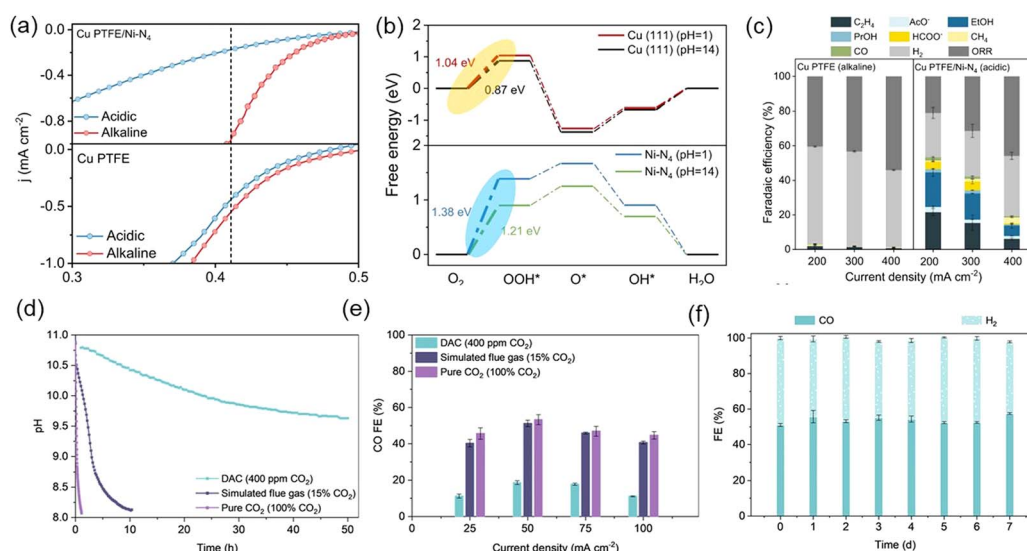


Fig. 7 (a) LSV curves in pure O<sub>2</sub> saturated acidic and alkaline electrolytes. (b) Free energy diagrams of ORR on Ni-N<sub>4</sub> and Cu PTFE at 1.23 V vs. SHE. (c) Product FE for Cu PTFE in 1 M KOH and Cu PTFE/Ni-N<sub>4</sub> in 0.05 M H<sub>2</sub>SO<sub>4</sub> + 1.5 M Cs<sub>2</sub>SO<sub>4</sub> under different current densities. (d) pH of a 2 M K-GLY with 0.1 M KH<sub>2</sub>PO<sub>4</sub> capture solution over time while capturing CO<sub>2</sub> from the atmosphere (400 ppm), CO<sub>2</sub> from a simulated flue gas (15%), and pure CO<sub>2</sub> (100%). (e) FE towards CO using the post-capture solutions of the capture processes in (d). (f) FE towards CO and H<sub>2</sub> of a newly assembled electrolyzer while continuously operating at 50 mA cm<sup>-2</sup> and exposing the same capture solution to 100 sccm of air for 7 days. (a-c) Reproduced with permission.<sup>54</sup> Copyright 2024, Springer. (d-f) Reproduced with permission.<sup>55</sup> Copyright 2024, Springer.



which was  $\sim 20$  times higher than that of bare Cu under alkaline conditions, enabling  $O_2$ -tolerant production of  $C_{2+}$  products in simulated flue gas.<sup>54</sup> DFT simulations suggested increases in the free energy change of the rate-determining step for the ORR on Cu and Ni- $N_4$  tandem sites in acidic media suppressing the ORR (Fig. 7a–c). Additives in the solution can interact with  $CO_2$  molecules, therefore promoting  $CO_2$  enrichment. When using amino acid salts (AAS) as additives, the amino groups can effectively adsorb  $CO_2$  and promote selective  $CO_2RR$ . Xiao *et al.*<sup>55</sup> developed a reactive capture strategy using AAS as additives to potassium glycinate capture solution. A maximum  $FE_{CO}$  of 19% for direct air capture experiments and a maximum  $FE_{CO}$  of 51% for the simulated flue gas experiment were achieved. The  $CO$  selectivities for the simulated flue gas and pure  $CO_2$  feed were comparable, demonstrating feasibility of reactive capture with dilute  $CO_2$  inputs when using AAS as the capture solutions (Fig. 7d–f).

## 4 Summary and outlook

### 4.1 Summary

Direct  $CO_2$  electroreduction using flue gas (typically containing 3–20%  $CO_2$  and 3–5%  $O_2$ ) offers a promising route to decarbonize industrial emissions by bypassing energy-intensive capture and purification steps. While strategies like  $O_2$  passivation, selective  $CO_2$  adsorption, and direct  $CO_2$ -selective catalysis have advanced aerobic  $CO_2RR$ , current research often lacks synthesis of cross-cutting design principles and industrial-relevant performance benchmarks. To bridge this gap, we emphasize two critical directions for practical implementation:

1. Breaking laboratory limits requires targeting the following thresholds. The activity, selectivity and stability parameters of  $CO_2RR$  are basic prerequisites for its commercial application. Industrial electrolyzers demand over  $200\text{ mA cm}^{-2}$  for current density. In addition, the FE should be higher than 85% with at least 200-hour durability under 5%  $O_2$  coexistence.

2. Multi-impurity tolerance: beyond oxygen challenges. Current research only focuses on a single impurity gas, but the actual situation will be more complex. Different impurity gases, such as  $SO_2$  and  $NO_2$ , have varying effects on the system. In addition, smoke may also carry some solid small particles (such as  $SiO_2$ ,  $Fe_2O_3$ ,  $Al_2O_3$ ,  $CaO$ ,  $MgO$ ), which may cause side reactions and even lead to the coverage of catalytically active sites. Integrated gas–solid separation could remove most particulates by pre-filtration and preventing active-site coverage.

### 4.2 Outlook

Despite the effort that has been made in  $CO_2RR$  under an aerobic environment, there are still many problems and challenges that need to be addressed for directly feeding with flue gas and applying the industrial-scale  $CO_2RR$ .

(i) The relationships among the electrocatalyst, electrolyte, and electrode should be studied. Typically, a more reliable reaction mechanism for effectively regulating the reaction shall be established. Current research on selective  $CO_2RR$  remains in its early stages, with only a limited number of studies providing

detailed explorations of the underlying mechanisms. Advanced *in situ* characterization techniques and computational methods are also necessary and efficient to obtain more useful information to investigate the structural and physicochemical properties.

(ii) The efficiency of  $O_2$ -tolerant  $CO_2RR$  is important to realize industrial-scale commercial application. The undesirable energy conversion efficiency caused by a high overpotential and low FE limits the near-future practical applications of  $CO_2RR$ . Achieving industrial-scale applicability for  $CO_2/O_2$  selective reduction is hindered by the challenge of achieving both high FE and  $j$ .<sup>56,57</sup> These performance benchmarks are essential for large-scale applications, but still difficult to satisfy, limiting the potential of directly feeding with flue gas. Additionally, long term stability tests (time > 200 h,  $j$  >  $200\text{ mA cm}^{-2}$ ) shall be studied.

(iii) More attention should be paid to study the selective  $CO_2RR$  using low concentration  $CO_2$  condition systems. For example, a typical exhaust gas generated by the combustion of fossil fuels has an  $O_2/CO_2$  ratio of 20%, the coal-fired gas is always composed of 5%  $O_2$ , 15%  $CO_2$ , 77%  $N_2$  and impurities, and the air contains 20%  $O_2$  and 400 ppm  $CO_2$ . Selective  $O_2/CO_2$  reduction using air directly as feed gas can save the cost of  $CO_2$  separation, which is attractive in the future. Therefore, developing electrocatalysts or reactors that can selectively separate and reduce  $CO_2$  directly from  $CO_2$ - $O_2$  mixed gas with various  $CO_2$  contents is crucial.<sup>58–60</sup>

(iv) The economic viability of oxygen-tolerant electrocatalyst manufacturing at an industrial scale faces significant challenges, primarily dictated by process methodology selection. Critical cost drivers—including raw material inputs, post-processing requirements, and waste management—must be holistically optimized, where the choice of synthesis techniques fundamentally determines operational efficiency and environmental impact. For example, electrosynthesis may emerge as a strategic alternative to conventional routes from some aspects. As a direct electrochemical redox platform, it utilizes electricity (ideally sourced from wind/solar) rather than thermal activation, achieving >40% reduction in carbon emissions *versus* thermochemical pathways. Contamination mitigation *via* electrode engineering inherently prevents metal leaching. This eliminates downstream further purification demand.

(v) Industrial-scale  $CO_2$  electroreduction faces system-level challenges beyond catalyst design, necessitating integrated engineering solutions for upstream gas conditioning and downstream product separation. On the upstream side, flue gas containing 3–5%  $O_2$  and particulate impurities competes with  $CO_2$  for catalytically active sites. A multi-stage purification system such as ceramic microfiltration membranes achieves >99% removal efficiency for particulates to prevent catalyst abrasion. Applying a pressure of 3–5 bar elevates local  $CO_2$  concentration >20%, thereby facilitating current densities that surpass the critical industrial benchmark of  $200\text{ mA cm}^{-2}$ . On the downstream side, liquid fuels (*e.g.*, formic acid, ethanol) present significant technoeconomic hurdles, particularly due to their dilute nature (<1 mol  $L^{-1}$ ) in electrolyte-laden catholyte streams (containing  $K^+/Na^+$  species). These challenges manifest



in three primary dimensions: (1) mandatory ion removal through electrodialysis or reverse osmosis processes, which elevate operational expenditures by 30–50%; (2) energy-intensive multi-effect distillation requirements for product concentration, adding 40–60% to the energy balance; and (3) substantially inflated logistics costs combined with the need for additional purification steps to achieve industry-mandated specifications (>90% purity).

Overall, developing electrocatalysts with high CO<sub>2</sub>RR ability in flue gas shall shed new light on the development of O<sub>2</sub>-tolerant electrocatalysis systems that would facilitate efficient CO<sub>2</sub>RR with high activity and selectivity in the presence of O<sub>2</sub>. We believe that the CO<sub>2</sub>/O<sub>2</sub> selective reduction shall offer new approaches to further improve efficiency and provide novel insights for directly operating CO<sub>2</sub>RR under O<sub>2</sub>-containing CO<sub>2</sub> feed gas.

## Data availability

The sources of the data discussed are all references cited within the article.

## Conflicts of interest

There are no conflicts to declare.

## Acknowledgements

This work was supported by the Science and Technology Development Fund from Macau SAR (FDCT) (0111/2022/A2, 0050/2023RIB2, 0023/2023/AFJ, 0002/2024/TFP, and 0087/2024/AFJ) and Multi-Year Research Grants (MYRGGRG2023-00010-IAPME and MYRG-GRG2024-00038-IAPME) from Research & Development Office at the University of Macau.

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