



Skipping the middleman: Direct electroreduction of amine-captured CO₂ via zwitterionic carbamate interfaces

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Excessive CO₂ emissions from fossil fuel combustion and industrial activities have intensified global warming and accelerated climate-related disruptions, making carbon capture and utilization an increasingly important strategy for achieving carbon neutrality^[1-3]. Among existing carbon capture technologies, amine-based absorption remains the industrial benchmark because of its high capture efficiency and technological maturity (Figure 1a). However, conventional processes require energy-intensive thermal regeneration to release captured CO₂, with solvent regeneration accounting for the majority of the energy consumption^[5-7]. Integrating CO₂ capture directly with electrochemical conversion (also known as electrochemical reactive capture) has therefore emerged as an attractive alternative^[5,8-12], as it bypasses thermal regeneration and enables captured carbon species to be directly upgraded into value-added products. Despite this promise, the intrinsic complexity of amine-CO₂ chemistry remains a major obstacle. Multiple dynamic species, including carbamates, bicarbonates, protonated amines, and dissolved CO₂, coexist in capture solutions, making it difficult to identify the true electroactive species and understand how capture chemistry influences the interfacial electrochemical process^[5,13].

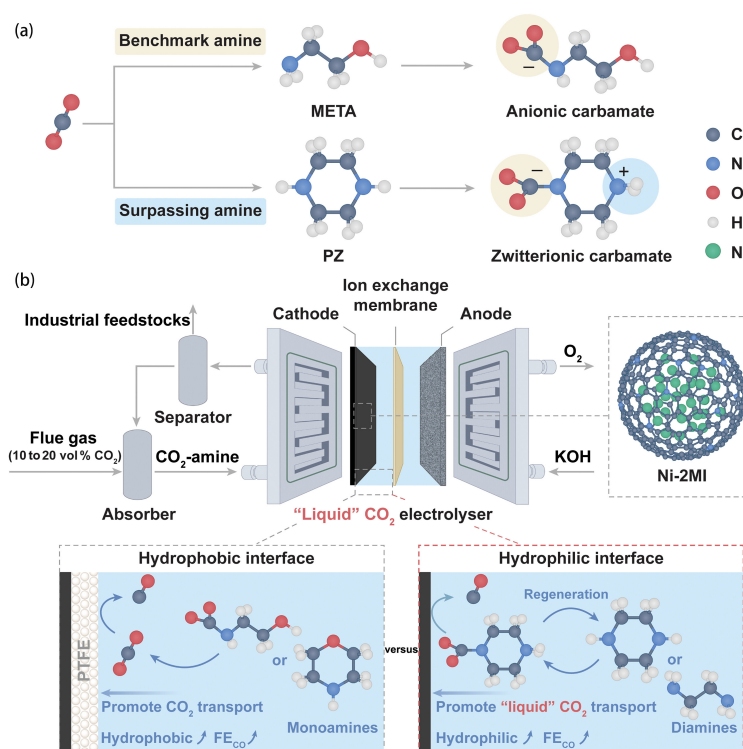


Figure 1. Capture chemistry and interfacial design for ICCE. (a) Comparison of carbamate formation pathways between benchmark (META) and surpassing (PZ) amines; (b) Schematic illustration of ICCE and electrode-electrolyte interfacial regulation of electroreduction. Reproduced from reference^[4]. CC BY 4.0. ICCE: integrated CO₂ capture and electrolysis; META: monoethanolamine; PZ: piperazine.



Prior to this work, research in this field had primarily focused on improving the electrochemical conversion step while treating the capture process as largely fixed^[13–15]. Considerable efforts were devoted to enhancing the performance of conventional amine- or amino-acid-based reactive-capture systems through catalyst engineering^[4], electrolyte optimization^[14,16,17], and interfacial modifiers such as alkali metal cations, surfactants and advanced electrocatalysts. Although these approaches improved catalytic activity to varying degrees, they shared a common assumption that the capture solvent merely served as a passive CO₂ reservoir. In 2025, Ma and co-workers proposed, for the first time, a tandem amine scrubbing and CO₂ electrolysis process that directly converts captured CO₂ into chemicals, thereby bypassing the energy-intensive desorption and compression stages. In their study, piperazine was employed as the absorbent, allowing for the direct electrochemical reduction of the resulting carbamate adducts over a nickel single-atom catalyst^[6]. However, the molecular structure of the amine itself and its influence on interfacial chemistry remained largely unexplored. As illustrated in Figure 1a, different amines possess fundamentally distinct CO₂-binding chemistries, suggesting that the capture solvent should not simply be regarded as a carbon carrier, but rather as a potential regulator of the electrochemical reaction environment^[4].

In a recent study published in *Science Advances*, Ma and co-workers^[4] challenge this conventional design philosophy by demonstrating that the molecular chemistry established during CO₂ capture can be deliberately exploited to engineer the catalytic interface. Through a systematic comparison of six representative amines, they identify piperazine as uniquely capable of stabilizing the protonated carbamate species that spontaneously assembles into a zwitterionic interfacial environment^[4,6]. Unlike conventional membrane electrode assembly (MEA) systems, where carbamates continuously undergo hydrolysis and exist in highly dynamic equilibria, the relatively stable protonated carbamates formed by piperazine provide a structurally organized interface that promotes efficient electrochemical conversion. Benefiting from this unique interfacial environment, the optimized piperazine-Ni nanoparticle system achieves a CO Faradaic efficiency of approximately 60%, while delivering stable CO Faradaic efficiencies of 30–45% and energy efficiencies of ca. 15–25% for over 150 hours in a 9 cm² zero-gap membrane electrode assembly electrolyzer.

The deeper value of this work lies in how it resolves the long-standing ambiguity of “what is actually being reduced” in amine-captured CO₂ electrolysis. Instead of inferring the reactive species only from product selectivity, Ma and co-workers build a coherent mechanistic chain across speciation, transport and surface reaction. The strong dependence of CO partial current on carbamate concentration, together with its much weaker correlation with CO₂ partial pressure, indicates that the reaction is not governed simply by released molecular CO₂. Through comparative experiments integrated with *in situ* spectroscopic investigations and computational simulations, the authors highlight the key role of protonated carbamates in the as-constructed integrated capture-conversion platform, which acts not only as CO₂ reservoirs but also as active participants in interfacial proton transfer and microenvironment regulation. Typically, piperazine exhibits exceptional ionic CO₂ loading capacity, which is beneficial for overcoming mass-transfer limitations, thereby providing high local species availability, a key factor for boosting electrochemical reaction rates (Figure 1b).

Overall, the present work highlights the key role of molecular structures generated during CO₂ capture in creating favorable interfacial environments for electrocatalysis. In this sense, capture chemistry is elevated from an upstream separation process to an active tool for electrochemical interface engineering. Such a perspective is likely to inspire future research that simultaneously considers capture solvent design, catalyst development and interfacial microenvironment regulation as a unified molecular system, rather than independent optimization targets. Beyond piperazine, this framework may also stimulate the exploration of new classes of capture molecules capable of simultaneously maximizing carbon capture efficiency and electrochemical reactivity, thereby accelerating the development of practical integrated carbon capture and conversion technologies. However, several challenges remain before this technology can be translated into practical applications. Current systems generally exhibit limited current densities insufficient product selectivity toward multi-carbon products. The complex interactions between capture solvents, catalysts, electrolytes, and membranes also hinder system optimisation. Future research should focus on developing integrated catalyst-capture media systems, advanced operando characterisation, and mechanistic modelling to guide materials innovation. Combining these advances with scalable reactor engineering, renewable electricity integration, and comprehensive techno-economic and life-cycle assessments will accelerate the commercial deployment of integrated CO₂ capture-electrolysis technologies.

Declarations

Authors contribution

Pan Q: Data analysis, investigation, validation, writing-original draft.

Sun Y: Conceptualization, project administration, data analysis, investigation, supervision, validation, writing-original draft, writing-review & editing.

Conflicts of interest

Ying Sun is an Editorial Board Member of *Smart Materials and Devices*. The other authors declare no conflicts of interest.

Ethical approval

Not applicable.

Consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data and materials

Not applicable.

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