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Advancing towards intelligent and scalable atomic catalysts: Insights and prospects

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Electrocatalytic CO_2 reduction (CO_2RR) toward multicarbon (C_{2+}) products represents a key pathway for closing the carbon cycle and producing renewable fuels and chemicals. However, the simultaneous achievement of high selectivity and high current density remains challenging.

Developing efficient and sustainable catalysts is thus central to addressing global energy and environmental challenges. Conventional heterogeneous catalysts suffer from limited atomic efficiency and ill-defined active sites. In contrast, single-atom catalysts (SACs) offer maximized atomic utilization, well-defined active sites, and unique electronic structures [1]. Nevertheless, the isolated single sites often struggle to effectively promote C–C bond formation, which typically requires the cooperative interaction between adjacent intermediates or multi-center active sites. Dual-atom catalysts (DACs), featuring two closely spaced metal atoms with synergistic geometric and electronic effects, have gained attention for facilitating C–C coupling and improving selectivity toward C_{2+} products [2].

Yet, challenges remain in achieving precise active-site engineering, enhancing reaction selectivity in multi-electron transfer processes, and bridging the gap between fundamental studies and industrial implementation. Among these challenges, atomic-level structural regulation has emerged as a pivotal strategy for addressing these limitations.

At the same time, computational chemistry, exemplified by density functional theory (DFT) and *ab initio* molecular dynamics (AIMD), has become an indispensable tool for probing reaction mechanisms and correlating atomic configurations with catalytic performance [3,4]. Machine learning (ML) accelerates catalyst discovery by rapidly exploring vast chemical spaces, identifying key descriptors and predicting the most promising SAC candidates. Compared with conventional trial-and-error experimental or computational screening strategies, the integration of ML with DFT dramatically reduces computational cost while improving predictive accuracy, enabling efficient exploration and identification of promising catalyst systems. When experimental characterization, theoretical modeling, and data-driven approaches are integrated within a closed-loop framework, the intelligent design and accelerated optimization of atomic catalysts become attainable realities, as shown in Figure 1.

Atomic-level structural regulation. Precise control of the atomic configuration of catalytic active sites is

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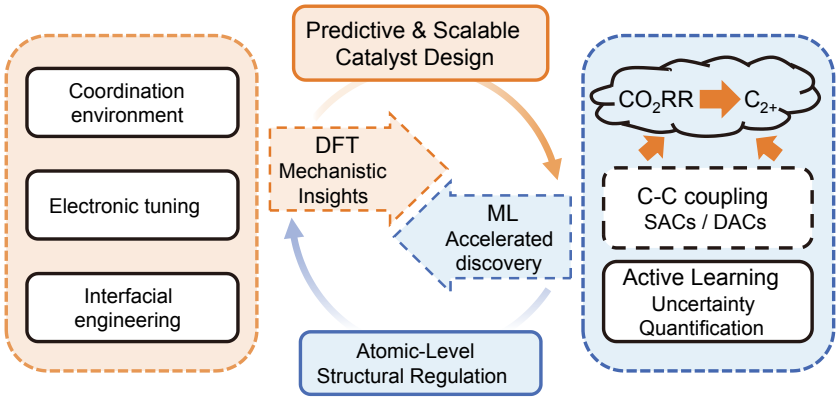


Figure 1 Hierarchical structural regulation and intelligent design framework for CO_2 electroreduction catalysts.

essential for elucidating reaction mechanisms and enabling the rational design of high-performance catalysts. For CO_2RR toward C_{2+} products, the atomic-level structural regulation of SACs can be viewed as a dual-lever strategy based on coordination environment and electronic structure, further amplified by reaction microenvironment and interfacial engineering. At the first coordination shell, fine-tuning of coordination type, coordination number, and geometric configuration effectively adjusts the d-band center and local charge distribution. This enables a balanced regulation between *COOH formation and *CO desorption energetics, thereby suppressing the hydrogen evolution reaction (HER) and enhancing *CO coverage. The second coordination shell and the extended carbon matrix fine-tune the oxidation state and local electric field through heteroatom doping, lattice strain, pore confinement, and dielectric or polar effects, maintaining the structural stability of the primary bonding framework.

Meanwhile, axial ligand tuning and electric double-layer engineering modulate proton-coupled electron transfer (PCET) pathways and intermediate stabilization. This cooperative regulation lowers the free-energy barriers of *CO-*CO and *CO-*CHO coupling under realistic potentials and explicit-solvent conditions, thereby facilitating C_{2+} product formation. Furthermore, surface defects, steps, strain, and heterointerfaces with oxides or hydroxides can create proton-relay channels and local pH microenvironments, mitigating *OH poisoning and stabilizing key transition states. These interfacial characteristics not only suppress undesired side reactions but also prolong the lifetime of key intermediates such as *CO, creating favorable conditions for subsequent C–C coupling. Beyond static coordination and microenvironmental regulations, recent studies have shown that nominally isolated SACs may undergo dynamic structural reorganization under operating conditions (Figure S1) [5].

Overall, coordinated regulation of local structure, electronic state, and interfacial environment promotes the evolution of SACs from isolated active sites into cooperative catalytic systems, providing guidance for predictive atomic-catalyst design.

Machine learning. Recent advances have made ML a powerful tool for catalyst discovery and design. To further enhance the physical interpretability and reliability of ML models, recent efforts have increasingly focused on incorporating attribution analysis techniques. For example, SHAP analysis has been applied to CO_2 electroreduction systems to identify dominant descriptors governing product selectivity [6]. These interpretable frameworks reveal feature importance and bridge data-driven prediction with electrocatalytic mechanisms and rational catalyst design [7]. Algorithms such as ensemble learning, kernel methods, and

graph neural networks (GNNs) effectively capture complex features derived from theoretical calculations, enabling robust and interpretable catalyst models (Figure S2) [8].

At the mechanistic level, DFT remains the cornerstone for elucidating reaction pathways and structure-activity relationships. It provides an atomic-scale magnifier for identifying key intermediates, rate-determining steps, and free-energy landscapes in C–C coupling and deep hydrogenation processes. A broadly accepted consensus has emerged that CO serves as the pivotal intermediate in C_{2+} product formation. CO is widely recognized as the key intermediate in C_{2+} product formation, undergoing either direct dimerization or hydrogenation-assisted coupling pathways prior to subsequent PCET processes that determine C_2 product selectivity. Standard simulations based on the computational hydrogen electrode (CHE) framework, combined with solvent models and transition-state calculations, enable quantitative evaluation of interfacial PCET kinetics and rate-limiting steps under realistic electrochemical conditions.

Beyond static energetics, dynamic simulations are essential for capturing realistic catalyst behavior under operating conditions (Figure S3) [9]. Coupling DFT with ML through microkinetic modeling and active-learning strategies establishes a scalable closed-loop workflow, where physics-informed surrogate models iteratively refine catalyst predictions via Bayesian optimization and operando validation. This closed-loop paradigm enables precise regulation of product distributions and enhanced selectivity toward C_{2+} species in CO_2 electroreduction.

To ensure the practical implementation of active learning in closed-loop catalyst design, establishing clear stopping criteria for iterative model updating is essential. One possible strategy is based on the convergence of model uncertainty, where the predicted uncertainty across the candidate space falls below a predefined threshold, indicating sufficient exploration of the chemical space.

Alternatively, stopping criteria can be defined by performance-based targets, where the predicted activity or selectivity of candidate catalysts reaches preset objectives, suggesting that further iterations may yield diminishing returns. In addition, model-centric criteria, such as the saturation of validation performance reflected by negligible improvement in metrics such as MAE or RMSE over successive iterations, can also be employed to monitor convergence. In practice, a combination of these criteria may provide a more robust and standardized stopping strategy, balancing model reliability, exploration efficiency, and practical relevance [10].

Outlook. Despite significant advances in elucidating the structure-activity relationships of SACs/DACs, translating these atomic-level insights into scalable, robust, and industrially relevant catalytic systems remains a formidable challenge. Future research should focus on several strategic directions toward practical implementation.

(1) Intelligent closed-loop design. Integrating high-throughput computation, *in-situ* and *operando* characterization, and ML within a feedback loop enables autonomous catalyst optimization. Future efforts should emphasize multi-objective optimization considering activity, C_{2+} selectivity, stability, and cost. Reinforcement learning and generative models may further enable automated catalyst design and optimization.

(2) Scalable synthesis and engineering integration. Atomically dispersed catalysts are often limited to small-scale synthesis with structural nonuniformity. Developing scalable synthesis methods is vital for industrial applications. Stabilizing metal-support interfaces under realistic potentials and prolonged operation is also crucial to maintain atomic dispersion and structural integrity.

(3) Standardization and open data. Establishing unified evaluation metrics and open databases is essential

for reliable benchmarking. Inconsistencies in electrolysis conditions, current normalization, and degradation criteria currently impede objective benchmarking. Building a community-wide framework that integrates experimental results, theoretical data, and ML models will accelerate knowledge accumulation and data-driven catalyst design.

In summary, atomic catalysis is evolving from empirical exploration toward intelligent, scalable, and data-centric design. The synergy between computation, characterization, and ML is expected to accelerate the practical deployment of atomic catalysts for CO_2 utilization and sustainable energy conversion.

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Author contributions

Q.Y. supervised the project and organized the collaboration. Y.Z. wrote the manuscript. Q.Y. and Y.Z. revised the manuscript. All authors contributed to the manuscript.

Conflict of interest

The authors declare no conflict of interest.

Supplementary information

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