Understanding the Dynamic Behavior of Oxide-Derived Copper in CO_2 Reduction with Machine Learning Based Large-Scale Simulation

Zan Lian[1], Federico Dattila[2] and Núria López[1]

[1]Institute of Chemical Research of Catalonia (ICIQ-CERCA), The Barcelona Institute of Science and Technology (BIST), Av. Països Catalans 16, 43007 Tarragona, Spain

[2]Department of Applied Science and Technology (DISAT), Politecnico di Torino, Corso Duca degli Abruzzi 24, 10129, Turin, Italy

zlian@iciq.es

Figures

Figure 1. Schematic of dynamic behavior of oxide-derived copper in electrochemical CO_2 reduction reaction.

Abstract

Closing the carbon cycle by converting CO_2 to long-chain hydrocarbons using renewable electricity is a beneficial process from both an environmental and economic perspective. Oxide-derived Cu (OD-Cu) catalysts have shown the potential to produce multicarbon species with low overpotentials[1].These OD-Cu catalysts are usually formed when Cu_2O loses oxygen under reduction potentials. It is believed that its unique reconstructed surface is responsible for its superior performance[2]. However, these materials extensively rearrange under reaction conditions, thus the nature of the active site remains controversial[3]. Here, we studied the reduction process of OD-Cu via large-scale molecular dynamics with an accurate neural network potential[4,5]. The oxygen concentration in the most stable OD-Cu increases with the increase of the pH, potential, or specific surface area. In long electrochemical experiments, the catalyst would be fully reduced to Cu, but it takes a considerable amount of time to remove all the trapped oxygen, although the highly reconstructed Cu surface provides various sites to adsorb oxygen under relatively stronger reduction potentials, the surface oxygen atoms are not stable under common experimental conditions. This work provides insight into the evolution of OD-Cu catalysts and residual oxygen during reaction and a deep understanding of the nature of active sites.

References

[1] Yang, Y. et al., Nature 614, 262–269 (2023)

[2] Dattila, F., García-Muelas, R. & López, N., ACS Energy Lett. 5, 3176–3184 (2020)

[3] Arán-Ais, R. M., Scholten, F., Kunze, S., Rizo, R. & Roldan Cuenya, B., Nat. Energy 5, 317–325 (2020)

[4] Behler, J., J. Chem. Phys. 134, 074106 (2011).

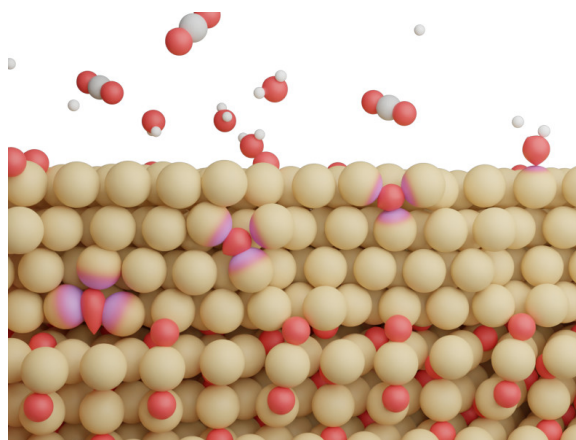[5] Lian Z, Dattila F, López N., Nat. Catal. Accepted (2024)