Selectivity Roadmap for Electrochemical CO₂ Reduction on Copper-based Alloy Catalysts

Xing Zhi, Yan Jiao*, Yao Zheng, Anthony Vasileff, Shizhang Qiao*
Centre for Materials in Energy and Catalysis, School of Chemical Engineering and Advanced Materials, The University of Adelaide, SA 5005, Australia

Introduction

- Electrochemical CO₂ reduction is a promising approach for converting CO₂ to valuable chemicals.¹
- Developing highly selective electrocatalysts for targeted products in CO₂ reduction reaction (CRR) remains a major challenge.²
- The search for effective selectivity descriptors by considering the intrinsic electrocatalyst properties are highly required.

![Figure 1. Reaction pathways to various products via key intermediates. Dashed arrows indicate multiple proton/electron transfer steps.](image)

Computational Methods

- Density functional theory (DFT) calculations
- Perdew-Burke-Ernzerhof (PBE) functional for electron exchange-correlation
- Projector-augmented wave (PAW) method
- Computational hydrogen electrode (CHE) model ³

Selectivity Roadmap

- Theoretical elucidation for CRR selectivity trends
- Electronic properties of CRR catalysts
- Rational design strategies for selective CRR catalysts

![Figure 3. Scheme of selectivity preference to various products based on different M-H and M-O affinities of the M@Cu catalysts](image)

Selectivity Descriptors

- Cu-based alloy models (M@Cu)
- M-H and M-O affinity in M@Cu catalysts
- Product Distribution

![Figure 2. Product grouping of the M@Cu catalysts according to their M-H and M-O affinities. The inset shows the investigated metal elements on the periodic table.](image)

Conclusion

- A novel descriptor-based approach to predict CRR selectivity of catalysts is developed.
- A series of Cu-based alloy models (M@Cu) are employed to explore their CRR selectivity by extensive thermodynamic analysis.
- The M-H and M-O affinity in M@Cu catalysts are found to be effective descriptors in determining CRR selectivity.
- The product distribution matches well with the reported CRR selectivity trends in Cu-based bimetallic catalysts

References


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