Selectivity Roadmap for Electrochemical CO_2 Reduction on Copper-based Alloy Catalysts

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Introduction

- Electrochemical CO_2 reduction is a promising approach for converting CO_2 to valuable chemicals.[1]
- Developing highly selective electrocatalysts for targeted products in CO_2 reduction reaction (CRR) remains a major challenge.[2]
- The search for effective selectivity descriptors by considering the intrinsic electrocatalyst properties are highly required.

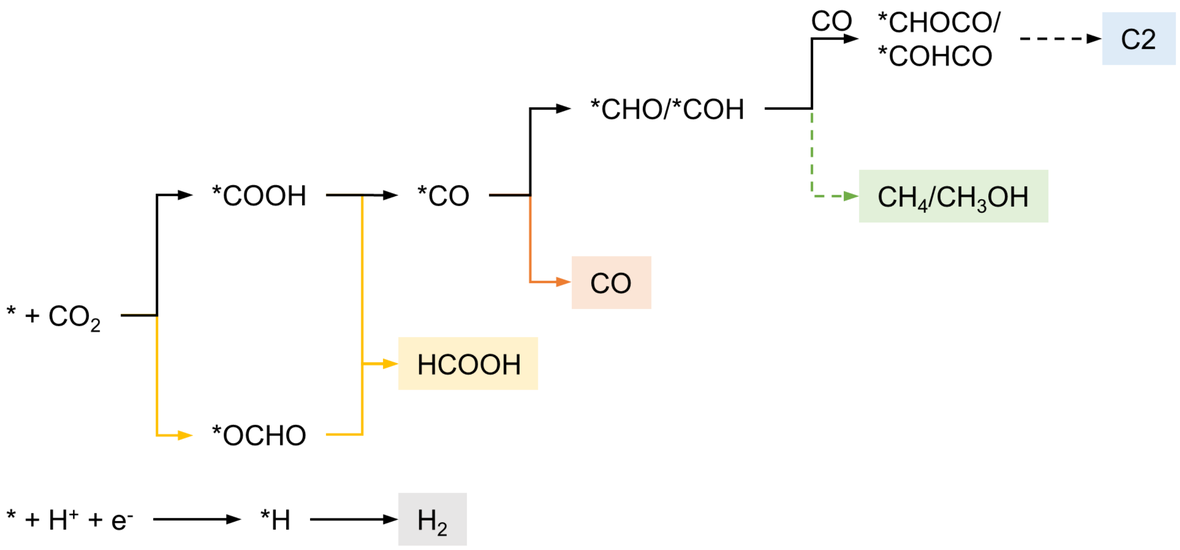

Figure 1. Reaction pathways to various products via key intermediates. Dashed arrows indicate multiple proton/electron transfer steps.

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 Descriptors

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

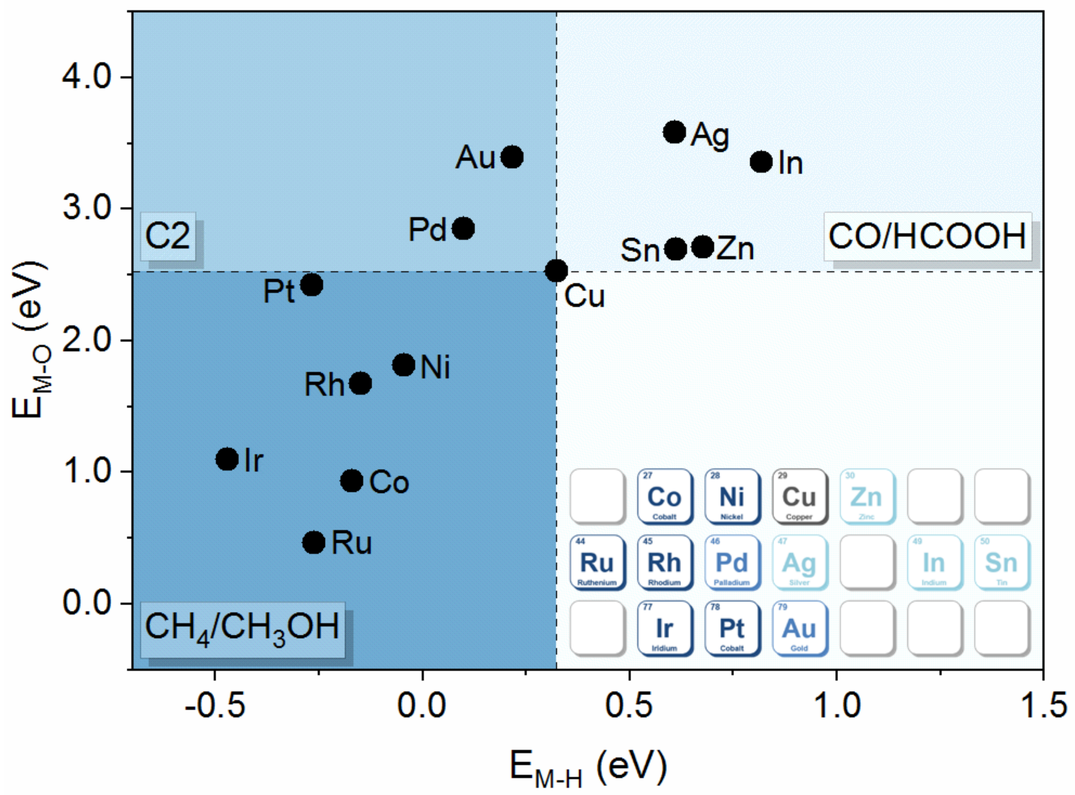

Figure 2. Product grouping of the M@Cu catalysts according to their M-H and M-O affinities. The inset shows the investigated metal elements on the periodic table.

Selectivity Roadmap

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

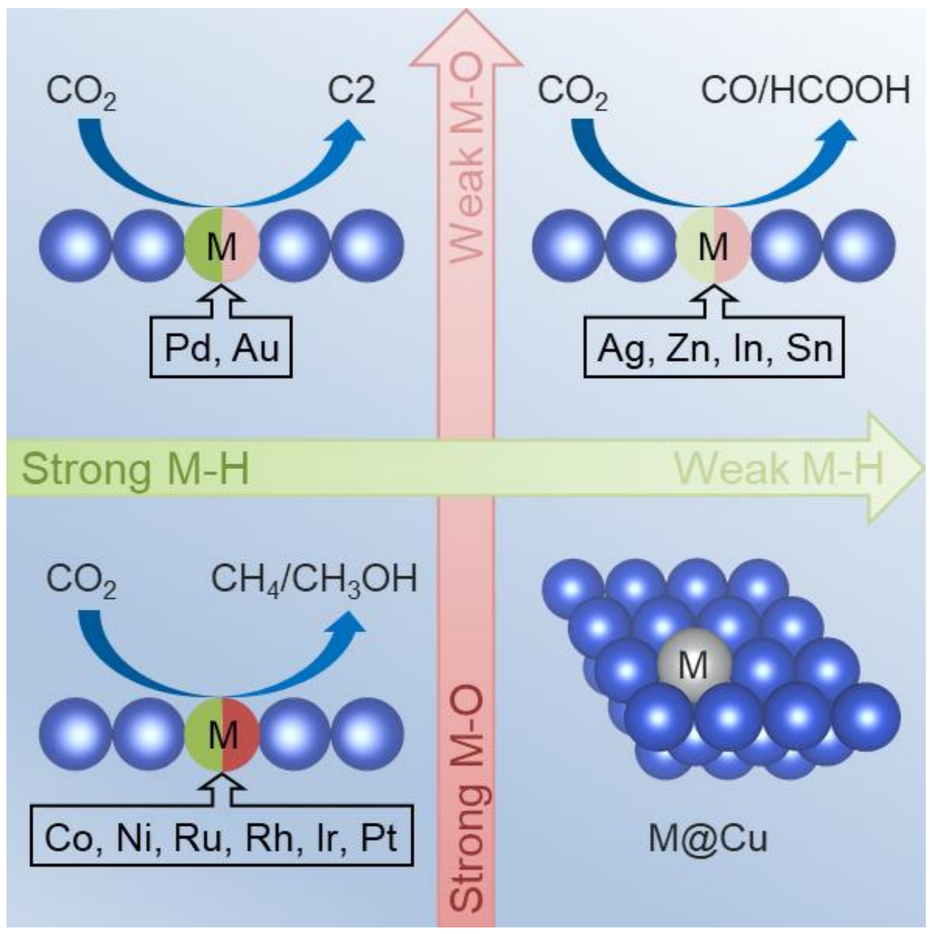

Figure 3. Scheme of selectivity preference to various products based on different M-H and M-O affinities of the M@Cu catalysts

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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3. J. K. Nørskov, et al., *J. Phys. Chem. B* **2004**, *108*, 17886.

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