Understanding Catalysis Through the Construction and Study of an Artificial Enzyme

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Nature’s Exemplary Enzymes as Inspiration
Nature has championed clean energy conversion reactions with high degree of selectivity and activity all while operating in water and utilizing earth abundant metals

Carbon Monoxide Dehydrogenase (CODH)

$\text{CO}_2 + 2\text{H}^+ + \text{HCOO}^- \rightarrow \text{CO} + \text{H}_2\text{O}$

$\text{CO}_2 + \text{H}^+ \rightarrow \text{HC} = \text{O}$

$\text{CO}_2 + \text{H}_2 \rightarrow \text{CO} + \text{H}_2\text{O}$

$\text{CO}_2 + \text{H}_2 \rightarrow \text{CO} + \text{H}_2\text{O}$

Nature’s enzymes are complex!

Can we identify the building blocks needed to achieve similar reactivity? What is the role of each complex element?

Building an Artificial Enzyme
Taking a top-down approach to build an artificial enzyme by utilizing a well-structured protein scaffold and a molecular complex

Structured Protein Scaffold
- Well characterized
- High yield expression
- Stable to mutagenesis
- Binding of exogenous molecules

Bio-inspired Molecular Complex
- Well studied and characterized
- Inactive in water

Artificial Enzyme
- Controlling outer-coordination sphere
- Rational design
- Tune catalysis?

CO$_2$ Hydrogenation by an Artificial Enzyme

$\text{CO}_2$ Hydrogenation by an Artificial Enzyme

Can we incorporate design principles conserved in native enzymes to modulate catalysis?

Artificial enzyme demonstrate catalytic competence for hydrogenation of CO$_2$ to formate, but what is the role of the scaffold?

Incorporation of positively charged residues near the Rh center results in increased activity while global negative charges have little effect on catalytic activity. Can we further define the mechanism?

Can we apply these design principles across different scaffolds?

Conclusions and Future Directions

Acknowledgements and References

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