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TEXAS
The University of Texas at Austin

Computational Design and Engineering of Biocatalysts for CO_2 Reduction

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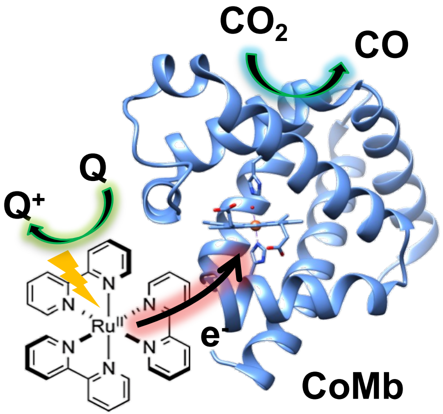
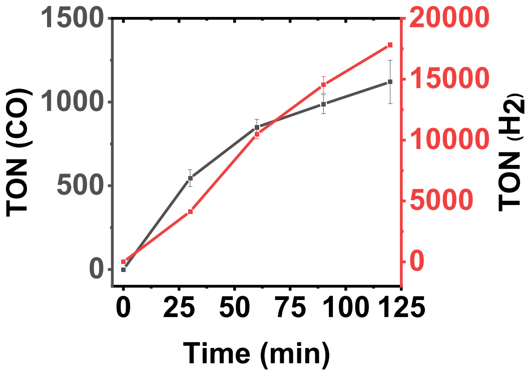
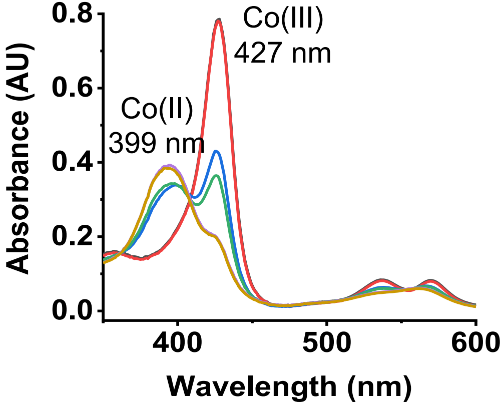
Introduction

❖ Designing biocatalysts for converting CO_2 into valuable products offers a green way to minimize greenhouse gas emissions and generate a sustainable net-zero carbon economy.







- Cobalt-substituted myoglobin (CoMb) can reduce CO_2 to CO under photocatalytic conditions.
- The catalytic activity can be enhanced by tuning the secondary coordination sphere (SCS).

Aims

1. Determine the oxidation states of CoMb system capable of CO_2 binding.
2. Calculate the energy landscapes associated with CO_2 activation and reduction.
3. Identify favorable SCS interactions for CO_2 binding and activation.

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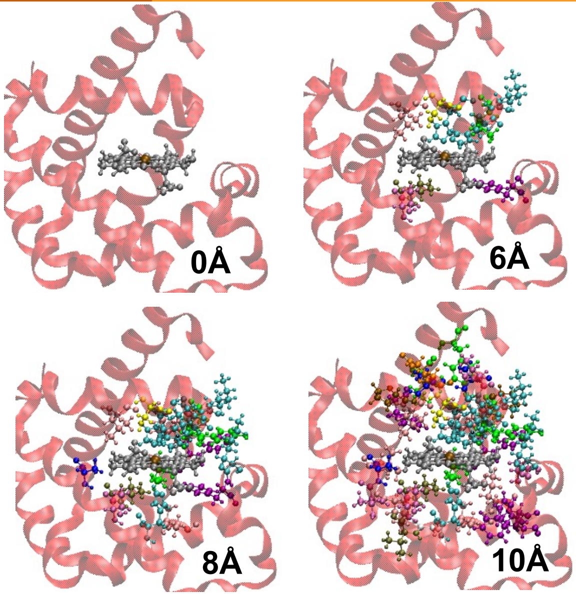
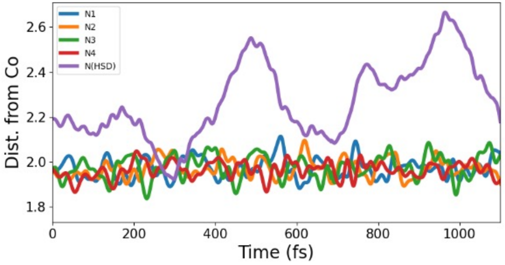
Methodology and Benchmarking

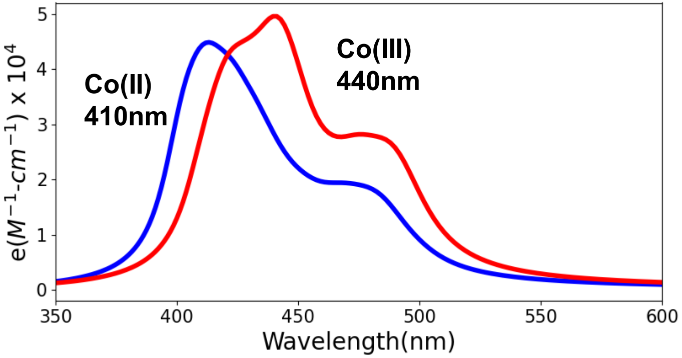
- QM/MM method was employed to deal with the large system sizes.
- Deviations from full scale DFT calculations can be minimized by careful selection of the QM region.
- Electrostatic potential at the metal center was used for QM region selection.
- Additionally, structural and opto-electronic properties were computed to benchmark with experiments.



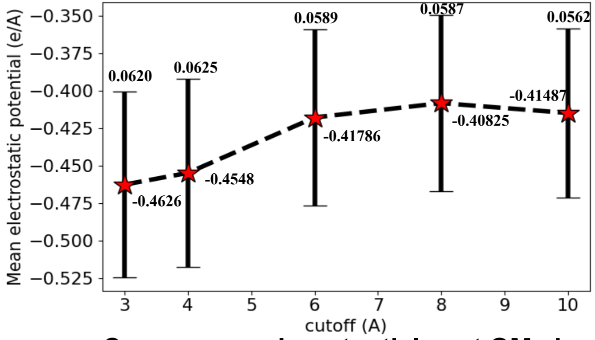


Averages of various bond lengths

Expt. (Å)	Sim. (Å)
1.98	1.9812 ± 0.052
1.94	1.9774 ± 0.0463
2.02	1.97945 ± 0.0495
1.95	1.9646 ± 0.0393
2.18	2.2032 ± 0.244

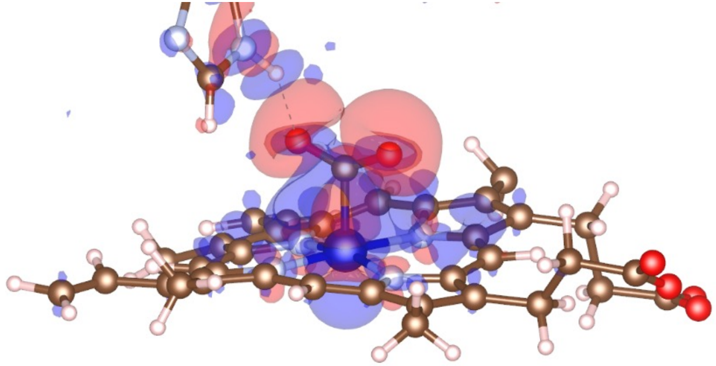


Absorption spectrum plots showing a red shift of 30nm between Co(II) and Co(III).



Convergence in potential w.r.t QM size

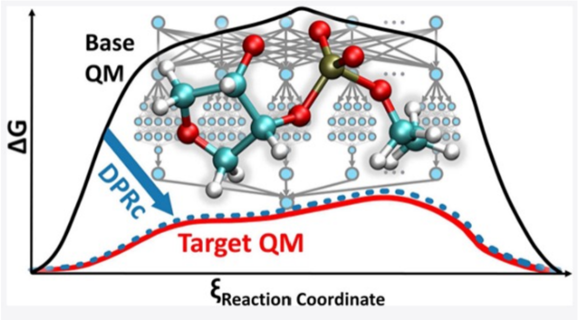
Results and Future Plans



- Preliminary QM/MM MD simulations show that CO_2 does not bind to Co(II)Mb system.
- Binding was observed in Co(0)Mb and Co(I)Mb systems and the binding energies were similar (< 5kcal/mol).
- Charge transfer from Porphyrin to CO_2 was observed upon binding.

Future plans:

- Calculate the energy landscapes associated with CO_2 activation and reduction to CO in CoMb systems.
- Use the established methodology to identify favorable SCS interactions for CO_2 binding and activation.
- Adopt machine learning framework to tackle the time scale limitations (e.g., in free energy calculations).



Zeng, Jinzhe, et al, *Journal of chemical theory and computation* 17.11 (2021): 6993-7009.