Synthesis of Two Novel Ligand Scaffolds

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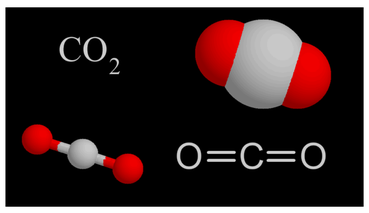
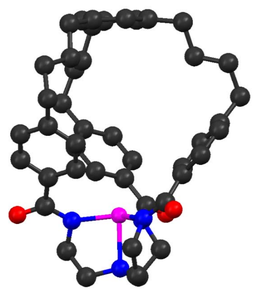
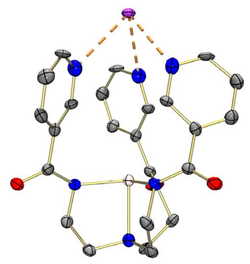
Overview



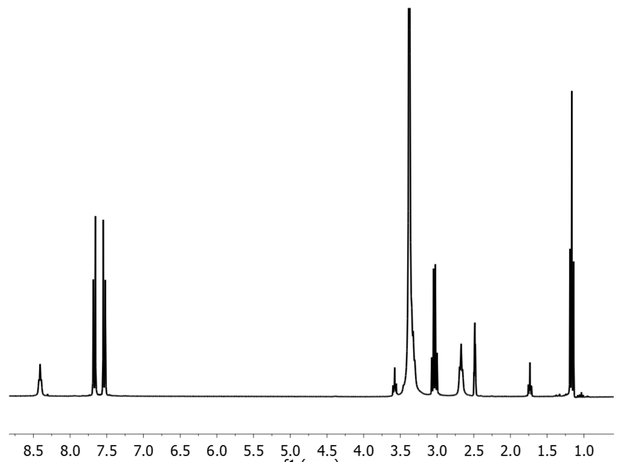
CO_2 is highly abundant, nontoxic, and easily stored. MeOH is a viable alternative liquid fuel. Developing a catalytic system to convert CO_2 to MeOH is of great interest in the search for renewable fuels. Cage ligands have been found to be potentially useful in catalysis for their ability to "trap" small molecules, such as carbon dioxide, and allow reduction to occur.

Target Ligands

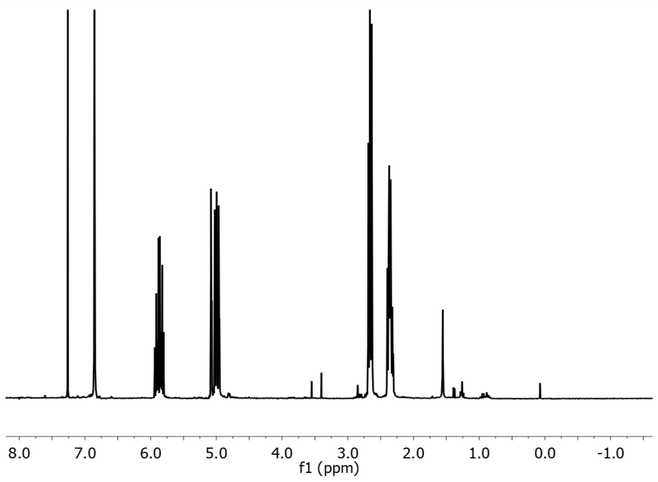




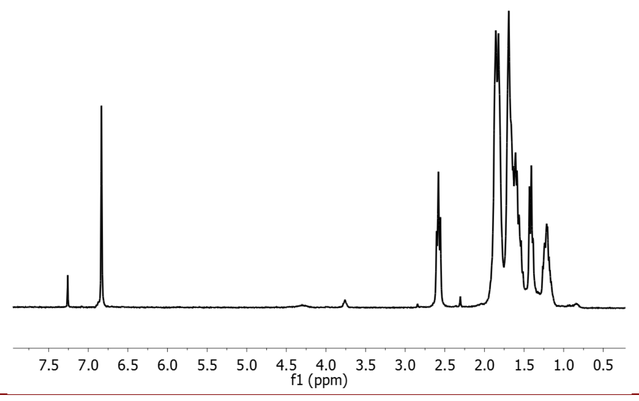
Synthesis of Base



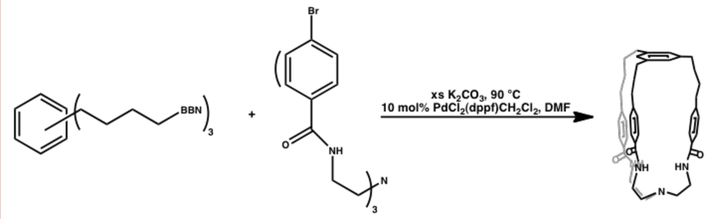
Synthesis of Cap Precursor



Synthesis of Cap

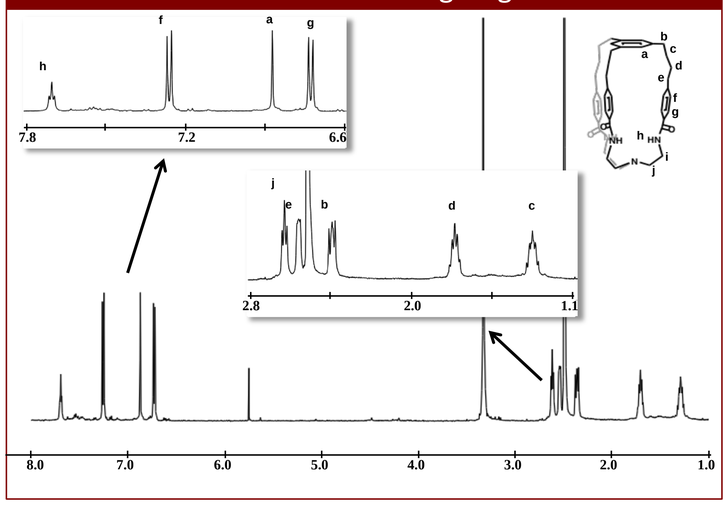


Synthesis of Amide Cage Ligand

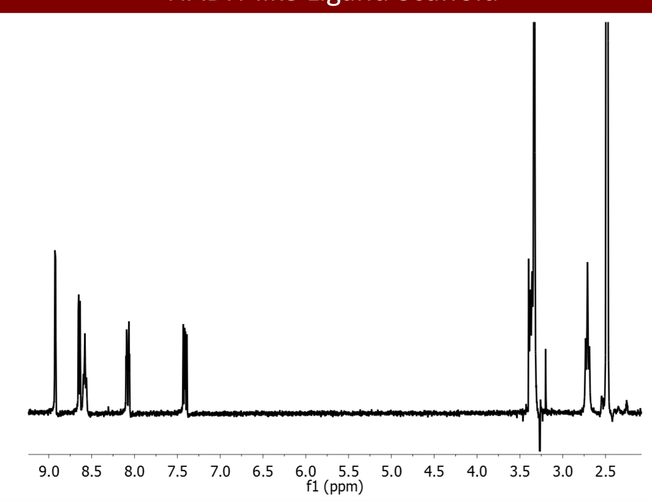


A Suzuki-Miyaura Coupling was utilized to couple the cap and base together to form the amide ligand. Dilute conditions must be used to prevent oligomerization. It was also discovered that the DMF used must be very dry in order to prevent hydrolysis at the amide linkage

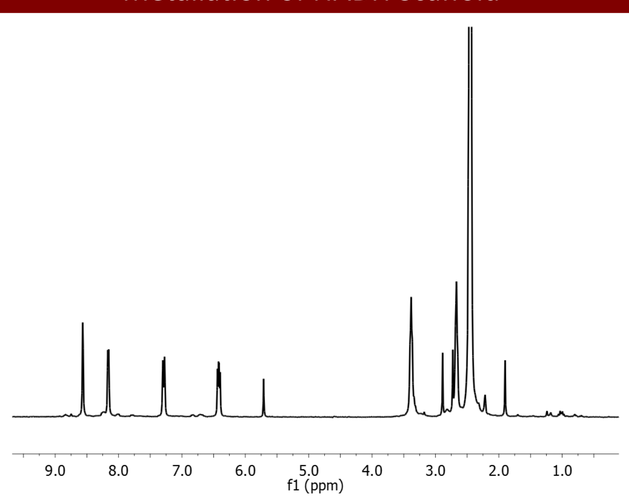
^{1}H NMR of Amide Cage Ligand



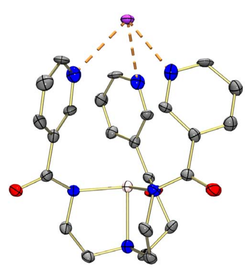
NADH-like Ligand Scaffold

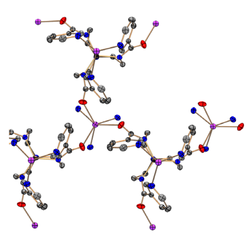


Metallation of NADH Scaffold



Crystal Structure of NADH-Zn Complex

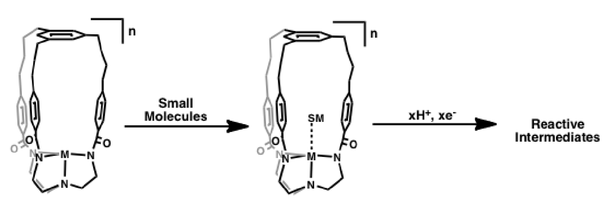




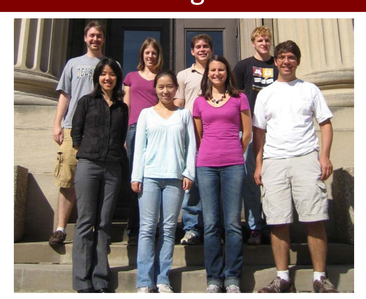
Selected bond lengths and angles	My Structure
Zn-N_{eq}	1.98 Å
Zn-N_{ax}	2.18 Å
N_{ax}-Zn-N_{eq}	85.68 °
N_{eq}-Zn-N_{eq}	119.44 °

Summary and Future Work

Summary: The synthesis of these cage ligands were carried out to aid in the development of catalytic systems for the conversion of CO_2 to MeOH. With the apprehension of these ligands, in high yields, it is our hope that they will serve as models for the development of future, novel cage systems for the reduction of small molecules.



Acknowledgements



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