

## **Investigating Hard-Soft Acid-Base Mismatch Catalysts for Renewable Fuel Synthesis**

**Eoin Gallagher, 2027**

The mounting threat of climate change expedites the necessity to eliminate fossil fuel emissions, a task that requires the development of carbon-neutral fuel alternatives. Current carbon-neutral fuels, such as hydrogen, suffer from low energy density, making meaningful storage impractical. A proposed alternative is to react hydrogen with carbon dioxide to produce a liquifiable hydrocarbon fuel with a similar energy density to fossil fuels. This reaction, however, proceeds unreasonably slowly, thus requiring a catalyst to provide a faster route to fuel production.

Our catalysts are designed to create a  $H^+$  and an  $H^-$  species based on the hard-soft acid-base mismatch theory. In essence, we will create a catalyst with a soft acid and a hard base, that will correspond to  $H_2$  which consists of a hard-acid and soft-base pair. The like species will bond to each other, creating a hard-hard match ( $H^+$  carrier) and a soft-soft match ( $H^-$  carrier). Due to the crystal lattice structures formed by our ideal catalysts and separation of their ions when dissolved in solution, we must affix these hard-acid soft-base pairs to an NHC (N-Heterocyclic Carbene) ligand to preserve the bond in solution and prevent undesired side reactions.

Previous work has focused on the NHC ligands 5-, 6-, and 7-DIPP for these catalysts, finding that they become more efficient as the ring size increases, which could be the influence of bulk, known as steric hindrance at the metal binding site, or the increased basicity of the ligand. My project alongside my partner Timmy Gee was centered around a different NHC ligand, IPR\*\*, which has the highest known degree of steric hindrance by some measures, while being less basic than the -DIPP series of ligands (Weber et al, 2012). By comparing IPR\*\*-bound catalytic efficiency to the efficiencies of the -DIPP series, we hope to distinguish whether steric hindrance or basicity are responsible for improved catalytic performance, helping us to design the best possible catalyst.

Over the 8-week research course this summer, my partner and I were able to complete the 5 synthetic steps necessary to produce a small sample of IPR\*\*, confirmed through nuclear magnetic resonance (NMR) spectroscopy. We ran into several challenges throughout the synthesis and spent much of our time making variations to the published protocol to obtain the desired products and remove impurities. The improved synthetic methods adapted as well as the synthesized IPR\*\* ligand precursors produced in the laboratory this summer will all serve useful in the continuation of my research into the academic year as I hope to conduct an independent study project to design and test a series of hard-soft acid-base mismatch IPR\*\* catalysts through hydrogen activation and  $CO_2$  hydrogenation to assess catalytic performance.

**Faculty Mentor: Professor Brandon Tate**

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References:

Weber SG, Loos C, Rominger F, Straub BF. 2012. Synthesis of an extremely sterically shielding N-heterocyclic carbene ligand. Maes B, editor. *Arkivoc*. 2012(3):226–242.