Synthesis of N-Heterocyclic Carbene Complexes of Coinage Metals and Their Application in the Activation of Hydrogen

Maryam Akramova, 2024

The development of renewable, carbon-neutral fuels is one prospective solution for the global transition from fossil fuels to sustainable energy sources. Renewable hydrogen (H_2) is a promising alternative to fossil fuels because H_2 burns cleanly and can be obtained sustainably from the electrolysis of water; however, due to its low energy density, H_2 is not a practical fuel for long-distance travel such as intercontinental flights and transoceanic shipping. Long-distance transportation requires high-density energy storage. Thus, the development of carbon-neutral fuels with high energy density is urgently needed to facilitate global energy sustainability.

My Honors project in Biochemistry aims to address the limitations of H_2 fuel by developing catalysts for the transformation of H_2 and CO_2 into energy-dense hydrocarbons, which can be used as fuel. Hydrocarbon fuels produced in this way would provide a sustainable, carbon-neutral means of storing renewable energy suitable for transportation applications. My project centers on catalysts featuring hard-soft acid-base mismatches (HSAB). By definition, hard acids and bases have low polarizability, while soft acids and bases have high polarizability. As such, hard acids prefer to pair with hard bases and form strong ionic bonds, whereas soft acids tend to pair with soft bases to form strong covalent bonds. A "mismatched" pair such as a soft acid paired with a hard base, results in a weak, reactive chemical bond, which we anticipate would provide a strategic platform for the catalytic transformation of H_2 and CO_2 to energy-dense fuels for long-distance transportation.

My project resulted in the development of a method to synthesize potential catalysts consisting of mismatched hard and soft acids and bases. Each of my target catalysts consists of a metal atom (M) serving as a soft acid, a negatively charged ion (X) serving as a hard base, and an organic molecule that stabilizes and supports the acid-base mismatch. The development of this convenient synthetic method will facilitate further studies of a variety of HSAB-mismatched compounds, whose reactivity and catalytic potential will be studied, particularly in the production of renewable fuels from H_2 and CO_2.

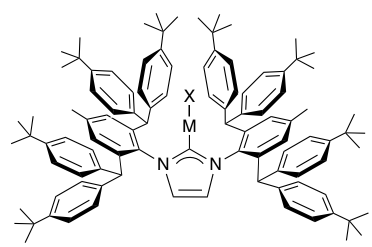

Figure 1: Chemical structure of our proposed catalyst where M represents the metal (a soft acid) and X the hard base.

Dr. Brandon Tate

Funded by the Bowdoin College Fall Research Award

Appel, A. M., et al. "Frontiers, opportunities, and challenges in biochemical and chemical catalysis of CO2 fixation." *Chem Rev* 2013, 113 (8), 6621-58.

Olah, George A., et al. "Anthropogenic chemical carbon cycle for a sustainable future." *Journal of the American Chemical Society*, vol. 133, no. 33, 2011, pp. 12881–12898

Tate, B. K.; Nguyen, J. T.; Bacsa, J.; Sadighi, J. P. "Heterolysis of Dihydrogen by Silver Alkoxides and Fluorides." *Chemistry* 2015, 21 (28), 10160–10169.

Weber, Simone G., et al. "Synthesis of an extremely sterically shielding N-heterocyclic carbene ligand." *Arkivoc*, vol. 2012, no. 3, 2012, pp. 226–242.