

Synthesis of Bimetallic Catalyst for Hydrogen Activation

Eliana Roberts, Class of 2023

A growing amount of chemical research and development is in the area of activating hydrogen gas (H_2) towards reactions with carbon dioxide (CO_2), by creating hydrogen-carbon bonds through a process called hydrogenation. With increasing concern about climate change and the detrimental effects of burning fossil fuels, efficient hydrogenation of CO_2 would enable sustainable production of carbon-neutral fuels, consisting primarily of simple hydrocarbons for the storage of renewable energy (Figure 1). However, the hydrogenation of CO_2 is a slow and challenging process in the absence of a catalyst, a reagent that allows for the reaction to progress faster and under industrially accessible conditions.

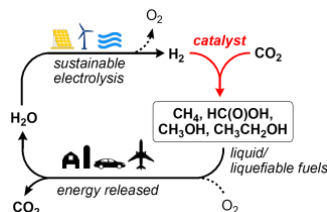


Figure 1. The cycle by which hydrogenation of CO_2 results in carbon neutral fuel¹

This project focuses on the synthesis a bimetallic catalyst, or a catalyst consisting of an organic compound bonded to two separate metal centers of different identities. Our desired ligand is a compound first published by Green et al.² in 2004 (Figure 2).

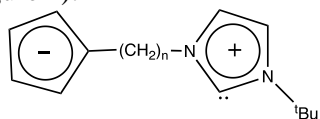


Figure 2. Proposed bifunctional ligand inspired by Green et al.²

Over the course of the summer, previous work from an independent study in the spring of 2022 was expanded upon in the synthesis of our desired bifunctional ligand. In the reaction scheme shown below (Figure 3), the total reaction scheme for the production of the tert-butyl substituted precursor ligand. Following the procedure outlined by Green et al.² with slight alterations, the overall yield of the reaction has been improved. These alterations include the use of a one-pot synthesis combining step 1 and step 2 of the synthesis scheme and changing the time conditions of the final step of the synthesis, as well as altering the steps by which the reaction mixture is purified following synthesis.

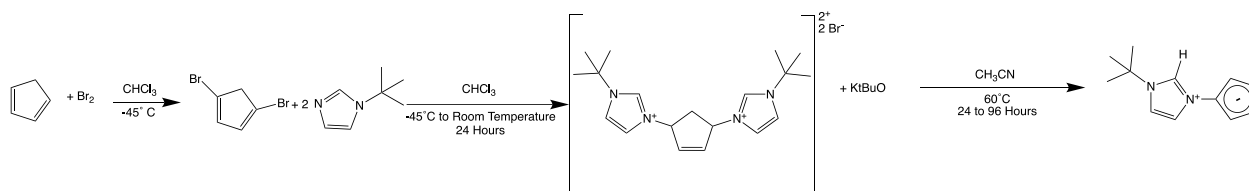


Figure 3. The final reaction scheme of the synthesis of the desired ligand precursor

Additional projects from this summer focused on the addition of other carbon-based (R) groups in place of the tert-butyl group, which would alter the steric properties of the ligand. We have had success following the same reaction scheme as initially proposed by Green et al.², but further work is needed to improve yield and purity of these reactions. Additionally, simple metalations of the final ligand were attempted with silver, with little success. Further work on this project will be continued towards the synthesis of similar ligands within the same compound, as well as further work towards the metalation of the successfully synthesized precursor ligand.

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References:

- (1) Olah, G. A.; Prakash, G. K. S.; Goepfert, A. *J. Am. Chem. Soc.* **2011**, *133* (33), 12881–12898.
- (2) Green, M. L. H., et al. *Tetrahedron Lett.* **2004**, *45* (47), 8695–8698.

