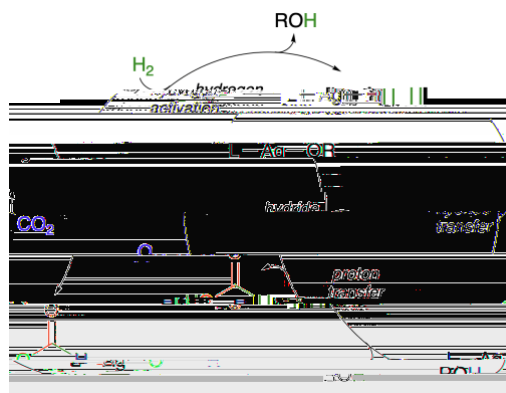
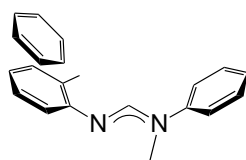


To mitigate the devastating environmental impacts of climate change in the coming decades, it is imperative that we replace the use of fossil fuels with renewable energy sources such as wind, solar, and hydroelectric. However, as these renewable energy sources are inherently intermittent, there exists a need for efficient, sustainable mechanisms to store renewable energy for later use. While the direct use of dihydrogen ( $H_2$ ) as a combustible fuel would allow for energy storage without the harmful release of carbon dioxide ( $CO_2$ ) upon combustion, the practicality of  $H_2$  as a synthetic fuel is limited by its low volumetric energy density. Instead, combining sustainable  $H_2$  production (e.g. electrolysis using energy from renewable sources) with subsequent carbon fixation (e.g. the hydrogenation of  $CO_2$ ) represents a promising pathway to the sustainable production of high-density synthetic fuels.<sup>1</sup> We hypothesize that such a process could be catalyzed by an IPr\*\*<sup>-</sup>-supported catalyst containing a hard/soft acid/base (HSAB) mismatch, with a polarizable coinage metal acting as a soft acid (Fig. 1A).<sup>2-4</sup> Following hydrogen production driven by energy generated from renewable sources, such a catalyst would facilitate the heterolysis of  $H_2$ , yielding one nucleophilic hydride poised to attack a carbon source such as  $CO_2$ , and one electrophilic hydride poised for a subsequent proton transfer event to complete the catalytic cycle (Fig. 1B).



$-Ag-Cl$  complex is an immediate precursor to our proposed silver catalysts, for which the chloride ion would be substituted for a stronger, harder base. Although our initial attempt to introduce a hard base ( $-pentoxide$ ) to the silver complex did not yield the desired product (IPr\*\*<sup>-</sup> $-Ag-O^iPent$ ), the replicable synthesis of IPr\*\*<sup>-</sup> $-Ag-Cl$  represents an important step towards our ultimate goal of constructing a catalogue of IPr\*\*<sup>-</sup>-supported HSAB mismatch catalysts.

