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Understanding the Role of Solvent in the Hydrogenation of Carbon Dioxide Using Molecular Catalysts

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The utilization of energy from intermittent and distributed sources, such as from solar and wind, would be facilitated by efficient energy storage with a high capacity. Storage in the form of chemical fuels can satisfy this requirement, but results in the need for the design of efficient catalysts for the relevant chemical transformations. The reduction of CO₂ can be advantageous over the use of H₂, as liquid fuels can be produced from CO₂ and thereafter used in transportation. The aim for our work is to develop knowledge of the parameters required for designing catalysts with unprecedented performance for the hydrogenation of CO₂. To accomplish this aim, we study each of the reduction steps in the overall transformation of CO₂ to liquid fuels.

Understanding the role of solvent upon catalysis is a route to controlling the activity of catalysts through modulation of the individual reaction steps. Towards this goal, catalysts for the hydrogenation of CO₂ to formate have been studied in both organic solvents and aqueous solution. For the relevant catalytic intermediates and overall reaction, the solvent has a substantial role in impacting the reaction thermodynamics and subsequently the catalytic mechanisms and rates. Studies of two series of bis(diphosphine) complexes will be presented: nickel hydrides¹ and cobalt hydrides.²

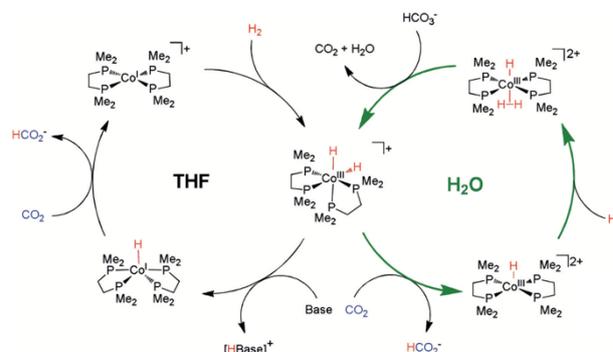


Figure 1. The mechanism for hydrogenation of CO₂ using HCo(dmpe)₂ changes between THF and H₂O, resulting in catalysis using a mild, inexpensive base.

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References

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