Does CO_2 bending and activation by a Frustrated Lewis Pair facilitate its reduction by hydride transfer?

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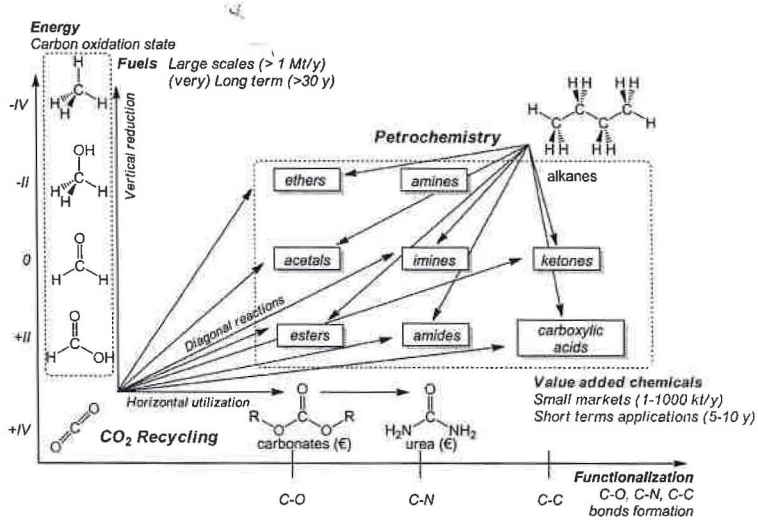
With 95 % of organic chemical commodities deriving from fossil resources, the chemical industry is currently exploring novel and renewable carbon feedstocks for the production of both bulk and fine chemicals.[1] In this context, the utilization of CO_2 or products derived from biomass wastes is an attractive strategy to access value-added products. Because these carbon sources feature carbon atoms in an oxidized state, the development of reduction methods is needed and they call for the design of efficient catalysts able to break strong C–O and C=O bonds.

Over the last years, our group has developed novel catalytic reactions for the conversion of CO_2 to formamides, N-heterocycles, methylamines and methanol, using hydroboranes, hydrosilanes or formic acid as reductants.[2-16] Extension of this methodology to SO_2 enabled the facile conversion of this gaz to sulfones, under metal-free conditions.[17] These new catalytic transformations rely on the use of simple organocatalysts, including nitrogen and phosphorus bases as well as Frustrated Lewis Pairs. The mechanisms at play in these transformations will be presented, based on DFT calculations and isolation of reactive catalytic intermediates.



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