

Metal-ligand cooperativity in homogeneous catalysis

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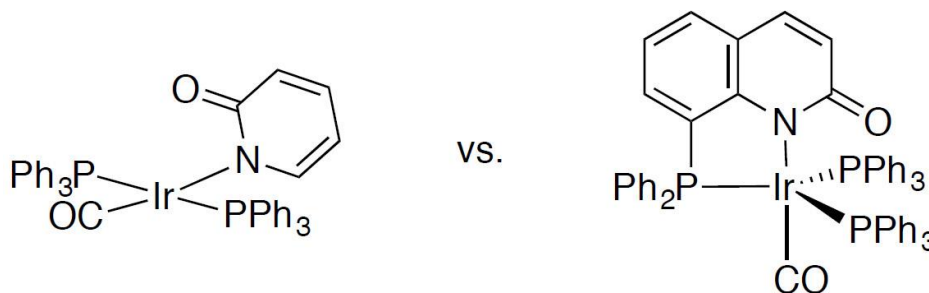
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Transition metals bearing cooperative ligands have been shown to vastly increase the scope and selectivity towards the activation of small molecules, in different fashions to traditional transition metal systems and leading to the development of new catalysts.^[1] We have become interested in the chemistry of hydroxypyridines and their application as ligands in transition metal catalysed hydrogenation and dehydrogenation reactions. In particular we are concentrating on 2-pyridones, as when ligated their deprotonated form has been shown to cooperate with the metal centre in the cleavage of dihydrogen^[2] and because the moiety is utilised by nature in the active site of iron hydrogenase.^[3]

Initially our research has been concerned with understanding the activation of dihydrogen by metal complexes bearing pyridone ligands. Similarly to previous reports we have found that pyridone iridium complexes undergo traditional oxidative addition of H₂, often leading to the dissociation of the pyridine moiety. We have used these results to influence the direction of our research, subsequently synthesising bidentate ligands which contain the pyridone moiety and exploring how iridium complexes of the ligands react with hydrogencontaining substrates.

We have also investigated a number of ligands containing the 2-hydroxypyridine moiety towards transition metal catalysed, acceptorless dehydrogenation and hydrogenation catalysis.



[1] J. R. Khusnutdinova, D. Milstein, *Angew. Chem. Int. Ed.* **2015**, *54*, 12236-12273.

[2] A. M. Royer, T. B. Rauchfuss, D. L. Gray, *Organometallics* **2010**, *29*, 6763-6768.

[3] C. M. Moore, E. W. Dahl, N. K. Szymczak, *Current Opinion in Chemical Biology* **2015**, *25*, 9-17.