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Hydrogen Atom Transfer as a Key Step in Base Metal Catalysis

Authors: Yadav, Sudha

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Abstract:

The major theme of the work presented in this dissertation is to develop an understanding of hydrogen atom transfer (HAT) and investigate it thoroughly. An inexpensive and green nickel catalyst does a wide range of catalysis including N- alkylation and formation of benzazoles starting from alcohol and amines. In case of N- alkylation, the reduced backbone of nickel catalyst extracts hydrogens during the alcohol oxidation process, stores it and redelivers it to imine in order to form amine. The alcohol oxidation step has been studied elaborately. The establishment of HAT is done by a series of experiments, as kinetic studies, KIE measurements, Kreevoy's criteria, Evans Polanyi plot and intermediate isolation. In the other part of thesis, the formation of benzazoles is discussed. A reaction sequence for the formation of benzazoles is alcohol oxidation, imine formation, ring cyclization and dehydrogenative aromatization. Both the alcohol oxidation and amine dehydrogenation steps are directly mediated by HAT, at fairly mild reaction conditions. By doing kinetic and KIE measurements, HAT is well established here too.

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