Abstract

This thesis investigates hexa-coordinated tin(IV) and tetra-coordinated tin(II) complexes as imine hydrogenation catalysts with molecular hydrogen as the reducing agent. Steric hindrance around the complexes prevents formation of a classical Lewis acid-base adduct between the Lewis acidic complexes and the imine substrate, leading to the formation of frustrated Lewis pairs (FLP). FLPs possess the unquenched reactivity of the Lewis acid and Lewis base and are capable of molecular hydrogen activation. Two factors affect FLPs' reactivity. First, highly acidic metal center with high hydride ion affinity may facilitate H_2 activation but may also bind the reaction products, and hence slow down the reaction. Second, ligand size controls the Lewis acid's steric hindrance. Too small ligands allow classical Lewis adduct formation, while too big ligands prevent substrate approach to the Lewis acid, and hence its reduction. This thesis focuses on Sn-based Lewis acids for FLP chemistry using modified tetradentate Schiff base ligands to improve the solubility of the complexes in non-polar media, and to reach appropriate steric hindrance. Mono-tert-butyl substituted complexes yield the imine hydrogenation product in 90 % yield, whereas with the di-tert-butyl substituted complexes the yield is 0 %. Various combinations of axially bound ligands are tested to influence the electronic and steric properties of the final complexes.