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Title: Redox-Active Ligand Catalyzed Borrowing-Hydrogen Reactions via Hydrogen Atom Transfer

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Abstract:

This thesis comprises a study to understand ligand-assisted single electron redox processes in cooperation with a 3d-metal, that can perform overall 2e - redox chemistry. In this endeavour, we have delineated how 2e - /2H + redox event can assist a reversible azo/hydrazo transformation, thus capturing the hydrogen from alcohol dehydrogenation. This facile redox couple has enabled us to carry out a variety of reactions via borrowing-hydrogen or oxidative dehydrogenation methodologies. The thesis is divided into six chapters. The first chapter introduces the borrowing hydrogen approach and provides a literature review of recent advances in transition metal-catalyzed alcohol oxidation and its relevance in C- and N- alkylation reactions. It further expands towards sustainable synthesis of N-heterocyclic compounds, often via oxidative dehydrogenation route. The catalytic activity of well-defined, azo-phenolate ligand-coordinated nickel catalysts for C-C and C-N bond-forming processes is the subject of chapters two to four. These reactions operate via borrowing-hydrogen methodology or oxidative dehydrogenation strategy with the liberation of H 2 O and H 2 / H 2 O 2. Chapter 5 describes a redox-active iminoquinone motif coupled to a delocalized pyrene core (PA), that also enables facile 2e - /2H + reversible redox cycle to accomplish alcohol dehydrogenation under photochemical conditions. Chapter 6 includes the experimental methodologies and spectroscopic data for the compounds mentioned in the thesis.

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