INDIAN INSTITUTE OF TECHNOLOGY GUWAHATI
SHORT ABSTRACT OF THESIS

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Thesis Title: **Exploring the Potential of Homogeneous Ru-SNS/NNS Complexes and Heterogeneous Ru-Hydrotalcite in De(hydrogenative) Transformations**

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Thesis Submitted to the Department/ Center	:	Chemistry
Date of completion of Thesis Viva-Voce Exam	:	13.10.2023
Key words for description of Thesis Work	:	Homogeneous Catalysis, Heterogeneous Catalysis, Transition Metal, De(hydrogenative) transformations

SHORT ABSTRACT

The contents of the present thesis entitled as “Exploring the Potential of Homogeneous Ru-SNS/NNS Complexes and Heterogeneous Ru-Hydrotalcite in De(hydrogenative) Transformations” have been divided into five chapters. The first chapter contains a brief literature study related to various de (hydrogenative transformations) and the last four chapters were based on the results achieved from the experimental works performed during the entire course of the PhD research program. Chapter 1 contains a brief introduction to the literature review of acceptorless dehydrogenation and borrowing hydrogen reaction of alcohols via homogeneous catalysis and heterogeneous catalysis. In 21st century, the rapid depletion of fossil fuels and growing environmental concerns urges chemists and chemical industries to search for alternative raw materials and to develop new methodologies to produce sustainable chemicals and important building blocks. In this regard, biomass-derived alcohols was found to be best candidate, as they are non-toxic in nature. Moreover, alcohols are considered renewable starting materials that can be used in organic synthesis for various organic transformations and the preparation of commodity chemicals. In this context, “acceptorless dehydrogenation (AD)” and “borrowing hydrogen (BH)” catalysis plays a key role. These approaches are sustainable because this process liberates water and in some cases (i.e., AD) molecular hydrogen as clean by-products. And, these types of reactions could be successfully performed by various types of homogeneous and heterogeneous catalysts.

Chapter 2 contains the synthesis and application of NNS and SNS ligand-derived air stable-Ru pincer complexes and their catalytic applicability towards deoxygenative coupling of primary alcohols. In this approach, the catalytic activity of various Ru-SNS/NNS was thoroughly studied for deoxygenative coupling of primary alcohols to selectively synthesise both alkenes and alkanes. Various control experiments and mechanistic and kinetic studies were done. Both homo- and cross-coupling reactions were successfully conducted and the expected product was obtained in good yield. The outcome of chapter 2 was published in *Organometallics*, **2023**, *42*, 55 – 61.

Chapter 3 contains the synthesis of 4-quinolone analogues, as 4-quinolone analogues are well known for their wide range of bioactivities and prominent applications in medical science and pharmacology. The reaction was selective towards 2,3-disubstituted-4-quinolone derivatives and barely formed C/N-alkylated products. Furthermore, the modified protocol could be utilized for a wide range of alcohols and diverse amino acetophenones like α-functionalised 2'-aminoacetophenone. The synthetic utility of the methodology was highlighted by synthesizing 4-quinolones with antibiotic properties. The outcome of chapter 3 was published in *Chem. Commun*., **2023**, *59*, 9267-9270.

Chapter 4 explores the catalytic potential of Ru-grafted hydrotalcite in a multicomponent reaction. With this catalyst, an efficient and atom-economic method for the facile synthesis of 1,8-Dioxo-decahydroacridine derivatives was developed via acceptorless dehydrogenative multicomponent reaction. This protocol was quite general to access the desired products in a wide range of substrates with good to excellent yields. In addition, mechanistic and kinetic studies were performed to understand the plausible reaction pathway involved for the target product formation which was discussed in this section in detail. A time-dependent product distribution experiment was also presented and the reaction scale-up was performed to highlight the practical utility of this strategy. The outcome of chapter 4 was published in *Asian J. Org. Chem*., **2021**, *10*, 2195.

Chapter 5 highlights the utilization of Ru-grafted hydrotalcite for the N-alkylation of benzamides and sulfonamides with alcohols via borrowing hydrogen catalysis. In this work, weakly nucleophilic amides were alkylated with various primary alcohols, including benzyl, heteroaryl and aliphatic alcohols. To shed light on the mechanistic details, several control studies and deuterium labelling experiments were performed. The reaction could be easily scaled up without any detrimental effect on the yield. Moreover, the catalyst was found to be capable of synthesizing quinazolinone directly from 2'- aminobenzamide and alcohols. The outcome of chapter 5 was published in *J. Org. Chem*., **2022**, *87*, 9, 5556–5567.