

## ABSTRACT

Among various onium salts, the pyridinium salt is an important heterocyclic motif which is widely present in a range of biologically active compounds as well as show applicability in the diverse fields of synthetic and material chemistry. On the other hand, imidazo[1,2-*a*]pyridines and amides represent significant class of compounds with extensive biological, medicinal and optoelectronic applications. The synthesis and functionalization of imidazo[1,2-*a*]pyridine with novel methods is desired both from economic and environmental point of view. The current thesis entitled “**Synthesis of Functionalized Imidazo[1,2-*a*]pyridines and Amides using Pyridinium Salts**” focuses on synthesis of functionalized imidazo[1,2-*a*]pyridines and amide derivatives from pyridinium salts *via* C–C and C-X (X = N, O, S) bond formation strategy using transition-metal catalysts as well as transition metal-free conditions. In this thesis, imidazolium-supported 2-chloropyridinium triflate has been synthesized and used for amide bond formation and further 2-amino-functionalized pyridinium salts were utilized to synthesize imidazo[1,2-*a*]pyridines *via* C-X (X = N, O, S) bond formation in presence of iodine, base or palladium/copper as catalyst. A brief outline of the work presented in the thesis is given below.

**The first chapter** of the thesis deals with the recent literature survey on the application of pyridinium salts as reagents and as efficient reactive partner for the synthesis of aza-fused heterocycles. Initially, importance of pyridinium salts and synthetic protocols to access pyridinium salts are discussed. Next, application of pyridinium salts in the synthesis of pyrazolopyridine, indolizine and imidazopyridines were discussed concisely. At last the application of pyridinium salts as efficient reagent in different organic transformation was summarized.

**The second chapter** of the thesis describes the synthesis and application of imidazolium-supported 2-chloro pyridinium salt as an efficient condensation reagent for synthesis of amides. The synthesised reagent was fully characterised using NMR, IR and mass spectroscopic technique and subsequently utilized for the synthesis of library of amides. All synthesized product were characterized using IR and NMR technique. The reaction conditions are benign, required shorter reaction time and furnished good yields of amides with a variety of substrates. The evasion of column chromatography, regeneration and reuse of the reagents are salient features of the methodology.

**The third chapter** of the thesis describes a simple and highly efficient protocol for the chemoselective synthesis of carbonyl-functionalized imidazo[1,2-*a*]pyridines *via* iodine-mediated 5-*exo-dig* type intramolecular cyclization of 2-amino-1-propargylpyridinium bromides in the presence of a 1N NaOH. Various substituted imidazo[1,2-*a*]pyridine derivatives were obtained in good to excellent yields (45-89%). Additionally, direct access to 2-methylimidazo[1,2-*a*]pyridines from pyridinium salts was carried

out in presence of base through intramolecular hydroamination reaction. A facile and greener approach combined with good to excellent yields of products are advantages of the protocol.

**The fourth chapter** of the thesis deals with the tandem intermolecular cyclisation of 2-aminopyridinium salts and 2-bromocarbaldehydes for the synthesis of chromone/pyrano-fused imidazo[1,2-*a*]pyridines. A direct one-pot tandem synthesis of chromeno-annulated imidazo[1,2-*a*]pyridines is accomplished by the reaction of 2-amino-1-(2-ethoxy-2-oxoethyl)pyridinium salts with 2-bromoarylaldehydes using Pd(TFA)<sub>2</sub> as a catalyst and Cu(OAc)<sub>2</sub> as an oxidant. The overall scheme comprises tandem base-mediated amidation and Knoevenagel condensation, followed by palladium-catalyzed Wacker type oxidation and intramolecular C–O coupling reaction. The method is simple, tolerates different functional groups to generate 33 examples of desired products with moderate to good yields (28-77%).

**The fifth chapter** of the thesis deals with the KOH-mediated cross-dehydrogenative C(sp<sup>3</sup>)-S bond formation between 2-aminopyridinium salt and thiols to access 3-sulfenylimidazo[1,2-*a*]pyridin-2-ol derivatives in acetonitrile at 30 °C. The reaction afforded 29 examples of differently substituted 2-hydroxy-3-sulfenylimidazo[1,2-*a*]pyridine derivatives in 56-95% yields. The reaction is believed to proceed through intramolecular amidation followed by cross-dehydrogenative C(sp<sup>3</sup>)-S coupling reaction.

The simultaneous inclusion of hydroxy functionality in the molecule give added advantages for further functionalization and generation of complex scaffolds. The hydroxyl group was further utilized to access 2-aryl-3-sulfenylimidazo[1,2-*a*]pyridines.

Finally, in the **sixth chapter** of the thesis, a summary of the thesis work is presented along with future scope of the research work.