ABSTRACT

The thesis entitled " ***Development of novel metal and non-metal catalysts for the sustainable synthesis of bis(indolyl)methanes, C-3 alkylated oxindoles, and benzoic acids*** " presents the results obtained from the research work carried out on the development, characterization, and application of transition metal and main group-based compounds as catalysts and reagents for oxidation and C-C bond formation reactions. Oxidative coupling of benzyl amines with indole was carried out using a water-soluble cobalt complex. The oxidative coupling of benzyl alcohols and oxindoles was carried out using a phosphine-free manganese complex as the catalyst. Main group compound I_2 was utilized for the oxidative coupling of methyl 3,3-dimethoxypropanoate with indoles. *In situ* generated halides (Cl^-, Br^-, I^-) were used as the main group element-based catalyst for the oxidation of benzyl halides to benzoic acids. The results discussed in this thesis have been broadly divided into four parts. The *first* part describes the oxidative coupling of benzylamines with indoles, using an inexpensive water-soluble cobalt complex, [Co(bpbH_2)Cl_2] [bpbH_2 = N,N'-bis(2'-pyridinecarboxamide)-1,2-benzene] as the catalyst. This catalyst was quite effective when air was used as the oxidant and water as the solvent under mild reaction conditions. Aromatic amines were successfully converted to the corresponding bis(indolyl)methanes (BIMs) in good yields. The *second* part of the work deals with a new air-stable phosphine-free 8-AQ (8-aminoquinoline) based Mn(I) carbonyl complex as the catalyst for the C(α)-alkylation of oxindoles with alcohols. The Mn complex [(8-AQ)Mn$(CO)_3$Br] works effectively as a catalyst for the α-alkylation of oxindoles by both secondary as well as primary alcohols. Ease of catalyst preparation using an inexpensive ligand with a Mn carbonyl precursor are additional interesting features of this methodology. The procedure has been used to synthesize pharmaceutically important C-3 alkylated oxindoles. The third part of the work describes an efficient and metal-free method for synthesizing bis(indolyl)methanes in the water medium. Water as an inexpensive and green solvent and iodine as a catalyst make this protocol economical, environmentally benign, and potentially suitable for commercial and academic applications. Various biologically active BIMs, such as streptindole and its derivatives, have been synthesized in a single step from indole in good yields. The fourth part of the work focuses on an efficient and metal-free methodology for the oxidation of benzyl halides to benzoic acids using an inexpensive and green oxidant (TBHP) in an aqueous basic medium. This protocol offers an excellent way to avoid added catalysts and involves the use of an *in situ* generated halide ion as the catalyst. It is also the first report on the oxidation of benzyl

iodides to benzoic acids. This methodology provides a broad scope for realizing carboxylic acids from benzyl halides in high yields under mild reaction conditions without the need of column chromatographic purification.