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Ruthenium Heteroleptic Complex Immobilized to SBA-15 for Visible Light Driven Oxidative Cyanation of Aromatic Tertiary Amines

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Abstract—*The direct oxidative cyanation of C-H bonds in tertiary amines to give corresponding α-aminonitriles has gained importance in recent years, prominently due to highly useful and versatile intermediates which find wide application in the construction of biologically active nitrogen compounds such as alkaloids. Dual reactivity shown by these bifunctional organic compounds, as nucleophilic addition provides an easy access to other compounds such as α-amino aldehydes, ketones and β-amino alcohols.[1] Considering the huge importance of these compounds in nature, scientists have dedicated their research in the development of clean and environmentally benign methods for the transformation of amines into the corresponding α- functionalized compounds. Sunlight is an inexhaustible source of energy and can provide activation energy required for the organic transformation.[2] Due to this, scientific community has been attracted towards the solar light harvesting for the organic transformations which is considered most economical source of energy. Till date, most of the work was carried out on semiconductor photocatalysis which works by generating electrons and hole pairs but in the semiconductor photocatalysis conversion efficiency and quantum yield was found low in visible light due to wide band gap. Metal complexes due to their visible light absorption efficiency are considered to be suitable catalysts for visible light driven organic transformations. But, the main challenge associated with these redox catalysts is their homogenous nature which restricts their recyclability. However, immobilisation of these homogenous metal complexes on photoactive supports like graphene, carbon nitride, SBA-15 etc. makes them recyclable as well increase their catalytic performance.*
Herein, we report a new microwave synthesized heteroleptic ruthenium previously synthesized by our group and characterised by ^{1}H and ^{13}C NMR, ESI-HR-MS and UV-Vis spectroscopy. The synthesized catalyst was immobilized on highly mesoporous SBA-15 material as efficient photoredoxcatalyst for oxidative cyanation of wide range of tertiary amines to corresponding α-aminonitriles via C-H activation and characterized by FTIR, TGA, XRD, ICP-AES, BET, CHNS, UV-Vis etc. The catalytic activity of the photocatalyst was tested for the oxidative cyanation of various tertiary amines under visible light irradiation. The developed catalytic system yields α-aminonitriles from corresponding amines in excellent yields. Due to the heterogenization of the synthesized catalyst on the photoactive support, it can be recovered and reused for further reactions without much loss in the activity.

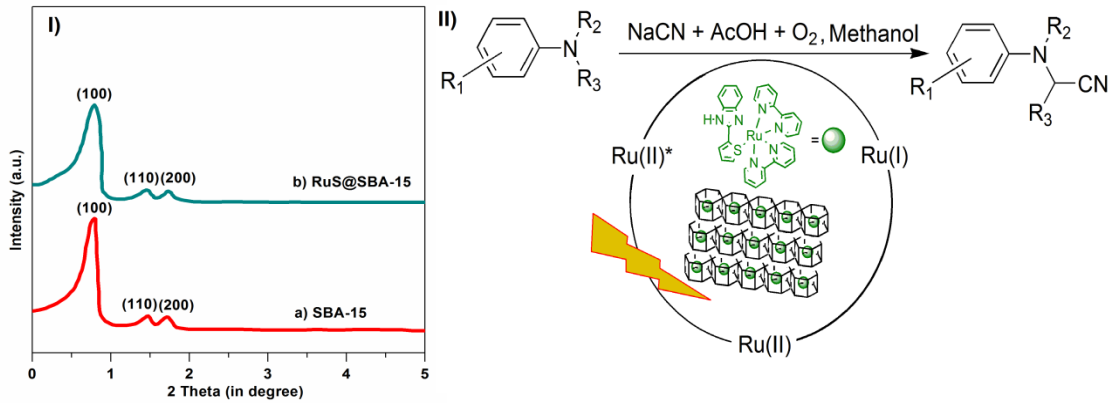

Fig. 1 I): Short angle XRD diffraction pattern of a) SBA-15 b) RuS@SBA-15 II) Schematics of visible light induced photocyanation of tertiary amines.

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