PREFACE

Heterocyclic compounds over the years have gained lots of attention due to their high biological and pharmaceutical applications. They are widely distributed in nature and possess many medicinal activities like anti-fungal, anti-cancer, anti-malarial, anti-tumor anti-HIV, anti-microbial etc. Because of these reasons, various research groups throughout the world have developed a number of methodologies for the synthesis of heterocyclic compounds. All methods have their own advantages, but at the same time many of them possess economic and environmental limitations. Hence development of newer methodologies which addresses the above mentioned problems are always desirable.

Because of the growing concerns on environment, since last few decades, scientific community worldwide is giving much importance on implication of greener protocols in organic synthesis. Sustainable chemistry is a branch of chemistry which deals with development of eco-friendly procedures. Recently synthesis of efficient cascades having high structural diversities with least number of steps is much demanding and the best way to achieve this is via multi-component reaction. Multi-component reactions allow creation of several new bonds in a single step and offer notable advantages like operational simplicity, convergence, simple extraction, easier purification of desired product etc.

Catalysis is the heart of many chemical reactions because it lowers the activation energy and makes the reaction feasible. Application of clean and reusable catalytic material is one of the principles of green chemistry. Surface coated metal nanoparticles and graphite oxide because of their heterogeneous nature offers the scope of easy separation and recyclability.

In this thesis entitled “Multi-component Synthesis of Heterocyclic Compounds of Biological importance using surface Coated Metal Nanoparticles and Graphite Oxide as Catalysts” I have discussed about the synthesis and characterization of various surface coated metal nanoparticles and graphite oxide, and their application in multi-component synthesis of biologically relevant heterocyclic compounds. The thesis consists of five chapters. In the first chapter, I have discussed about importance of green and sustainable chemistry, advantages of multi-component reaction, significance of applying surface coated metal nanoparticles and graphite oxide as catalysts. Recent literature reports on the synthesis of various biologically active heterocyclic moieties by the application of surface coated metal nanoparticles and
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graphite based oxides via one-pot multi-component reaction procedure has also been incorporated in the first chapter. In the second chapter several drawbacks corresponding to application of organocatalyst like loss of organic component during work up and difficulty in recyclability was discussed. And to tackle these problems, grafting of glutathione on nano-ferrite has been reported. Nano-ferrite supported glutathione (nano-FGT) after being synthesized was characterized by various analytical techniques like Fourier transform-infrared spectroscopy (FT-IR), scanning electron microscopy (SEM), transmission electron microscopy (TEM), energy dispersive X-ray analysis (EDX), powder X-ray diffraction (PXRD), thermogravimetric analysis (TGA) and inductively coupled plasma optical emission spectroscopy (ICP-OES). After successful characterization it was applied as an efficient nano-organocatalyst for the one-pot multi-component synthesis of phthalazine derivatives under solvent-free reaction condition (SFRC). Nano-FGT (nano-ferrite supported glutathione) being magnetic in nature was easily recycled from the reaction mixture and was applied in subsequent reactions. In the third chapter, synthesis of nano-FDP (nano-Fe$_3$O$_4$-DOPA-L-proline) from readily available starting materials was discussed and its characterized by FT-IR, SEM, TEM, EDX, PXRD techniques have been reported. In order to determine the magnetic nature of the catalyst, a vibrating sample magnetometer (VSM) analysis was also performed. The prepared nano-FDP efficiently catalyzed the reaction for synthesis of pyran derivatives using water as solvent at room temperature, providing good to excellent yield of the product via one-pot multi-component reaction process under ultra-sonic irradiation. After completion of reaction nano-FDP was easily recycled from the reaction mixture by using external magnetic field and reused in subsequent runs without much decrease in catalytic activity. The fourth chapter discusses about synthesis of nano-Fe$_3$O$_4$-DOPA-SnO$_2$ and its characterization by using common analytical techniques. Further, an X-ray photoelectron spectroscopy (XPS) analysis was also performed to find out the oxidation state of Sn (tin) present in the catalyst. Turn over number (TON) and turn over frequency (TOF) of nano-Fe$_3$O$_4$-DOPA-SnO$_2$ was also calculated based on the amount of the active metal. The prepared magnetically recyclable catalyst was then efficiently applied for the one-pot multi-component synthesis of dihydroquinazolinone and bis-dihydroquinazolinone derivatives by using water as solvent. To further extend the scope of present methodology, a gram scale reaction and conversion of dihydroquinazolinones to
biologically more important quinazolinones have also been performed. In order to check whether there is any leaching of metal from catalyst, a hot filtration test as well as an inductively coupled plasma atomic emission spectroscopy (ICP-AES) was also carried out. Reusability of nano-Fe$_3$O$_4$-DOPA-SnO$_2$ was also successfully checked till five consecutive runs. Finally the fifth chapter, which discusses about one-pot multi-component synthesis of dihydropyrimidine derivatives, has been divided in to two parts. Part A: discusses about synthesis, characterization and application of nano-Fe$_3$O$_4$@SiO$_2$@SO$_3$H (nano-ferrite coated with silica sulfonic acid) for the synthesis of dihydropyrimidine derivatives. Wide substrate scope employing various aldehydes, amine sources and β-dicarbonyl compounds were investigated for the present catalytic process. A recyclability experiment for the synthesized catalyst was performed and it was found that catalyst which was magnetically recyclable can be easily reused in subsequent runs without much depreciation in catalytic activities. In part B, graphite oxide, a highly active metal-free carbocatalyst was employed for the synthesis of above mentioned heterocyclic moiety. Graphite oxide was synthesized by oxidation of graphite powder and characterized by several techniques like FT-IR, SEM, TEM, EDX, PXRD, Raman, TGA and XPS analyses. Catalytic activity of graphite oxide was investigated for the synthesis of dihydropyrimidine derivatives under solvent-free reaction condition at 60 °C. Good result of present methodology in gram scale reaction proved that this procedure can be also applied in industries. Graphite oxide was also found to be effective in recycling experiments and was reused in nine consecutive runs without much decrease in catalytic activity.