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MINI REVIEW

Recent Advances in Metal Nanoparticles for the Synthesis of *N*-Containing Heterocyclic Compounds

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Heterocyclic compounds are indispensable organic structures because of their promising potential applications in medicinal, pharmaceutical and other related fields of chemistry. Exploring these heterocyclic compounds by evaluating metal nanoparticles as efficient nanocatalysts have attracted the researchers over the past decades, due to their stability, recyclability, reusability rather than conventional catalysts. Most of the nano-catalyzed organic reactions are taking place in benign and under green conditions. In this mini-review, we have documented synthesis of some of the recent *N*-containing heterocyclic compounds by utilizing advanced nanoparticles as effective catalysts. It has been concluded that exercising these nanocatalysts for the synthesis of *N*-containing heterocyclic compounds not only enhanced the yield of the product but has extensively increased the selectivity of the desired products *via* greener approach.

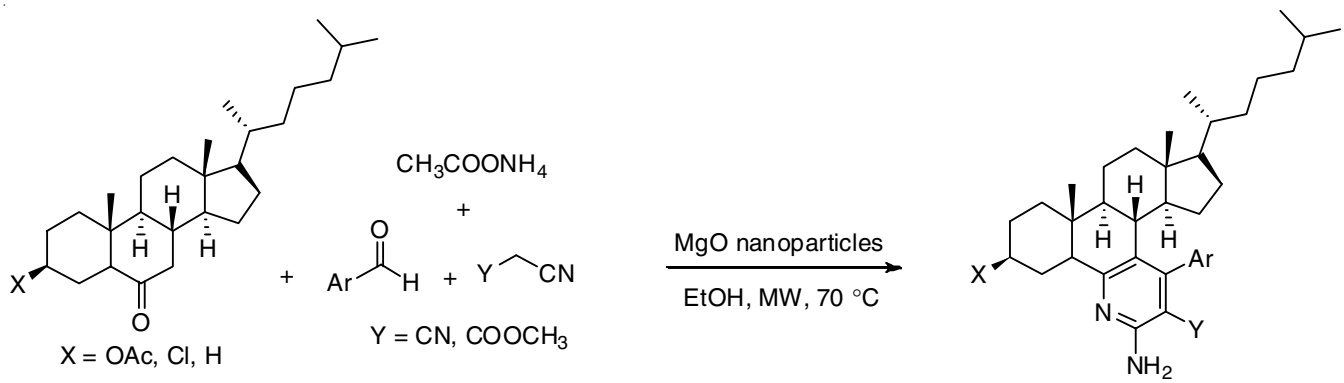
Keywords: Heterocyclic compounds, Metal nanoparticles, Nanotechnology, Pharmaceutically important compounds.

INTRODUCTION

Heterocyclic compounds are the cyclic systems, which contain two or more than two different kind of hetero atoms. The syntheses of these heterocyclic compounds are useful for the researchers at laboratory level as well as industrial purposes. These reactions can be affected by the reagents, which are immobilized on the nanocatalyst support having many advantages over conventional methods. The design, stability, selectivity and recyclability of several nanocatalysts produces lower toxic byproducts and making the reactions more environmentally friendly, which is indeed a step towards the green approach of syntheses of *N*-heterocyclic compounds [1].

Heterocyclic compounds have a wide range of applications in pharmaceuticals, agrochemicals, veterinary products, *etc*. They can also be used as sanitizers, developers, antioxidants, *etc*. Nitrogen-containing heterocyclic compounds have vital applications because of their abundance in nature. Several nitrogen containing heterocyclic compounds exhibit many biological activities like as antibacterial, antiarthritis, antiasthamatic [2-7]. Nitrogen containing heterocycles also show their important role as ligands, explosives, stabilizers in photography, *etc*. [8-13]. Other nitrogen containing heterocycles *viz.* dihydropyrimidinones and diazepines show biological and medicinal importance such as calcium channel blocker, antihypertensive agents, antitumor compounds [14-19], antibiotics [20,21], viral infection such as HIV [22-25], control cardiovascular disorders [26,27], medical importance in cancer treatment [22].

These compounds can be synthesized by using multicomponent reactions (MCRs). Approximately, all atoms contri-



Catalyst	Solvent	Temp. (°C)	Time (h)	Yield (%)
MgO NPs	Methanol	70	6.0	77
MgO NPs	DMSO	70	7.0	68
MgO NPs	DMF	70	7.0	70
MgO NPs	Acetonitrile	70	6.5	72
MgO NPs	Chloroform	70	8.0	67
MgO NPs	Toluene	70	8.0	62
MgO NPs	Dichloromethane	70	8.0	65
MgO NPs	Ethanol	70	6.0	79
MgO NPs	Ethanol	70, MW	20 (min)	89

Scheme-I

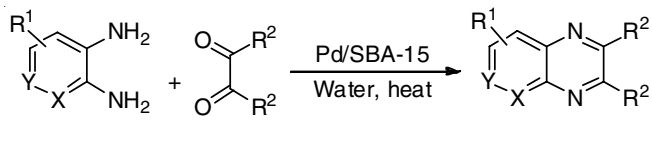
bute to form a new products [28]. Multi-component reactions are important for the fast and ecofriendly syntheses of various heterocycles. Multicomponent reactions are efficient for synthesis of compounds with pharmaceutical and biological properties [29-32]. Such kind of reactions display a significant number of advantages compared to conventional linear-step syntheses [32].

In recent years, nanocatalysts are being very helpful to upgrade the selectivity and simpler procedure for the synthesis of heterocycles [33-37], which among them *N*-bearing heterocycles as structurally subunit of naturally occurring compounds provided well-designed biologically active products. In continuation, we attempted to highlight some recent literatures which focused on metal nanoparticles as effectual catalysts.

Metal nanoparticles for synthesis of heterocyclic cores: Design of metal nanoparticles allows the developing of organocatalysts to functionalized metal heterogeneous retrievable catalysts, which can be easily synthesized using metal salt as a low-cost precursor. Various techniques studied for the synthesis of metal nanoparticles [38,39]. In recent years, green synthesis of metal nanoparticles remarkably developed by researchers [40-43]. These metal nanocatalysts can be utilized in multicomponent reactions for the synthesis of heterocyclic compounds as significant precursor of pharmaceuticals. Herein, we have summarized some of the syntheses using metal based nanocatalyst such as Mg, Cu, Ni, Co, Fe, *etc.* for *N*-bearing heterocyclic compounds:

Pyridine cores: Pyridine derivatives are significant class of bioactive molecules that have shown dramatically treatment properties in medicinal chemistry [44-46]. The MgO nanocatalyzed microwave assisted multicomponent reaction for synthesis of polysubstituted steroid pyridines reported by Ansari *et al.* [47]. A mixture of steroidal ketones aromatic aldehyde, malononitrile, ammonium acetate was examined in the presence of MgO nanoparticles as a catalyst in ethanoic media. When the reaction mixture was irradiated in a microwave oven at 70 ºC led to higher yield (89%) and short reaction time (20 min) (**Scheme-I**) [47].

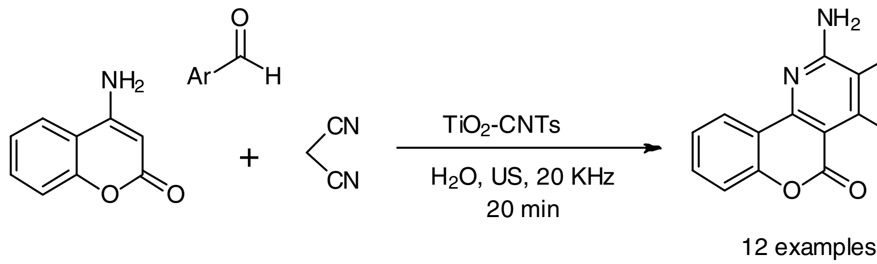
Polyheterocyclic systems containing nitrogen atoms are valuable scaffolds owing to their fascinating therapeutic properties. Cyclocondensation of 1, 2-diamines and 1,2-diketones by means of a catalytic amount of the SBA-15-supported palladium nanocatalyst provided pyridopyrazine derivatives in green conditions with high yields (**Scheme-II**) [48].



Scheme-II

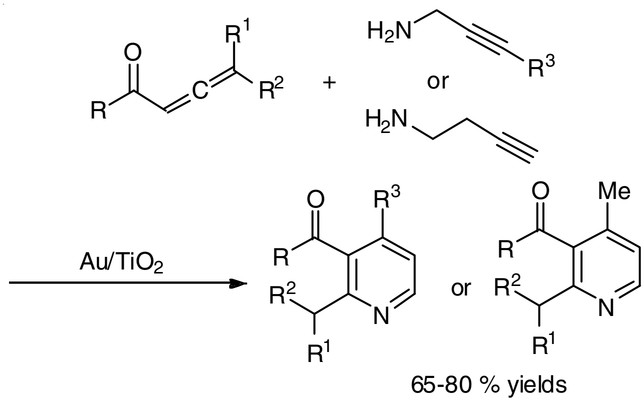
A new synthesis of chromeno[*b*]pyridines was reported by Abdolmohammadi *et al.* [49]. The condensation reaction of 4-aminocoumarin, malononitrile and aromatic aldehydes under ultrasonic irradiation in green media using TiO_2-CNTs as an effective catalyst led to a series of high yielded pyridine derivatives in a short reaction time (**Scheme-III**). This well-designed metal nanoparticle catalyst made by TiO_2 nanoparticles immobilized on carbon nanotubes [49].

A valuable one pot cyclization reaction of *N*-propargyl or *N*-homopropargyl with β-naminones and then dehydrogenation provided good yields of substituted 3-keto pyridines or 4-picolines by aid of catalytic activity of gold nanoparticles on TiO_2 (**Scheme-IV**). Reaction of conjugated allenone or

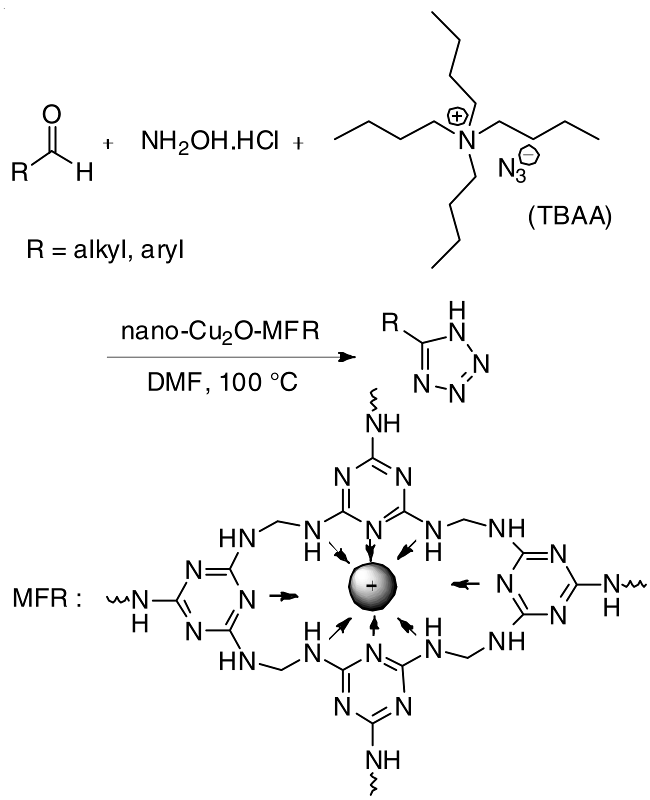


Ar	Yield (%)
C_6H_5	94
3-Cl-C_6H_4	93
2,4-Cl_2-C_6H_3	93
4-NC-C_6H_4	95
4-F-C6H4	96
3-HO-C_6H_4	94
4-HO-C_6H_4	95
4-CH_3O-C_6H_4	96
4-CH_3-C_6H_4	95
3-O_2N-C_6H_4	96
Pyridin-4-yl	96
Thiophen-2-yl	95

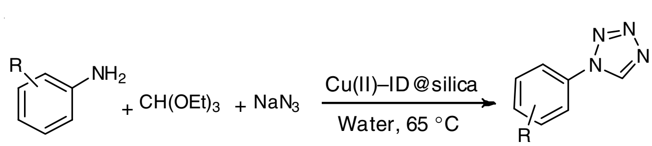
Scheme-III



Scheme-IV



Scheme-V



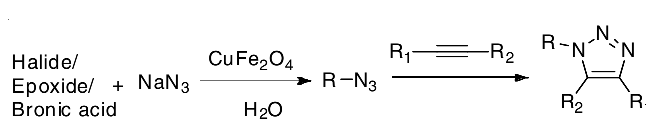
Scheme-VI

allenyl ester with the alkynylamine formed *in situ* enaminones. The outstanding protocol developed a reusable Au/TiO_2 catalyst for pyridine core synthesis, whereas the known cyclization methods give 1,4-oxazepines in the presence of Au(I) or Au(III) catalysts [50].

Triazole and tetrazole cores: Today, triazole and tetrazole containing compounds have highly regarded attention due to their individual properties such as antibacterial, antifungal, anticancer and HIV protease inhibitors [51-64]. Three component one pot synthesis of aldehyde, hydroxylamine hydrochloride and tetrabutylammonium azide (TBAA) was taken for synthesis of 1*H*-tetrazoles using doped copper oxide nanoparticles on melamine-formaldehyde resin (Cu_2O-MFR) as catalyst under 100 ºC and DMF solvent described by Behrouz [65] (**Scheme-V**).

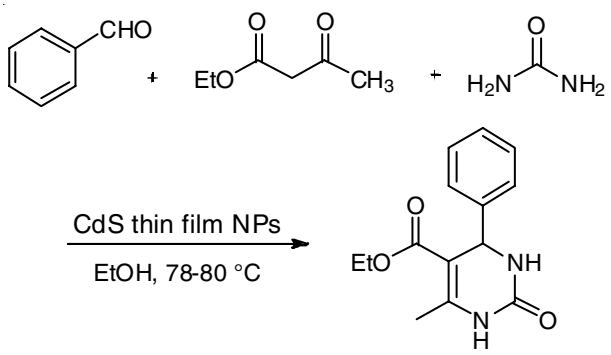
A novel nano-heterogeneous copper catalyst was developed by Sharghi *et al.* [66]. A nano-silica supported copper(II)-2-imino-1,2-diphenylethan-1-ol complex was used for the synthesis of some five-membered *N*-heterocycles through C–N bond formation reactions. Three component reaction of amines, triethyl orthoformate and sodium azide in the presence of 1.0 mol% nono catalyst at 65 ºC gave high yield products under mild conditions [66] (**Scheme-VI**).

An efficient and green protocol for three-component Huisgen 1,3-dipolar cycloaddition click reaction using $CuFe_2O_4$ nanoparticles as a heterogeneous catalyst in aqueous media as reported by Mondal *et al.* [67]. Cycloaddition reaction of alkyl halide, epoxide or boronic acid with sodium azide and alkynes under aqueous media afforded 1,4-disubstituted 1,2,3-triazoles in high yields and short reaction time (**Scheme-VII**).



Scheme-VII

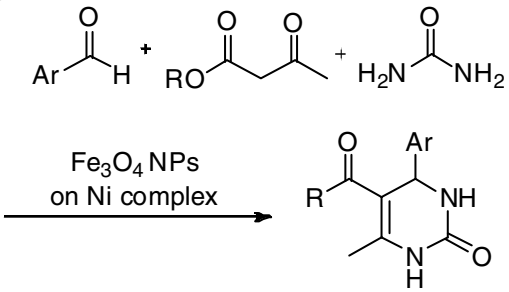
Dihydropyrimidinone cores: The chemistry of dihydropyrimidinones synthesis due to their vast spectroum applications as anti-inflammatory, antiviral, antibacterial, antitumor, antihypertensive agents and calcium channel blockers have emerged abundantly in last decades [14,15,68-73]. Lavanya *et al*. [74] reported an efficient synthesis of dihydropyrimidinones in high yields using nanocrystaline CdS thin film *via* condensation of benzaldehyde, ethyl acetoacetate and urea (or thiourea) in ethanol solvent at reflux condition at 78-80 ºC (**Scheme-VIII**).



Catalyst (g)	Solvent	Time (h)	Yield (%)
0.25	EtOH	10	25
0.5	EtOH	7	78
1	EtOH	4	94

Scheme-VIII

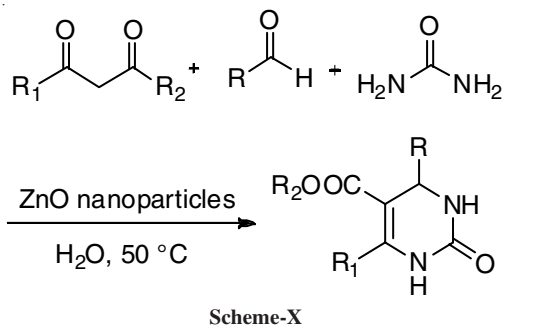
The Biginelli reaction of ethyl acetoacetate, urea and aromatic aldehyde occurred in presence of Fe_3O_4 nanoparticle supported on Ni(II) complexes as an efficient catalyst under solvent free condition in the microwave reactor at 130 ºC provided products the desired in good to high yields in short reaction time (6 min) (**Scheme-IX**) [75].



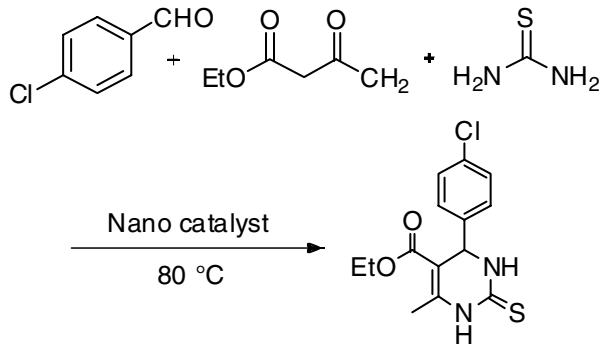
Scheme-IX

A green protocol for Biginelli reaction of benzaldehyde, ethyl acetoacetate and urea was reported by Hassanpour *et al*. [76] in water media at 50 ºC. In the absence of catalyst observed the trace amount of desired product after 28 h, instead when ZnO nanoparticles used as catalysts this reaction gave good yields during 30 min (**Scheme-X**).

An efficient synthesis of 3,4-dihydropyrimidin-2(*1H*)-ones-thiones *via* Biginelli reaction between benzaldehyde, ethylacetate and thiourea in the present of cobalt manganese oxide nanocatalyst at 80 ºC was reported by Karami *et al*. [77] (**Scheme-XI**).



Scheme-X



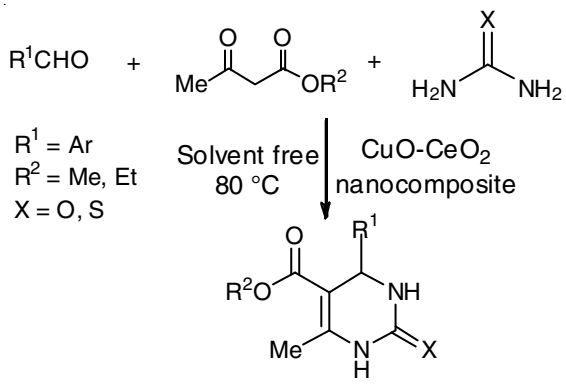
Scheme-XI

The nano-catalyzed reaction of aldehydes, α-ketoesters, urea or thiourea reported by Albadi and Mansournezhad [78]. A varity of 3,4-dihydropyrimidin-2(*1H*)-ones/thiones produced by catalytic activity of CuO-CeO_2 nanocomposite at 80 ºC under solvent free condition. The recyclability of CuO-CeO_2 was investigated for 10 consecutive runs that led to high yields of product after 10 runs (**Scheme-XII**).

Diazepine cores: Diazepine as a key building block of heterocyclic compounds have recognized as anticancer, antiviral, antibiotic and antituberculosis agents in medicinal chemistry [22,24-26,79]. Maleki [80] reported the synthesis of diazepine derivatives by using terminal alkynes, 1,2-diamines and isocyanide in the presence of silica-supported iron oxide nanoparticles at room temperature in a ethanolic media. Using of SiO_2-supported super-paramagnetic iron oxide nanoparticles as an efficient protocol in synthesis of diazepines afforded good to high yield products under benign conditions (**Scheme-XIII**).

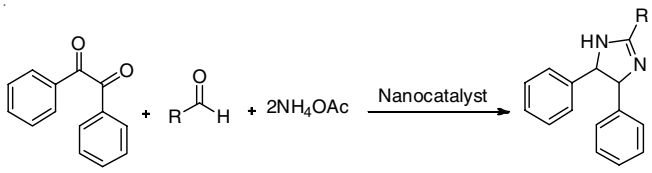
Benzodiazepines scaffold by having biological activities are one of important motifs in medicinal chemistry [81-84]. In 2017, a developing method for synthesis of benzodiazepine reported by Nasir *et al*. [85] using NiO-SiO_2 nanocomposites as catalyst. Benzodiazepine derivatives obtained by condensation of *o*-phenylenediamine, dimedone and aldehydes at 70 ºC in a microwave reactor for 20-25 min (**Scheme-XIV**).

Imidazole cores: Multi-substituted imidazoles are an important class of compounds in the field of pharmaceuticals and exhibit a wide spectrum of biological activities [86-88]. Sengupta *et al*. [89] synthesized some imidazole derivatives by developing of γ-Fe_2O_3@TiO_2EGCu(II) as an effective nanocatalyst. The condensation reaction of benzil, benzaldehyde and ammonium acetate (NH_4OAc) afforded highly substituted imidazoles in the presence of Cu(II) nanocatalyst and aniline as a solvent at 100 ºC (**Scheme-XV**).



Run	Time (min)	Yield (%)
1	10	91
2	10	91
3	10	90
4	10	90
5	12	90
6	12	90
7	15	90
8	15	89
9	18	89
10	20	88

Scheme-XII

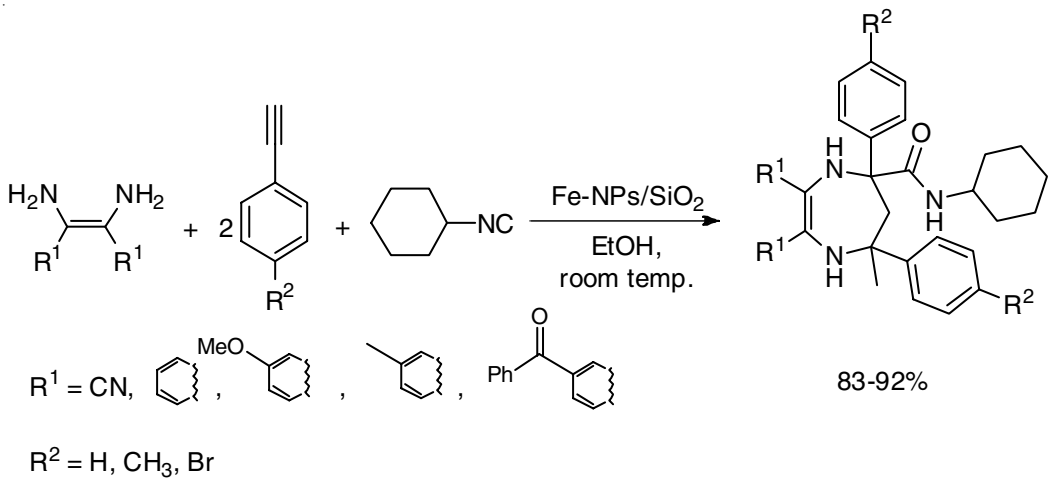


Scheme-XV

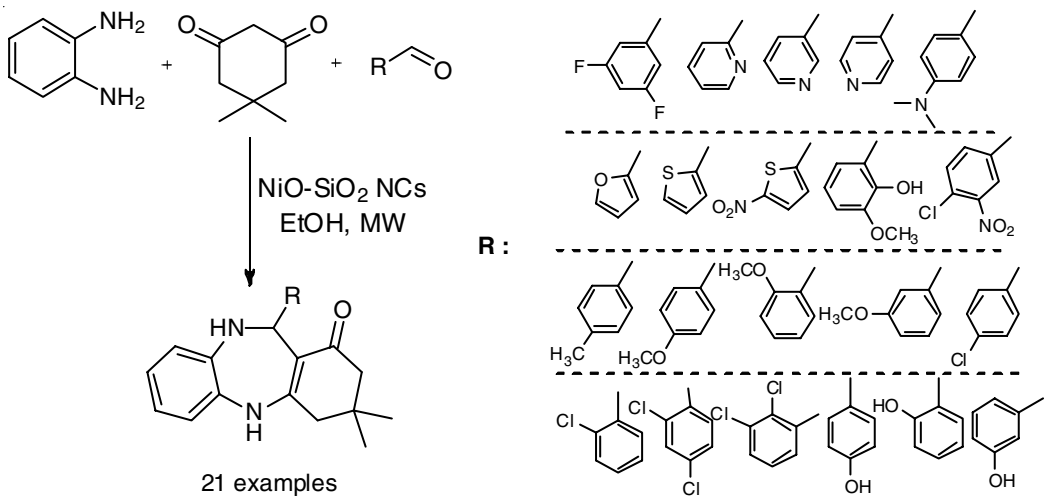
The $ZnFe_2O_4$ nanoparticles have been used as a catalyst for the effectual synthesis of imidazole derivatives through the reaction of benzyl, aromatic aldehydes, ammonium acetate and aliphatic amines under mild and solvent-free conditions [90] (**Scheme-XVI**). The hydrothermal prepared $ZnFe_2O_4$ magnetic nanoparticles as a cost-effective catalyst recovered simply and reused in five runs without loss of activity.

Conclusion

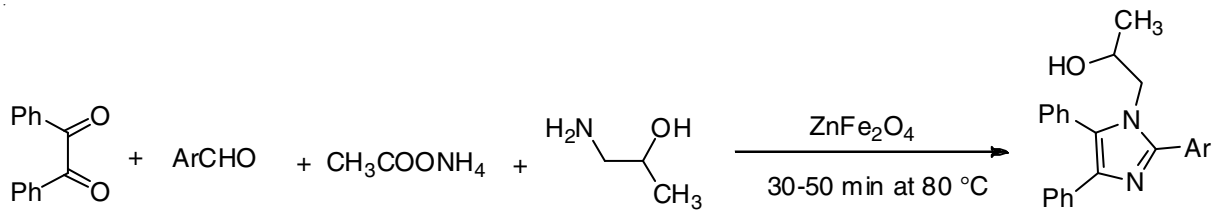
Herein, the syntheses of *N*-containing heterocyclic compounds using advanced nanomaterials as promising potential catalysts have been explored. Several protocols showed numerous advantages compared to conventional methods. They have been occurred in short reaction times, higher yields under green conditions. Metal nanoparticles can be easily separated and reused without much loss in their activities. The environmental



Scheme-XIII



Scheme-XIV



Scheme-XVI

friendly catalysts, which have been explored in the synthesis process are inexpensive and commercially available. In conclusion, the ability of metal nanoparticles in different aspects such as activity, selectivity, stability, simple recovery, recyclability and reusability is promising new roads for researchers in medicinal, pharmaceutical and other related fields of science.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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