

Review Article

Function of Nanocatalyst in Chemistry of Organic Compounds Revolution: An Overview

Kanagarajan Hemalatha,¹ Gunabalan Madhumitha,¹ Amir Kajbafvala,² Narayanan Anupama,¹ Rajesh Sompalle,³ and Selvaraj Mohana Roopan³

¹ *Organic Chemistry Division, School of Advanced Sciences, VIT University, Vellore, Tamil Nadu 632 014, India*

² *Department of Materials Science and Engineering, North Carolina State University, Engineering Building I, Raleigh, NC 27695-7907, USA*

³ *Chemistry Research Laboratory, Organic Chemistry Division, School of Advanced Sciences, VIT University, Vellore, Tamil Nadu 632 014, India*

Correspondence should be addressed to Selvaraj Mohana Roopan; mohanaroopan.s@vit.ac.in

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Heterocyclic motif is an important scaffold which has both industrial and pharmaceutical applications. These motifs can be prepared using wide variety of reaction conditions such as the use of expensive catalyst, toxic solvent, harsh reaction condition like the use of base, high temperature, and multistep reaction. Although various methods are involved, the chemistry arena is now shifted towards the greener way of synthesis. Nanocatalyst constitutes an important role in the green synthesis. This is because the activity of the catalyst resides in the exposed portion of the particles. By decreasing the size of the catalyst, advantages such as more surface area would be exposed to the reactant, only negligible amount would be required to give the significant result and selectivity could be achieved, thereby, eliminating the undesired products. The current review enlists the various types of nanocatalyst involved in the heterocyclic ring formation and also some other important functionalization over the ring.

1. Introduction

The new era of chemistry is shifting towards the path of innovative techniques which mainly concentrates on environmental aspects [1, 2]. Each and every component of the reaction is investigated on the basis of ecofriendly concepts such as use of nonhazardous solvent (water) and solvent-free synthesis or inexpensive catalyst, without affecting the yield and quality of the reaction. Synthesis of heterocyclic core constitutes the important portion of organic synthesis because it has wide variety of pharmacological actions [3–6]. Various methods have been adopted for the synthesis which includes the use of catalyst [7, 8], ultrasound irradiation [9–11], and microwave irradiation [12, 13]. Although these methods have their own advantages, it also possesses certain disadvantages like expensive instruments, inaccessible materials, nonrecyclable and non-selectivity, and so forth. To overcome these, the role of nanocatalyst holds its application [14]. Nanoscience is the cram of phenomenon on a nanometer

range. Atoms are a few tenths of a nanometer in diameter, and molecules are typically a few nanometers in size. The smallest structures humans have been made have dimensions of a few nanometers and the smallest structures we will ever make will have the dimensions of a few nanometers. This is because as soon as a few atoms are placed next to each other, the resulting structure is a few nanometers in size. Chemistry is the study of molecules and their reactions with each other. Since molecules typically have dimensions of a few nanometers, almost all of nanoscience can be reduced to chemistry. Chemistry research in nanotechnology concerns carbon nanotubes, self-assembly, C₆₀ molecules, and structures built using DNA. Sometimes the chemical description of a nanostructure is insufficient to describe its function. Owing to the hasty progress of nanoscience and nanotechnology, the primeval colloid science is given a new life. Because of their great differences from single molecules and bulk materials, nanoscale materials, including colloids, have attracted much attention since the last decade, especially

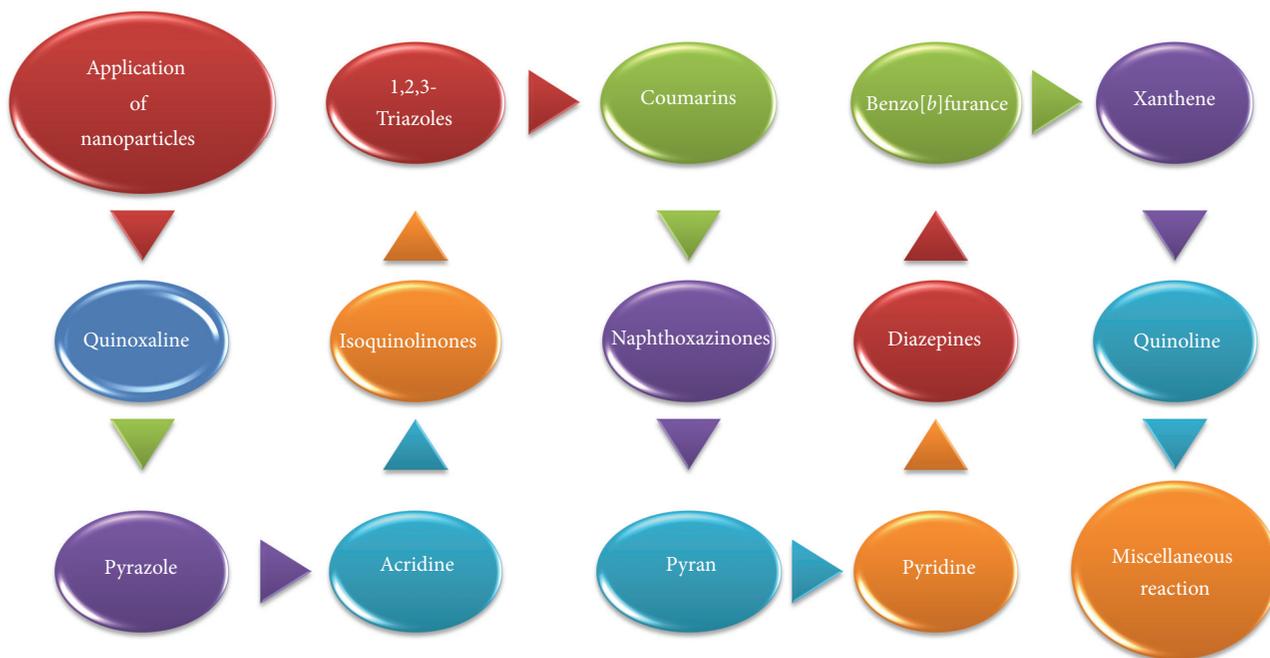


FIGURE 1: Application of nanoparticles in organic synthesis.

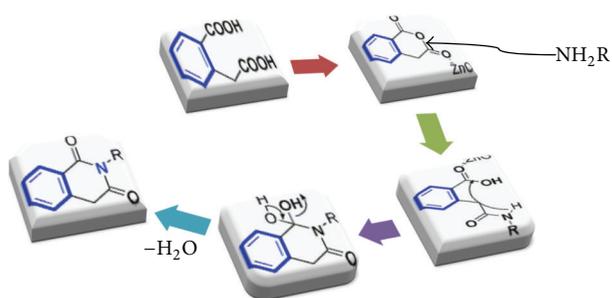


FIGURE 2: Possible mechanism for the formation of *N*-arylmophthalimide, 2.4.2.

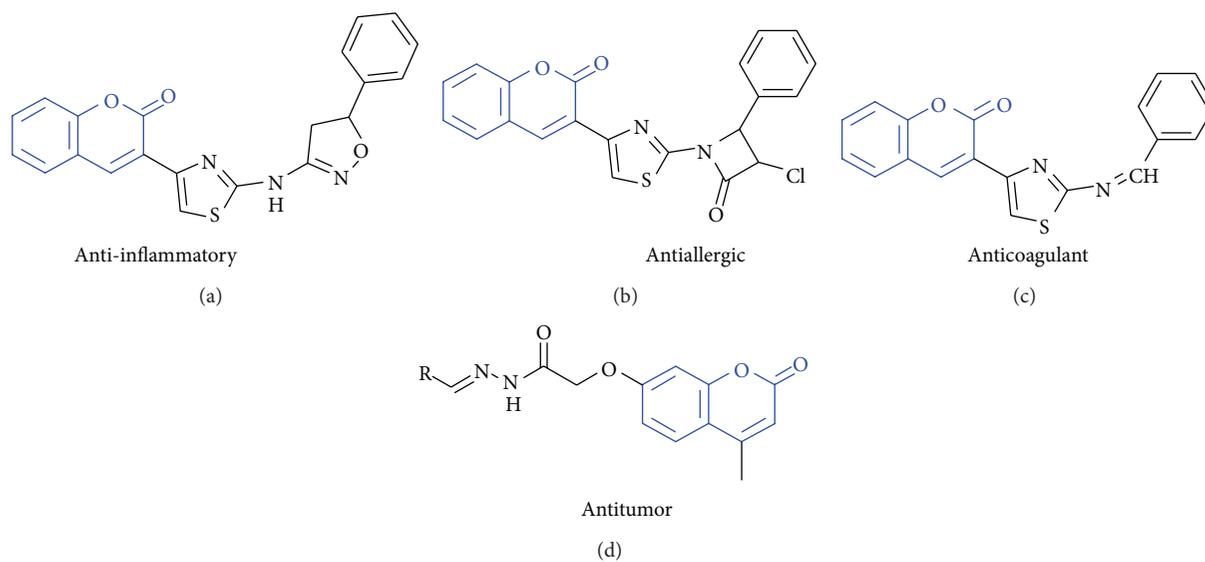


FIGURE 3: Pharmacological activities of coumarins.

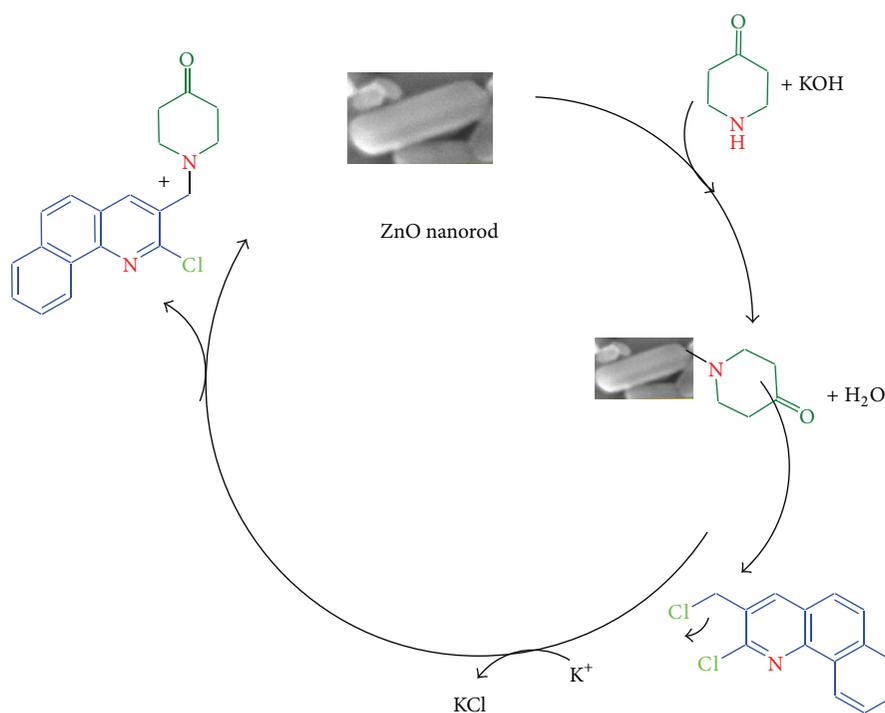


FIGURE 4: Role of ZnO nanorods in synthesis of *N*-alkylated products (adapted from [1]).

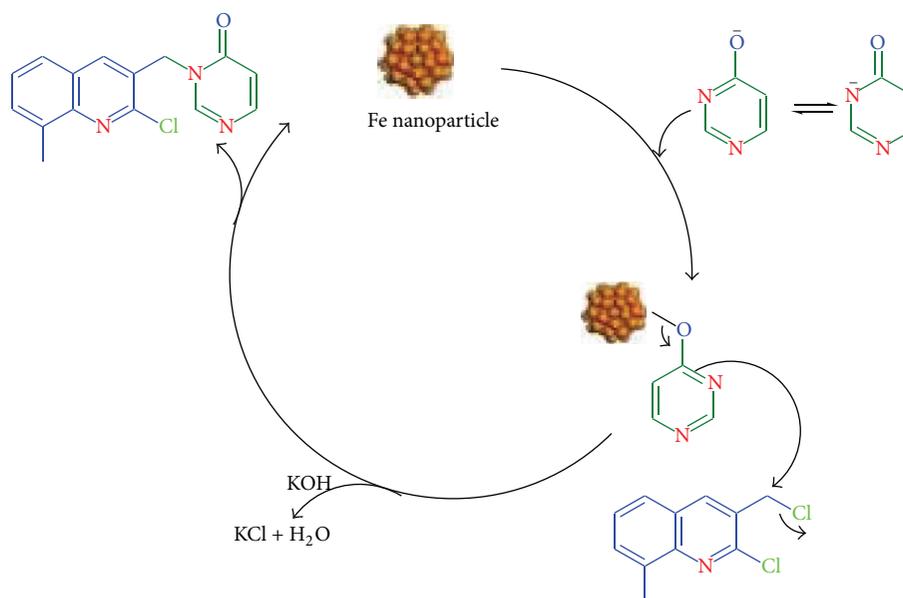


FIGURE 5: Role of Fe nanoparticles in *N*-alkylation reaction (adapted from [2]).

in the field of catalysis. Over the several past decades, catalysts and catalytic reactions have attracted considerable attention with the aim of finding meaningful applications in the pharmaceutical and fine chemical industries. The nanocatalysts are highly selective, reactive, and stable; thereby it supersedes the conventional catalyst. Nanoparticles with a diameter of less than 10 nm have generated intense interest over the past decade due to their high potential applications in areas such

as sensors, nanoscale electronics, catalysis, and optics. The catalytic activity of nanoparticles is affected by size; therefore, the relative ratio of surface atom types changes dramatically with varying particle size. In many cases, the activity increases as the particle size decreases due to favorable changes in the electronic properties of surface atoms, which are located mainly on edges and corners in small particles. On the other hand, the reactivity and selectivity of metal nanocatalysts

also depend strongly on the different crystallographic planes present on the nanoparticles and which can be achieved by controlling the morphology of these nanoparticles. Size and surface of the nanocatalyst play a major role because it is the reason for its selectivity and reactivity. Also, in some cases the enhancement by doping and surface chemical modifications would be done to increase its performance [15]. Nanocatalyst is not only used in organic transformation but also it has various applications [16, 17]. These nanocatalysts can be prepared by various methods such as thermal decomposition, micro-arc oxidation irradiation, chemical vapor synthesis, non-sono and sonoelectrooxidation, sol-gel technique, chemical precipitation, photochemical method, hydrothermal method, antisolvent precipitation, glow discharge plasma electrolysis, wet-chemical method, microwave irradiation, and sonochemical method [16–21]. The size and nature of nanocatalyst varies on the type of method used for preparation [22–27]. Based on the requirement, the method of preparation can be selected. In this paper, we will review recent examples of nanoparticles used in organic transformation such as quinoxaline, naphthoxazinones, coumarins, 1,2,3-triazoles, acridine, pyrazole, and isoquinolinones. (Figure 1). The heart and soul of this paper is Section 2.23, where we cover miscellaneous functionalization on heterocycles; this is a challenge for nanocatalyst researchers to engage.

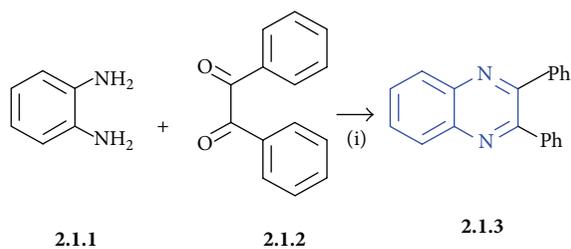
2. Application of Nanoparticles in Organic Synthesis

2.1. Synthesis of Quinoxaline Analogues. Quinoxaline is an important chemical entity which has interesting biological properties such as trypanocidal property [28], antimycobacterial agent [29], and cytotoxic agent [30]. The synthesis of quinoxalines (Scheme 1) was carried out by oxidative coupling of 1,2-diamines, **2.1.1** and 1,2-dicarbonyl compounds, **2.1.2** using gold nanoparticles supported on nanoparticulated ceria (Au/CeO) or hydrotalcite (Au/HT) as catalysts and air as an oxidant. The use of nanoparticles led to the mild reaction conditions such as base-free reactions, using mild temperature and air as an oxidant. The catalyst could be reused only with a little loss in activity [31]. The use of inexpensive and recyclable SiO₂ which has highly reactive –OH group on its surface has its application in the synthesis of quinoxaline, and it produces high yield in less reaction time. Because of its reusable nature, it supersedes the other catalyst [32]. Quinoxalines can also be synthesized by advantageous nano-BF₃·SiO₂ and nano-TiO₂ catalyst systems. The reaction was carried out at varied temperatures and different moles of reactants to optimize the reaction condition and concluded that solvent-free conditions at room temperature could be the optimal one. In addition, the report concluded that the reaction time could be reduced by performing the reaction under sonication [33]. In nano-TiO₂ system, the same authors carried out the synthesis in the presence of nano-TiO₂ and compared with bulk TiO₂ and other applied catalysts. The satisfactory results were obtained in solvent-free condition at room temperature using 12 mol % as a catalyst [34]. Lü and coworkers synthesized quinoxalines using magnetic Fe₃O₄ nanoparticles. The result shows that the reaction could be

performed well in water using 10% Fe₃O₄ nanoparticles as catalyst at room temperature, and the catalyst can be recovered easily by using external magnet and reused with consistent activity [35]. Polyaniline/SiO₂ nanocomposite material was prepared and it was used as a catalyst for the synthesis of quinoxalines. They reported that 10% catalyst was found to be optimal for the reactant transformation, and the catalyst activity was found to be consistent even after three runs [36]. Another popular method to synthesize quinoxalines is by using TiO₂ nanoparticles as a catalyst. The optimal protocol system was found out by using dichloroethane as an efficient solvent with 2.5 mol % catalyst to give the highest yield. Also *o*-phenylenediamine with electron-withdrawing group gave the higher rates and yield than the electron-donating groups [37]. The quantitative yield of quinoxaline was obtained in 10 minutes by using acetonitrile solvent system, 10 mol % of Ni-nanoparticles as catalyst at 25°C stirred under N₂ atmosphere [38]. Bardajee and coworkers prepared SBA-15 supported on Pd (II) Schiff-base complex nanocatalyst for the synthesis of 2, 3-disubstituted quinoxalines derivatives [39].

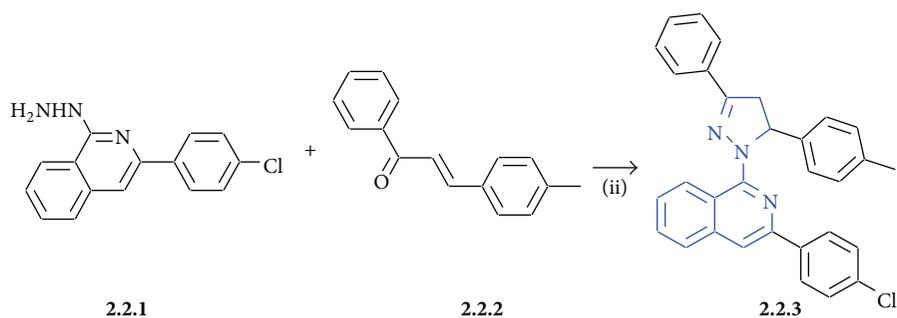
2.2. Synthesis of Pyrazole Analogues. Pyrazole is an important novelty which has reported insecticidal [40], antimalarial [41], anti-inflammatory, and antimicrobial [42] activities. Khan and coworkers synthesized 1-(4,5-dihydropyrazol-1-yl) isoquinolines, **2.2.3** from chalcones, **2.2.2** and 1-hydrazinylisoquinoline, and **2.2.1** using iron-oxide nanoparticles and this method of synthesis eliminates the autooxidation of the desired pyrazolines to the corresponding pyrazoles (Scheme 2) [43]. (α-Fe₂O₃)-MCM-41 catalysts is impregnated with 10% of iron oxide nanoparticles as a recoverable and reusable catalyst (Scheme 3) for the synthesis of pyrazolo [3,4-*c*] pyridine, **2.2.6** derivatives from 3,5-dibenzylidenepiperidin-4-one, **2.2.4** and hydrazine derivatives, **2.2.5a–c**. The optimum amount of catalyst was found to be 0.015 g, and further increase in the amount of catalyst has no effect on rate of the reaction and yield. Even though pure MCM-41, amino-functionalized MCM-41, and Fe₃O₄ produced satisfactory result, the ease of recoverability and reusability of (α-Fe₂O₃)-MCM-41 made it to prefer this catalyst for the pyrazole analogue synthesis [44].

2.3. Synthesis of Acridine Analogues. Acridine, one of the important nitrogen heterocycle, shows activity such as anti-herpes [45], antimalarial and antitumor [46], and larvicidal action [47]. Roopan and Khan performed an ecofriendly synthesis of 9-chloro-6, 13-dihydro-7-phenyl-5H-indolo [3, 2-*c*]-acridines, **2.3.3** by Friedlander condensation (Scheme 4) of 2-amino-5-chlorobenzophenone, **2.3.1** and 3,4-dihydro-2H-carbazol-1(9H)-one, and **2.3.2** in the presence and absence of SnO₂ nanoparticles under microwave irradiation. The reaction was not initiated in the absence of catalyst [48]. A comparative study of various nanoparticles (Mn₃O₄, CuO, CaO, MgO, and Fe₃O₄), optimization of solvent, and temperature showed that nano-Fe₃O₄ (10 mol %) + solvent-free conditions + 120°C as an efficient protocol for the synthesis (Scheme 5) of 1,8-dioxo-decahydroacridines, **2.3.6**, from aldehyde, **2.3.5**, dimedone, **2.3.4**, and aromatic amine



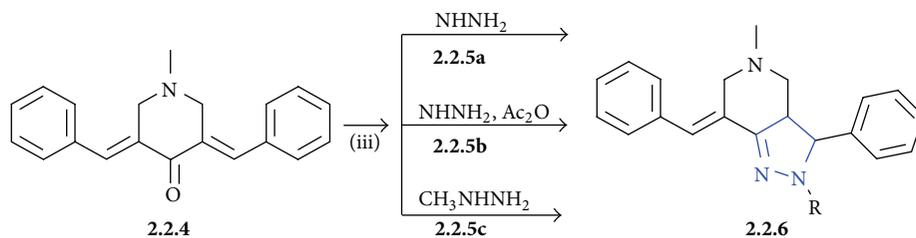
(i) Au, SiO₂, BF₃-SiO₂, Fe₃O₄, polyaniline/SiO₂, TiO₂, Ni, Pd/SBA-15

SCHEME 1: Synthesis of quinoxaline analogues, 2.1.3.



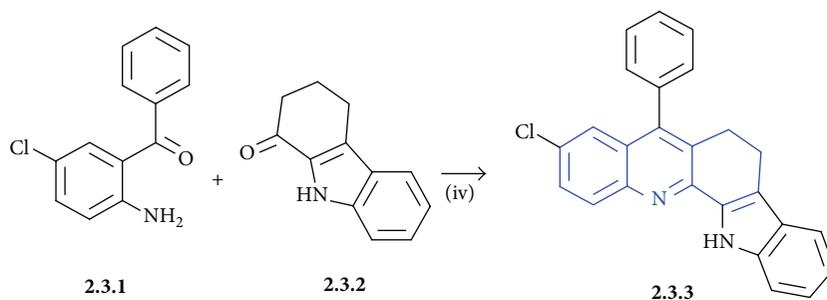
(ii) Iron oxide, ethanol, 30 min, reflux

SCHEME 2: Synthesis of pyrazole analogues, 2.2.3.



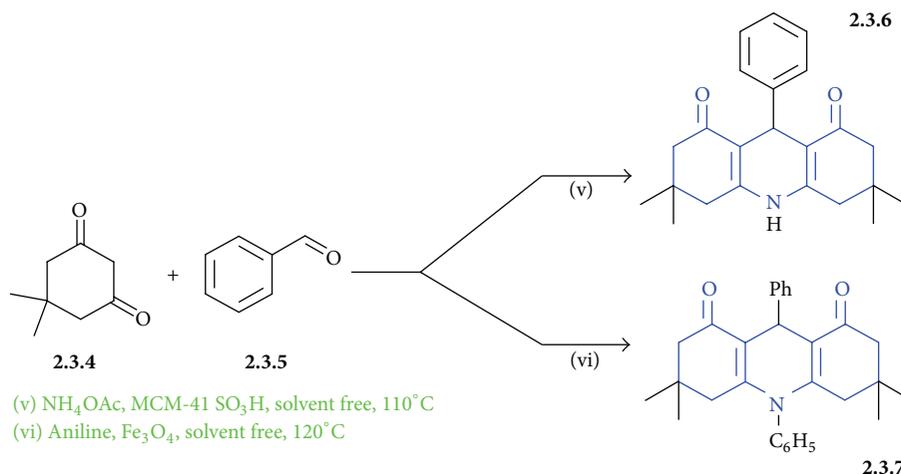
(iii) (α -Fe₂O₃)-MCM-41-SO₃H, ethanol, 30 min, rt

SCHEME 3: Synthesis of pyrazole analogues, 2.2.6 using dibenzylidenepiperidin-4-one.

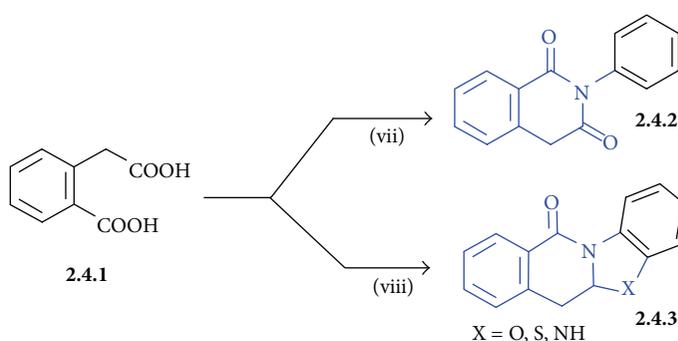


(iv) SnO₂ nanoparticles, H₂SO₄, 500 W, MW

SCHEME 4: Synthesis of acridine analogues, 2.3.3 using 2-amino-5-chlorobenzophenone.



SCHEME 5: Multicomponent strategy to synthesis acridine analogues, 2.3.6-2.3.7.



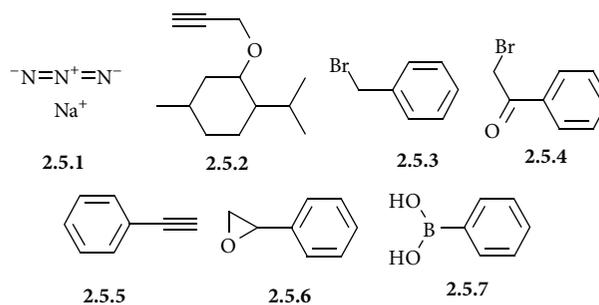
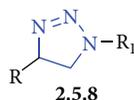
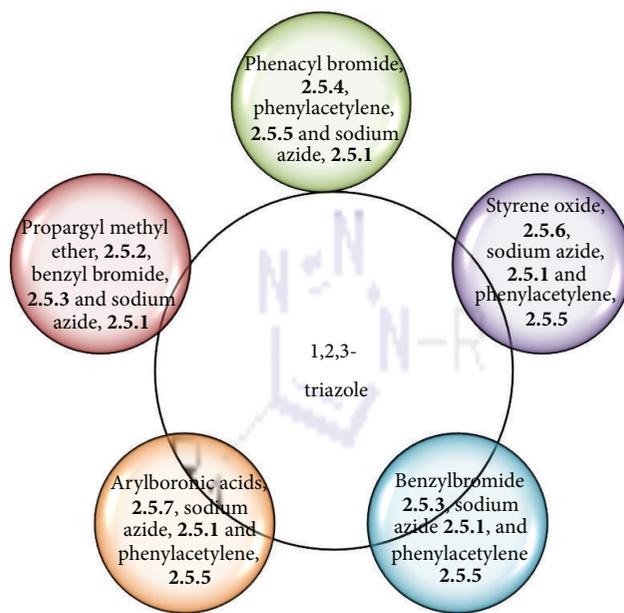
SCHEME 6: Synthesis of *N*-arylhomophthalimides, 2.4.4 and Isoquinolinones, 2.4.5.

in the presence of ammonium acetate [49]. 0.005 g of MCM-41- SO_3H , 110°C and solvent-free condition is another effective combination for obtaining 1, 8-dioxo-9-aryl decahydroacridines nucleus, 2.3.7 [50].

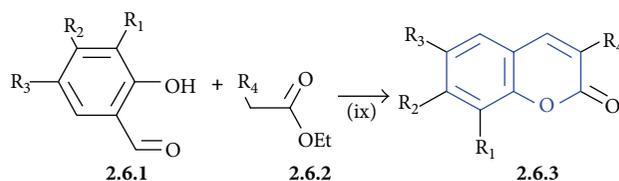
2.4. Synthesis of *N*-Arylhomophthalimides and Benzannelated Isoquinolinones. Isoquinolinones are reported to cause allosteric modulation of metabotropic glutamate receptor 2 [51], and also it has JNK inhibitory action [52]. An emerald procedure was developed utilizing an efficient catalyst; that is, ZnO nanoparticles mediated the synthesis of *N*-arylhomophthalimides and benzannelated isoquinolinones. Krishnakumar and coworkers synthesized flower-shaped ZnO nanoparticles and used them in the reaction between homophthalic acid, 2.4.1, and substituted anilines, benzyl amine, for the ecofriendly synthesis of *N*-arylhomophthalimide 2.4.2, and benzannelated isoquinolinones, 2.4.3 (Scheme 6). They carried out the reaction using various catalysts, solvents, and different concentration of the catalyst. Nano ZnO at a concentration of 5% mol in the toluene system was found to be effective [53]. The ZnO nanoparticles exhibit

admirable catalytic action, and the proposed methodology was capable of providing the desired products in good yield and purity. The possible mechanism for the formation of this product is illustrated in Figure 2.

2.5. Synthesis of 1,4-Disubstituted 1,2,3-Triazoles. 1,2,3-triazoles (Scheme 7), an important entity, has reported antimicrobial activity [54], anti-HSV-1 activity [55], and antifungal activity [56]. This moiety can be synthesized by an innovative concept called “click chemistry.” It is a reaction between sodium azide, 2.5.1, and alkyne to give 1,4-disubstituted 1,2,3-triazoles in the presence of copper and water. This reaction is also known as Cu-catalyzed alkyne azide cycloaddition. Since copper acts as a catalyst in the synthesis of triazoles, copper supported on various materials can be made into nanoparticles, and its yield and specificity can be increased further. Alonso and coworkers synthesized 1,2,3-triazoles through various heterocycles derived from natural product such as (–)-menthol, lactic acid, D-glucose, oestrone, and cholesterol converting them into alkynes by introducing propargyl groups. Propargyl methyl ether,



SCHEME 7: Synthesis of 1,2,3-triazoles, 2.5.8 using various strategy.



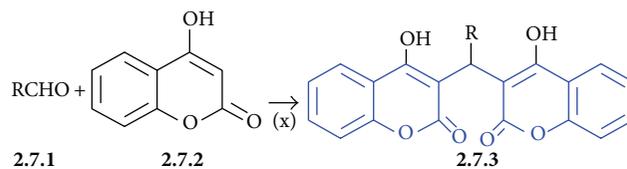
(ix) ZnO nanoparticles, MW

R₁ = H, OH, OMeR₂ = H, OH, OMe, Et₂NR₃ = H, Cl, Br, NO₂, OHR₄ = CO₂Et, COMe, CN

SCHEME 8: Synthesis of coumarins, 2.6.3.

2.5.2, benzyl bromide, 2.5.3, and sodium azide, 2.5.1, under click reaction condition to give triazole derivatives. These derivatives of natural products having wide variety of application were obtained in high yield using copper nanoparticles [57]. CuI supported on poly(4-vinylpyridine) [P₄VPy-CuI] acts as a heterogenous catalyst for the synthesis of triazoles. Using the optimized ratio of 1 : 1 : 1.1 of phenacyl

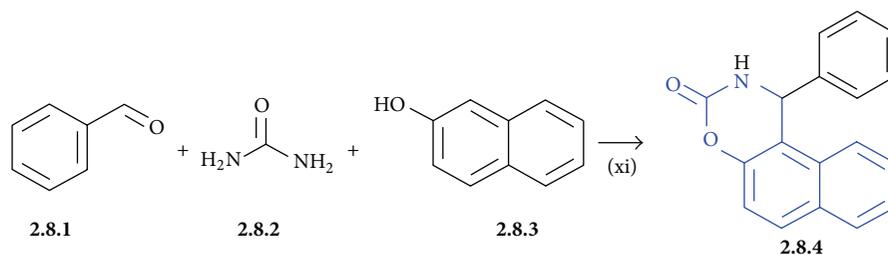
bromide, 2.5.4, phenyl acetylene, 2.5.5 and sodium azide, 2.5.1, 0.1g of P₄VPy-CuI and water, required triazoles were obtained after refluxing. Also, this catalyst can be reused up to 8 runs without losing its efficiency [58]. Metalloanthraquinone complex, an important catalyst for the synthesis of 1,4-disubstituted 1,2,3-triazole, was prepared, and various reaction conditions were studied. Various metal



(x) PVP-Ni, ethylene glycol, rt

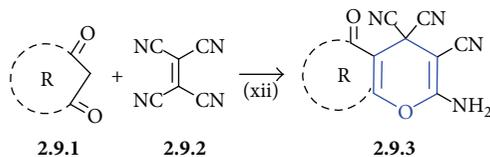
R = 4-ClC₆H₄, 4-NO₂C₆H₄, 4 and 2-OHC₆H₄, 3,4(CH₃O)₂C₆H₃, piperonyl

SCHEME 9: Synthesis of biscoumarins, 2.7.3.



(xi) Cu nanoparticles, K₂CO₃, PEG-400

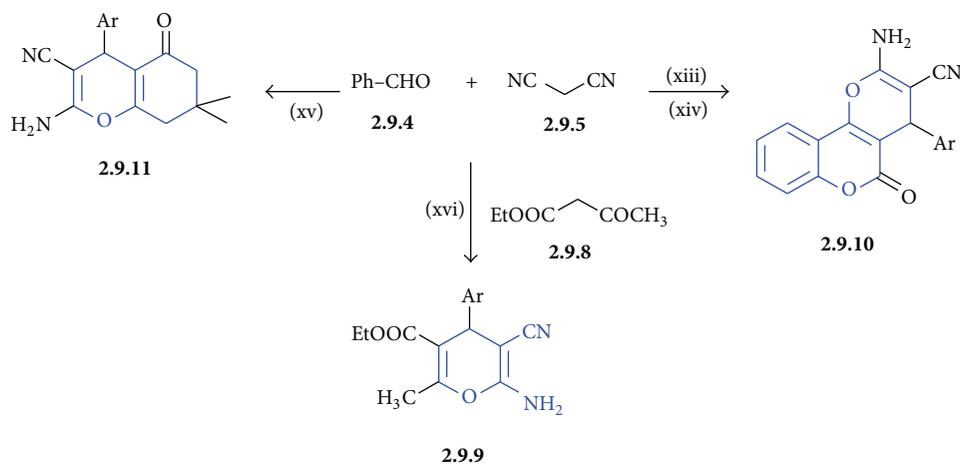
SCHEME 10: Synthesis of naphthoxazinones, 2.8.4.



R = Alicyclic/heterocyclic

(xii) TiO₂ NPs, H₂O, RT

SCHEME 11: Synthesis of pyran analogues, 2.9.3.



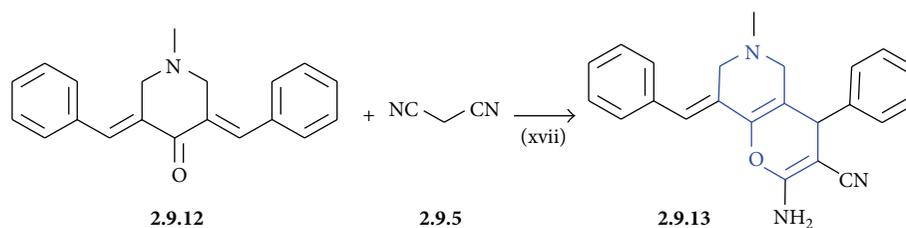
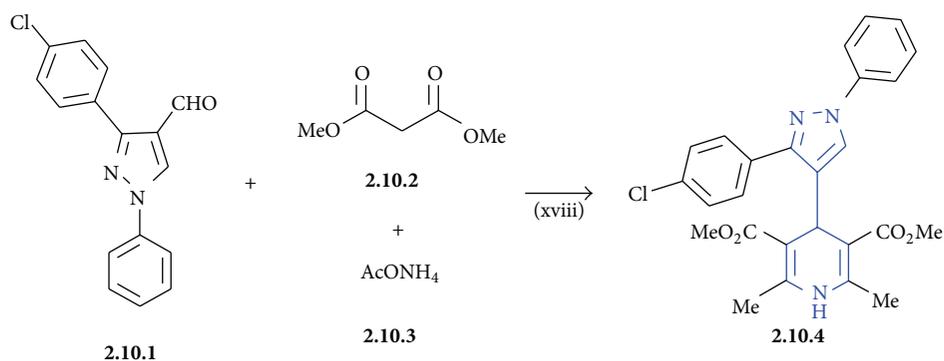
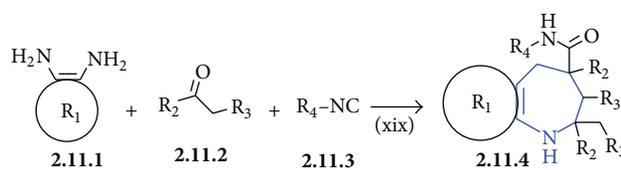
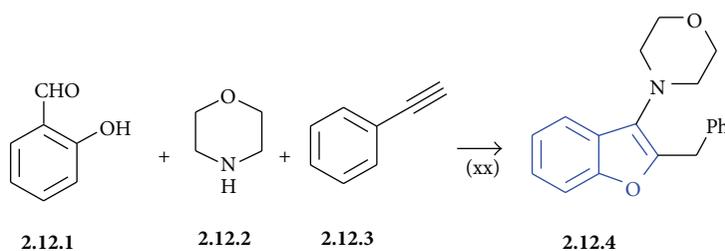
(xv) Dimethylcyclohexane-1,3-dione, 2.9.6, Fe₂O₃

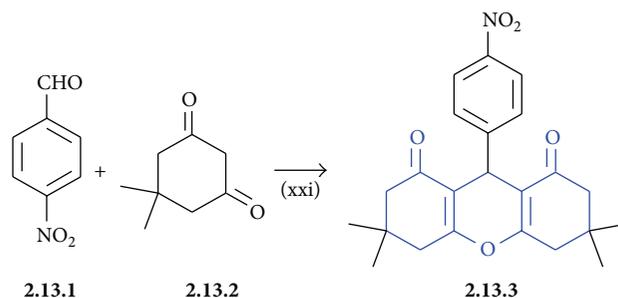
(xvi) ZnO nanoparticles

(xiii) 4-Hydroxy-2H-chromen-2-one, 2.9.7 CuO

(xiv) 4-Hydroxy-2H-chromen-2-one, 2.9.7 Fe₂O₃

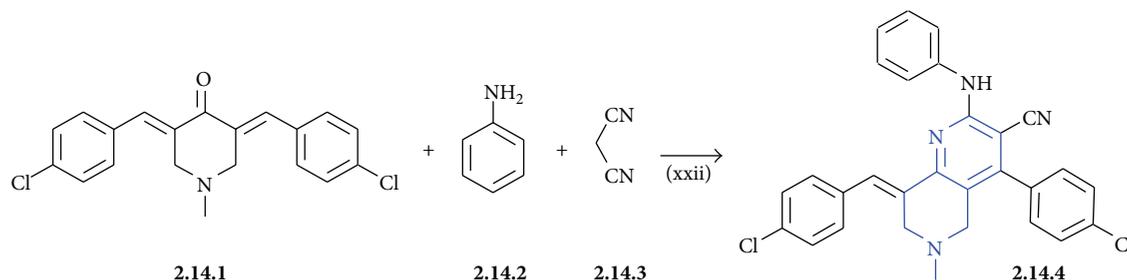
SCHEME 12: Multicomponent reaction for the construction of pyran analogues, 2.9.9–2.9.11.

(xvii) MCM-41-SO₃H, solvent freeSCHEME 13: Synthesis of pyrano pyridine, **2.9.13**.(xviii) MgO nanotube, CH₃CN, refluxSCHEME 14: Synthesis of 1,4-dihydropyridine derivatives, **2.10.4**.R₁ = CN, Ar-Cl₂, ArNO₂, Ar-COOHR₂ and R₃ = acetone, cyclohexanone, acetophenoneR₄ = aliphatic, alicyclic, aromatic(xix) Fe₃O₄/SiO₂, EtOH, rt., 3–6 h, refluxSCHEME 15: Synthesis of diazepines, **2.11.4**.(xx) CuI nanoparticles, K₂CO₃, reflux, 1.5 hSCHEME 16: Synthesis of benzo[*b*]furans, **2.12.4**.



(xxi) MCM-41-SO₃H, H₂O, sonication

SCHEME 17: Synthesis of 1,8-dioxo-octahydroxanthenes, **2.13.3**.



(xxii) α -Fe₂O₃-MCM-41-SO₃H, solvent free, 120°C

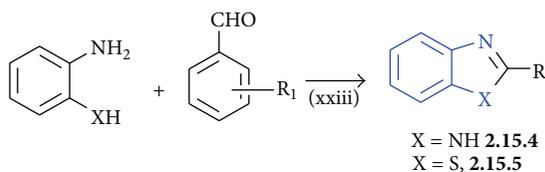
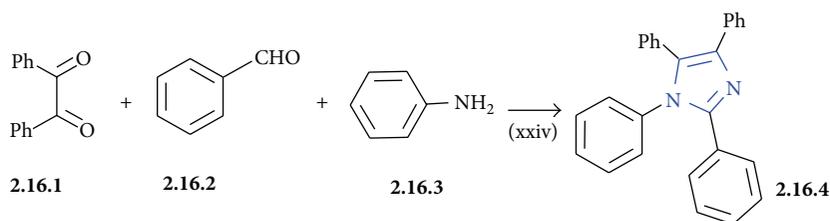
SCHEME 18: Synthesis of 1,6-naphthyridine analogues, **2.14.4**.

ligands complexes were tested, but only copper was found to be catalytically active due to the richness of electron on metal. Water was found to be an effective solvent, and also the amount of water is also an important criterion. The optimum amount of water required was found to be 5 mL for the reaction between styrene oxide, **2.5.6**, sodium azide, **2.5.1** and phenyl acetylene, **2.5.5** [59]. Another environment friendly synthesis of triazoles was the cyclisation reaction between three components benzyl bromide **2.5.3**, sodium azide **2.5.1**, and phenyl acetylene **2.5.5** in the presence of magnetically separable CuFe₂O₄ nanoparticles, water at 70°C. The catalyst can be separated easily and reused effectively [60]. In an alternative method, various copper salts [CuI, CuSO₄, CuCl₂, Cu(NO₃)₂, Cu₂- β -CD complex] were used for the synthesis of 1,2,3-triazoles of phenyl boronic acid from coupling of aryl boronic acids, **2.5.7**, sodium azide, **2.5.1** and phenyl acetylene, **2.5.5**. Among these copper catalyst Cu₂- β -CD complex gave excellent yield of 1,2,3-triazole, **2.5.8** without adding any additives [61].

2.6. Synthesis of Coumarins. Coumarins are attractive molecule in chemistry with anti-inflammatory activity [62], antioxidant and lipoxygenase inhibitory activity [63], and antifungal activity [64]. Coumarin has been used as an aroma enhancer in pipe tobaccos and alcoholic drinks although in general it is banned as a flavorant food additive, due to concerns about coumarin's hepatotoxicity in animal models. The synthesis of coumarins and its analogues has attracted

extensive thought from organic and medicinal chemists for many years as a large number of natural products contains this heterocyclic nucleus. Moreover, coumarins have various pharmacological activities (Figure 3). Knoevenagel condensation is one of the widely used reactions for the synthesis of coumarins (Scheme 8). Since it involves the use of acids and bases, an alternative approach for carrying out the condensation is essential. The reaction between *o*-hydroxy benzaldehyde, **2.6.1**, and 1,3-dicarbonyl compounds, **2.6.2**, is an effective reaction for the formation of coumarins, **2.6.3**. ZnO nanoparticles were found to be an effective alternative in 10% mol concentration. Increase or decrease in the concentration of the ZnO extends the time taken for the reaction with fewer yields [65].

2.7. Synthesis of Biscoumarins. Transition metal nanoparticles have gained tremendous importance due to their interesting electrical, optical, magnetic, chemical properties, and especially catalytic properties, which cannot be achieved by their bulk counterparts. Recently, there has been growing interest in using nickel nanoparticles in organic synthesis owing to their easy preparation, potent catalytic activity, possible process ability, and high stability. Heterocyclic systems are common structural motifs in many biologically active substances and natural products and therefore warrant the design of newer and efficient protocols for their synthesis. In view of this biscoumarins is an important molecule which possesses anticoagulant activity (Scheme 9) [66]. Khurana

SCHEME 19: Synthesis of benzimidazoles, **2.15.4** and benzothiazoles, **2.15.5**.SCHEME 20: Synthesis of imidazoles, **2.16.5** and **2.16.6**.

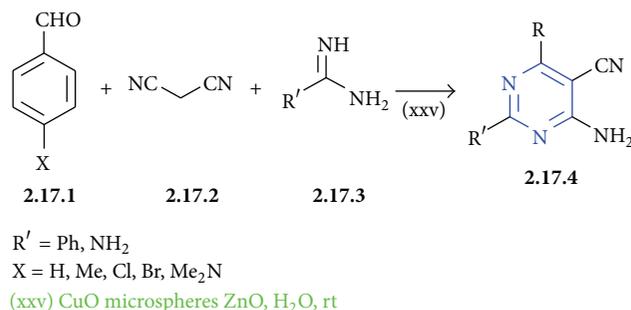
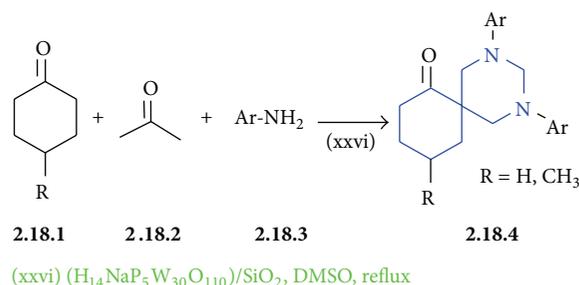
and Vij performed the synthesis of biscoumarins, **2.7.3**, via Knoevenagel condensation followed by rapid Michael addition using polyvinyl pyrrolidone-(PVP)-stabilized nickel nanoparticles for the reactions of aldehydes, **2.7.1**, with 4-hydroxycoumarin, **2.7.2**, in ethylene glycol at room temperature [67].

2.8. Synthesis of Naphthoxazinones. Naphthoxazine, an important motif in heterocyclic chemistry, has reported cytotoxic and antifungal activities [68]. An efficient protocol for the synthesis of 2-naphthol-condensed 1,3-oxazinone, **2.8.4**, (Scheme 10) by the reaction between benzaldehyde, **2.8.1**, urea, **2.8.2**, and β -naphthol, **2.8.3**, was carried out in the presence of K_2CO_3 and copper nanoparticles stabilized by PEG-400. In the absence of Cu, the reaction was not initiated. When the same reaction was carried out without PEG-400, the yield was only 30%. Due to various drawbacks of results with the solvents such as DMSO, acetonitrile, ethanol, THF, and ethylene glycol, the ideal solvent for the synthesis of naphthoxazinones was found to be PEG-400. Not does only it act as a solvent, but also it provides stability to Cu nanoparticles [69].

2.9. Synthesis of Pyran Analogues. Pyran has reported activities such as molluscicidal activity [70] and anthelmintic activity [71]. The synthesis of pyran-annulated heterocyclic systems, **2.9.3** (Scheme 11) can be carried out from various alicyclic/heterocyclic 1,3-dione, **2.9.1**, and tetracyanoethylene, **2.9.2**, using ecofriendly TiO_2 and TiO_2 nanoparticles as a catalyst with high yield. Because of the heterogenous nature of TiO_2 , the work-up process will be carried out easily, and the catalyst can be recovered without any difficulty

[72]. The three-component reaction between aromatic aldehyde, **2.9.4**, malononitrile, **2.9.5**, 4-hydroxycoumarin, **2.9.7**, and CuO nanoparticles (15% mol) in 10 mL water is an effective protocol for the synthesis of 3,4-dihydropyrano[*c*]chromenes, **2.9.10**, (Scheme 12). The same reaction was carried out in the presence of MgO, ZnO, and NiO, but the reaction in the presence of CuO was proved to be best in yield [73]. Khoobi and coworkers carried out the synthesis of 4H-benzopyrans, **2.9.11**, and 2-amino-5-oxo-4-aryl-4,5-dihydropyrano[3,2*c*]chromene-3-carbonitriles, **2.9.10**, (Scheme 12) using the new concept of magnetically inorganic-organic hybrid nanocatalyst, hydroxyapatite-encapsulated Fe_2O_3 [74]. A new way of synthesizing 4H-pyrans, **2.9.9**, was carried out in ionic liquid using ZnO/MgO solid sample containing ZnO nanoparticles as an innovative catalyst [75]. The α - Fe_2O_3 nanopowder was prepared by combustion method and it was used in the synthesis of 3,4-dihydropyrano[*c*]chromenes, **2.9.10** [76]. MCM-41- SO_3H has functional groups which forms bonding with 3,5-dibenzyl idenepiperidin-4-one, **2.9.12**, and the reactions are initiated inside the nanoreactor along with malononitrile, **2.9.5**. The rate of the reaction is increased in the compound, **2.9.12**, with electron-withdrawing group and decreases with electron-donating group. The combination of nanosized MCM-41- SO_3H and solvent-free atmosphere for the ecofriendly synthesis of pyrano [3,2-*c*] pyridine derivatives, **2.9.13**, (Scheme 13) [77].

2.10. Synthesis of 1,4-Dihydropyridine Derivatives. 1,4-dihydropyridine possesses activity such as calcium channel antagonist activity [78] and antioxidant activity [79]. Synthesis of pyrazolyl 1,4-dihydropyridines (Scheme 14), **2.10.4**, was

SCHEME 21: Synthesis of pyrimidone carbonitriles, **2.17.4**.SCHEME 22: Synthesis of spirohexapyrimidines, **2.18.4**.

carried out by multicomponent reaction between pyrazolyl-4-carbaldehyde, **2.10.1**, acetoacetic ester, **2.10.2**, ammonium acetate, **2.10.3**, using 15% MgO nanotube in the presence of acetonitrile. Even though the reaction was carried in various solvents, usage of acetonitrile-made MgO nanotube gave the expected product in high yield [80].

2.11. Synthesis of Diazepines. The development of new approaches for the construction of number of heterocycle continues to be essential for accessing natural products and their structural analogues. Among them, 1H-1,4-diazepines derivatives scaffolds over the years have gained an ongoing interest for biological activities as antileukemic, antiviral, antiplatelet, anticancer, anticonvulsant, psychotropics, and herbicidal [81, 82]. Maleki synthesized one-pot multicomponent synthesis of diazepine derivatives, **2.11.4**, (Scheme 15) from readily available 1,2-diamine, **2.11.1**, a linear or cyclic ketone, **2.11.2**, and an isocyanide, **2.11.3**, using magnetically recoverable Fe₃O₄/SiO₂ nanocatalyst [83].

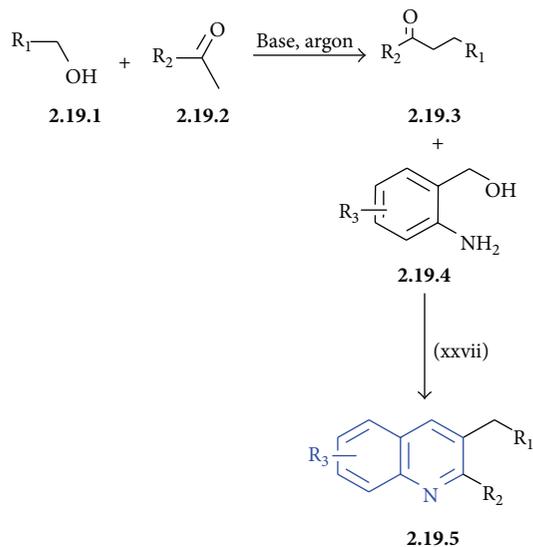
2.12. Synthesis of Benzo[b]Furans. Furan ring possesses some important activity such as cytotoxic activity [84] and antibacterial activity [85]. An ecofriendly multicomponent synthesis of benzo[b]furans (Scheme 16) was carried by the condensation reaction between salicylaldehyde, **2.12.1**, morpholine, **2.12.2**, and phenyl acetylene **2.12.3**, using copper iodide nanoparticles as a specific catalyst. The reaction was standardized with various aldehydes, amines, and acetylenes. The result concluded that salicylaldehyde with electron-

withdrawing groups, aromatic alkynes and aliphatic amines, gave the desired benzo[b]furans [86].

2.13. Synthesis of 1,8-Dioxo-Octahydroxanthenes. Octahydroxanthenes act as anticancer agents [87]. A classical method for synthesis of 1,8-dioxo-octahydroxanthenes, **2.13.3**, (Scheme 17) was the condensation reaction between 4-nitrobenzaldehyde, **2.13.1**, and dimedone, **2.13.2**, using a combination of ultrasound irradiation and nanosized MCM41-SO₃H catalyst which leads to increase in the rate of the reaction and yield [88].

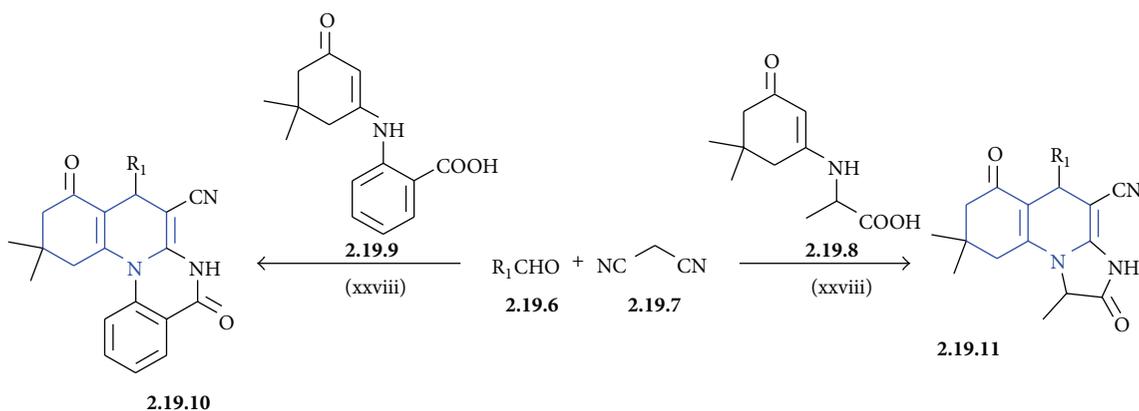
2.14. Synthesis of 1,6-Naphthyridine Analogues. Naphthyridine derivatives are reported with antitumour activity [89] and antimicrobial activity [90]. The reactants such as 3,5-bis(4 chlorobenzylidene)-1-methylpiperidin-4-one, **2.14.1**, aniline, **2.14.2**, and malononitrile, **2.14.3**, are mixed together in solvent-free condition. A novel magnetic (α -Fe₂O₃)-MCM-41-SO₃H acts as a nanocatalyst which could be reused even after 5 runs without decrease in activity. This acts as an efficient catalyst for the synthesis of *N*-aryl-2 amino-1,6-naphthyridine derivatives, **2.14.4**, (Scheme 18) [91].

2.15. Synthesis of Benzimidazoles and Benzothiazoles. The mixture of *o*-phenylenediamine, **2.15.1**, aminothiophenol **2.15.2**, and aromatic aldehydes, **2.15.3**, in water was stirred at 90°C using prepared CdS and manganese-doped CdS nanoparticles for the chemoselective synthesis of benzimidazoles, **2.15.4**, (Scheme 19) and benzothiazoles, **2.15.5**.



$\text{R}_1 = \text{R}_2 = \text{Ph, 2/3/4-MeC}_6\text{H}_4, \text{4-OMeC}_6\text{H}_4, \text{4-CF}_3\text{C}_6\text{H}_4$
 $\text{R}_3 = \text{Me}$
 (xxvii) Ag-Pd/C, base, O₂

SCHEME 23: Synthesis of quinoline, 2.19.5.



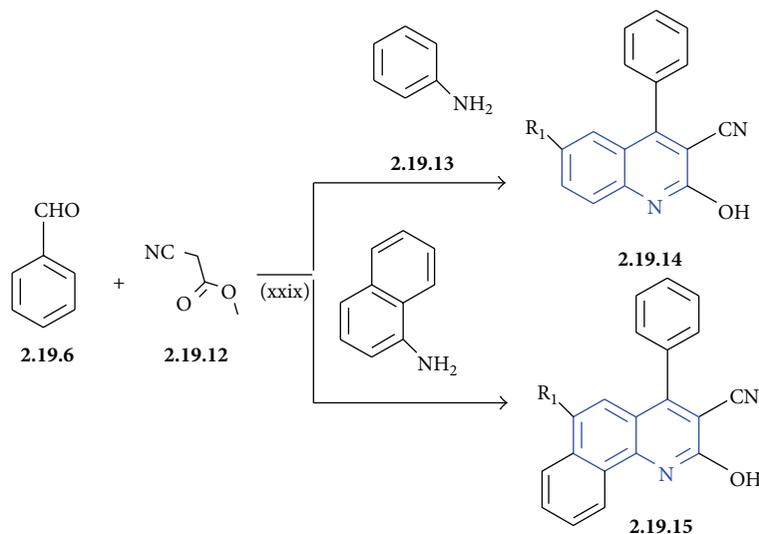
$\text{R}_1 = \text{C}_6\text{H}_5, \text{4-MeOC}_6\text{H}_4, \text{4-BrC}_6\text{H}_4, \text{4-ClC}_6\text{H}_4, \text{4-MeC}_6\text{H}_4$
 (xxviii) CuO nanoparticles, 50°C, 30–45 min

SCHEME 24: Synthesis of imidazo[1,2-*a*]quinoline, 2.19.11 and quinolino[1,2-*a*]quinazoline, 2.19.10.

The doping of Mn increased the activity and selectivity of nanoparticles [92].

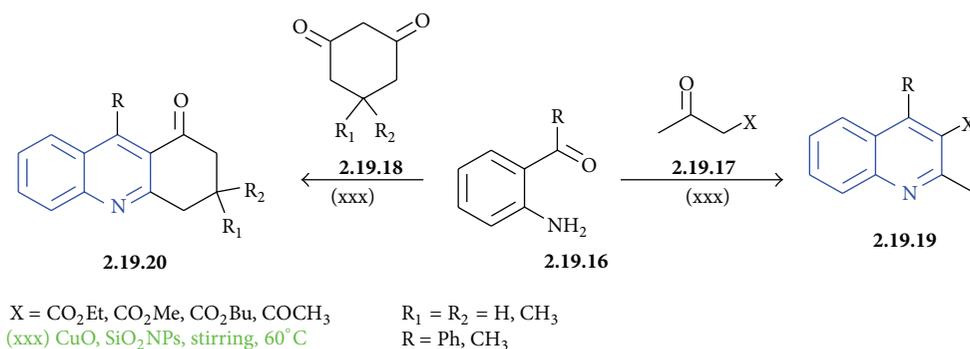
2.16. Synthesis of Imidazoles. Imidazoles are present in various pharmacologically active compounds which act as anti-tuberculosis agent [93] and antibacterial agent [94]. They were synthesized as either-trisubstituted or -tetrasubstituted imidazoles by using various reaction conditions such as ultrasonic irradiation [95], TBAB catalyst [96], and HClO₄-SiO₂ catalyst [97]. Imidazoles (Scheme 20) can also be obtained by multicomponent reaction using benzil, 2.16.1, aldehydes, 2.16.2, and amines, 2.16.3, in the presence of

metal nanoparticles as a catalyst. TiCl₄ supported on silica was used as a mild solid Lewis acid for the synthesis of triphenylimidazoles. This catalyst system can be prepared, handled, and stored without any special precautions by maintaining its efficiency. They carried out the reaction under solvent-free condition at 110°C for 30 minutes [98]. The solvent-free synthesis of imidazoles was explored with sulfonic acid functionalized SBA-15 as a catalyst. It was found that aliphatic aldehyde gave moderate yield and the aromatic aldehyde with electron-withdrawing and electron-donating groups gave excellent yield in the presence of catalyst and it could be recovered by continuous washing with dilute acid, water, and acetone [99]. A mild Lewis acid catalyst, MgAl₂O₄,



(xxix) TiO₂NPs, MW

SCHEME 25: Synthesis of quinoline-3-carbonitriles, **2.19.14** (or) benzo[*h*]quinoline-3-carbonitrile, **2.19.15**.



X = CO₂Et, CO₂Me, CO₂Bu, COCH₃
(xxx) CuO, SiO₂NPs, stirring, 60°C

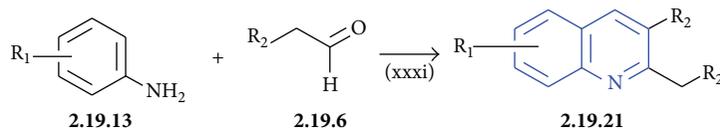
R₁ = R₂ = H, CH₃
R = Ph, CH₃

SCHEME 26: Friedlander methodology to synthesis of quinoline analogues, **2.19.19-2.19.20**.

was utilized for the efficient synthesis of substituted imidazoles under ultrasound irradiation. Because of the decrease in size of the crystal magnesium aluminate, a defect was produced in the coordination of constituent atoms which increases the reactivity of the catalyst, and thereby it leads to cyclocondensation reaction for the formation of imidazoles [100]. The synthesis of imidazole was carried out using clay and zeolite and also with nanocrystalline-sulfated zirconia catalyst in the presence of ethanol at room temperature. The optimization of the reaction condition was performed and found that the yield was increased up to 93% by the SZ catalyst [101]. The Bronsted acid nanoreactor, MCM-41-SO₃H, was involved in the solvent-free synthesis of trisubstituted and tetrasubstituted imidazoles. In this experiment, it was found out that the solvents have no role on the synthesis of imidazoles. The modified action of the nanoreactor increased its efficiency and resulted in higher yield and good reusability [102]. An efficient catalyst, magnetic Fe₃O₄ nanoparticles, can also be used for the synthesis of imidazole derivatives. Magnetic Fe₃O₄ nanocatalyst and temperature (80°C) play

a crucial role in this reaction under solvent-free condition and gave a maximum yield of up to 96% [103]. Rostamizadeh and coworkers developed a toxic-free solvent reaction for the synthesis of 2,4,5-trisubstituted and 1,2,4,5-tetra-substituted imidazoles using nanosized MCM-41-SO₃H as a catalyst [88].

2.17. Synthesis of Pyrimidine Carbonitriles. The three-component reaction involving aldehydes, **2.17.1**, malononitrile, **2.17.2**, and amidines, **2.17.3**, in the synthesis of 4-amino-5-pyrimidinecarbonitriles, **2.17.4**, (Scheme 21) was catalyzed using CuO microspheres. CuO microspheres are made by granulation of nanoparticles using immobilization-calcination method [104]. Even though the surface area of microspheres is lesser than nanoparticles, they are larger than bulkier substances. The main purpose of microspheres is to avoid the physical instability of nanoparticles such as agglomeration. The polar solvents such as THF, CH₃CN, and CH₃CH₂OH gave fewer yields than water in the synthesis of 4-Amino-5-pyrimidinecarbonitriles. 4-amino-5-pyrimidinecarbonitriles can also be synthesized using ZnO

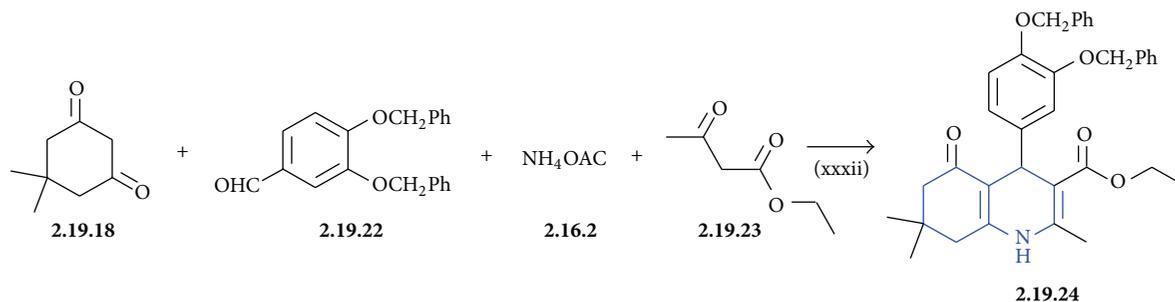


R₁ = CH₃, CH(CH₃)₂, OCH₃, Ph, Cl

R₂ = C₆H₅

(xxx) AuNP/SiO₂, O₂ bubbling, 110°C, 6 h

SCHEME 27: Role of Gold and SiO₂ in quinoline synthesis, 2.19.21.



(xxxii) Fe₃O₄-Cys, EtOH, 25 min

SCHEME 28: Multicomponent approach in quinoline synthesis, 2.19.24.

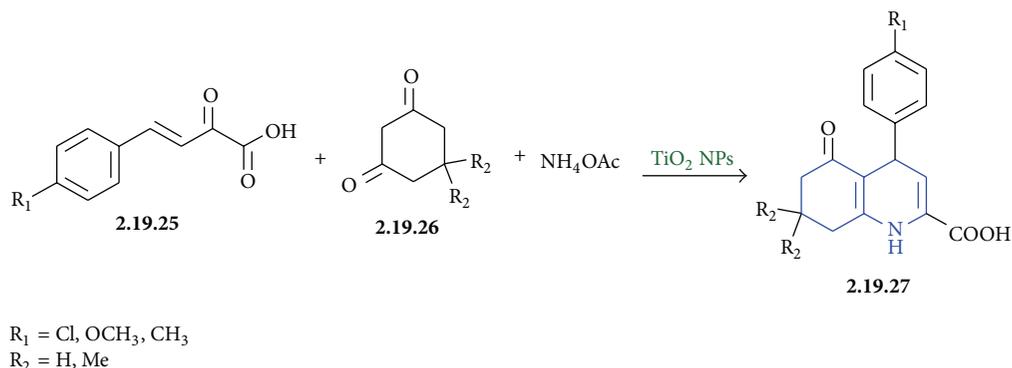
nanoparticles. Being insoluble in water and other organic solvents it can be easily recovered from the reaction mixture immediately after the reaction [105].

2.18. Synthesis of Spirohexahydropyrimidines. A one-pot condensation of cyclohexanone, 2.18.1, ketone, 2.18.2, and aniline, 2.18.3, using innovative preyssler nanoparticles, H₁₄[NaP₅W₃₀O₁₁₀]/Si as an efficient catalyst system for synthesizing 1,3-diaryl-5-spirohexahydropyrimidines, 2.18.4, (Scheme 22) [106].

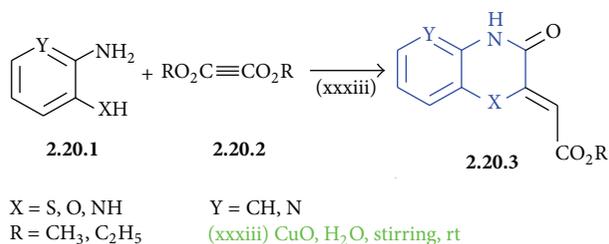
2.19. Synthesis of Quinoline Analogues. Quinoline nucleus is one of the important constituents which is present in the naturally occurring alkaloids. It has proven antimalarial activity along with many other important pharmacological actions. In the synthesis of polysubstituted quinolines, 2.19.5 (Scheme 23) first step involves the α -alkylation of ketone, 2.19.2, with alcohol, 2.19.1, to give saturated ketone, 2.19.3. In the second step, ketone, 2.19.3, undergoes modified Friedlander annulations process with 2-aminobenzyl alcohol, 2.19.4, to get the desired product. Ag-Pd alloy nanoparticles supported on carbon, a comparison of the activity of Ag-Pd/C catalyst with that of palladium-based nanocatalysts-core-shell Ag@Pd/C and Pd/C were studied. At 125°C, all the catalyst produced more- or less-same yield whereas at 90°C Ag-Pd/C catalyst superseded the other two catalysts in yield. This was explained due to the transfer of charge from less-electronegative Ag metal to more electronegative Pd [107]. An alternative method to synthesize quinoline derivatives such as imidazo[1,2-*a*]quinoline, 2.19.11, and quinolino[1,2-*a*]quinazoline, 2.19.10, (Scheme 24) is to heat the mixture of

aldehyde, 2.19.6, malononitrile, 2.19.7, enamines, (2.19.8, 2.19.9) at 50°C for 30–45 minutes in the presence of known solid base catalysts, bulk CuO, and CuO nanoparticles. All the catalyst is except CuO nanoparticles underwent reaction for a long time with moderate to poor yield. Whereas CuO nanoparticles produced excellent yield because of its insoluble nature in water, it can be recovered easily [108]. TiO₂-catalyzed synthesis of quinoline-3-carbonitriles, 2.19.14, (or) benzo[*h*]quinoline-3-carbonitrile, 2.19.15 and (Scheme 25) in the presence of water and microwave irradiation was carried out in an ecofriendly way. These derivatives can be synthesized from arylaldehyde, 2.19.6, cyanoacetate, 2.19.12, anilines, 2.19.13, using knoevenagel condensation, Michael addition followed by aromatization. The report indicated that the nanocatalyst showed superior reactivity than the conventional method [109].

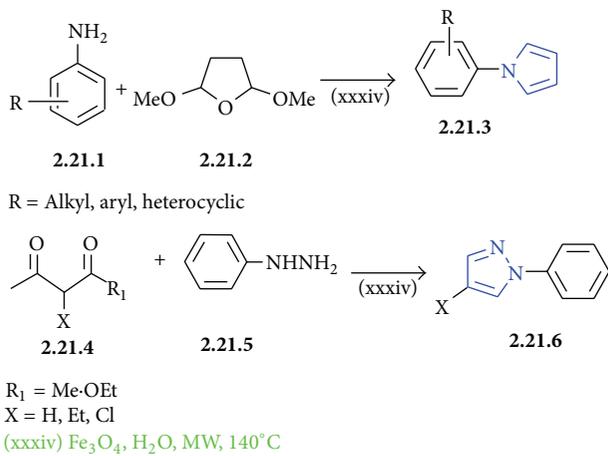
The optimization of Friedlander synthesis of quinolines (Scheme 26) was carried out on various of 2-aminoaryl ketones, 2.19.16, active methylene compounds, 2.19.17, and simple cyclic ketones, 2.19.18, under different catalysts (TiO₂, SiO₂, Al₂O₃, ZnO, MgO, CuO bulk and nano-CuO) in solvent-free condition at 60°C, and nano-CuO was found superior to all the other catalysts [110]. So et al. explored that AuNPs/SiO₂ + O₂ as an efficient catalyst system for the synthesis of polyheterocyclic compounds containing nitrogen, 2.19.21 (Scheme 27) from aniline, 2.19.13, and aldehyde, 2.19.6. They performed the mechanistic studies of quinolines and reported that the reaction does not follow the radical pathway, and the yield was very less in the presence of silica alone. Therefore, AuNPs/SiO₂ + O₂ protocol is the optimal one for the quinoline synthesis [111]. Ferrite magnetic



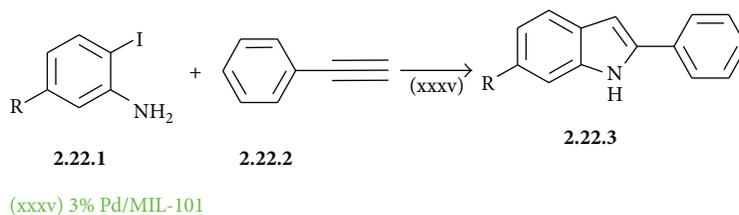
SCHEME 29: Metal oxide-mediated quinoline analogue synthesis, 2.19.27.



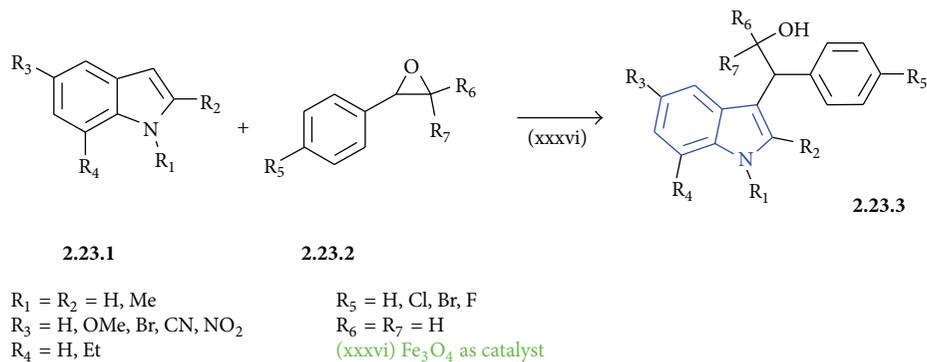
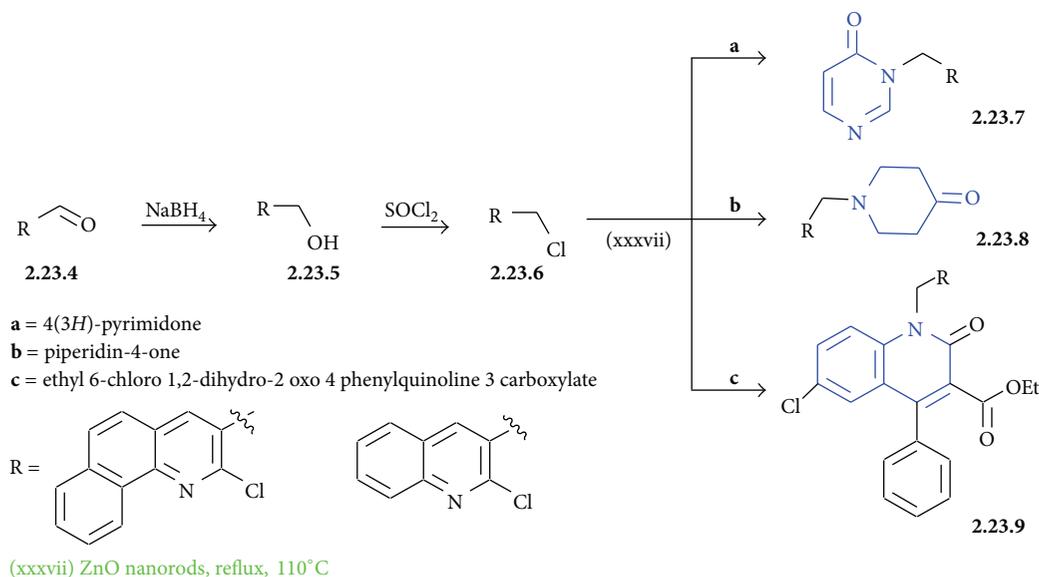
SCHEME 30: CuO-mediated various benzoheterocycle synthesis, 2.20.3.



SCHEME 31: Synthesis of pyrrole, 2.21.3, and pyrazole, 2.21.6.



SCHEME 32: Green synthesis of indole, 2.22.3.

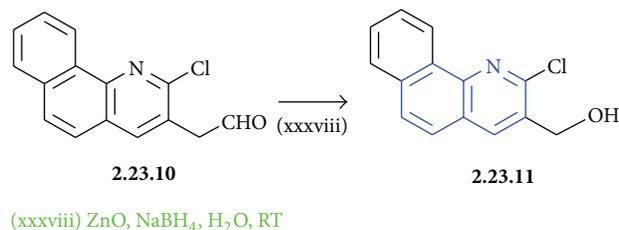
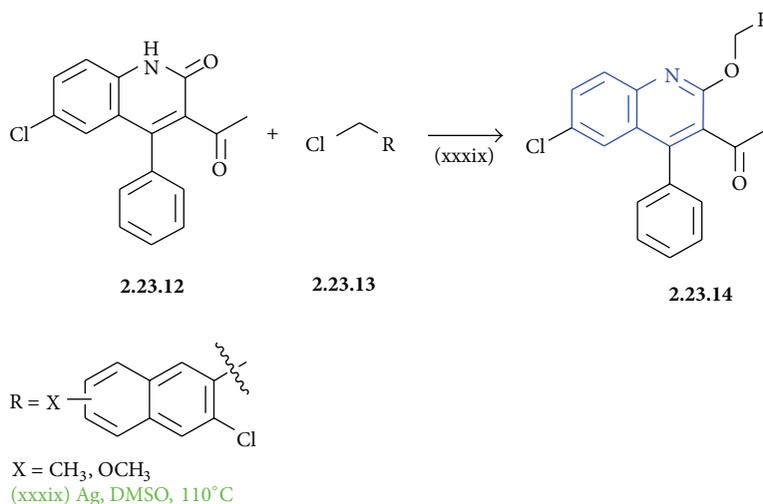
SCHEME 33: C-alkylation of indole using Fe₂O₃, **2.23.3**.SCHEME 34: Cross-coupling reaction, **2.23.7–2.23.9**.

nanoparticles with cysteine can be used as catalyst for the Hantzsch synthesis of hydroquinolines, **2.19.24**, (Scheme 28) in a multicomponent reaction between 3,4-diphenoxybenzaldehyde, **2.19.22**, ammonium acetate, **2.16.2**, ethyl acetoacetate, **2.19.23**, and 5,5-dimethylcyclohexane-1,3-dione, **2.19.18**. The yield was up to 88%, and it could be reused with unaltered activity until 9 cycles [112]. Abdolmohammadi developed a green friendly protocol for the synthesis of 5-oxo-4-aryl-1,4,5,6,7,8-hexahydro-2-quinolinecarboxylic acid, **2.19.27** (Scheme 29) using TiO₂ nanoparticles under solvent-free conditions [113].

2.20. Synthesis of Benzoheterocycle Derivatives. CuO nanoparticles are capable of synthesizing various compounds which have both pharmacological and industrial applications. In the benzoheterocycles, **2.20.3** (Scheme 30) formation, reaction between aromatic amine, **2.20.1**, and dialkyl acetylenedicarboxylate, **2.20.2**, catalyzed by CuO nanoparticles was optimized by various solvents (water, dichloromethane, ethanol, and acetonitrile). Water excels in

yield more than the other solvents. CuO has both oil- and water-resistant character. So it can be reused by easy recycling process without losing its efficiency [114].

2.21. Synthesis of Pyrrole and Pyrazole. Nanoparticles combined with organic compounds constitute a vital role in organic synthesis. Here, the magnetic nanoparticles are modified with aminoacids such as cysteine and glutathione. Both aminoacids have highly reactive thiol groups which can easily functionalize the nanoferrite surface. Of the two aminoacids, glutathione is superior to cysteine in reactivity. The active sites of the nanocatalyst were left free for catalyzing the reaction. This catalyst can be applied in the Paal-Knorr reactions of pyrrole, **2.21.3**, (Scheme 31) synthesis between a variety of amines, **2.21.1**, and tetrahydro-2,5-dimethoxyfuran, **2.21.2**. Also this catalyst can be used in the synthesis of pyrazole, **2.21.6**, between 1,3-diketone, **2.21.4**, and hydrazines, **2.21.5**. The entire reaction was carried out in toxic-free solvent (water) and effective microwave irradiation [115].

SCHEME 35: ZnO nanoparticles mediated reduction of quinoline-3-carbaldehyde, **2.23.11**.SCHEME 36: O-alkylation of quinoline, **2.23.14**.

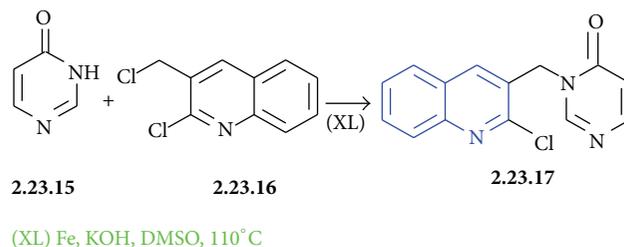
2.22. Synthesis of Indole. Synthesis of indole was carried out using an innovative catalyst, Pd supported on the cages of MIL-101 [Cr₃F(H₂O)₂O(bdc)₃]. This support is more water soluble than the other inorganic supports. Thereby, the reactions in aqueous solution will be catalyzed effectively. The reaction between 2-iodoanilines, **2.22.1**, and phenyl acetylene, **2.22.2**, in the presence of 3%-Pd/MIL-101 will lead to the formation of indole, **2.22.3**, (Scheme 32). In addition, the substituents on the ring will have an effect on the current reaction medium [116].

2.23. Miscellaneous Functionalization on Heterocycles. Parella and coworkers carried out the C-alkylation reaction (Scheme 33) of indoles, **2.23.1**, with epoxide, **2.23.2**, using the magnetic nano-Fe₃O₄ as a catalyst [117]. Roopan and Khan explored an efficient ligand-free cross-coupling reaction of 2-chloro-3-(chloromethyl) benzo[*h*]quinoline, **2.23.6**, with *N*-heterocycles such as piperidin-4-one, 4(3*H*)-pyrimidone, and ethyl 6-chloro-1,2-dihydro-2-oxo-4-phenylquinoline-3-carboxylate using a catalytic amount of ZnO nanorods as a recyclable catalyst to give its corresponding derivatives, **2.23.7–2.23.9** (Scheme 34), respectively [118]. The possible mechanism is described in Figure 4. The chloromethyl derivative of quinoline, **2.23.6**, is obtained from hydroxymethyl derivative, **2.23.5**, which in turn is obtained from aldehyde, **2.23.4**. The reduction of 2-chloroquinoline-3-carbaldehydes, **2.23.10**, into (2-chloroquinolin-3-yl)

methanol, **2.23.11**, occurs using zinc oxide nanoparticles as a catalyst in an ecofriendly way (Scheme 35) [1]. Roopan and coworkers synthesized 1-{2-[(2-chloroquinolin-3-yl) methoxy]-6-chloro-4-phenylquinolin-3-yl} ethanones, **2.23.14**, (Scheme 36) from heteroalkylhalides, **2.23.13**, and cyclic amides, **2.23.12**, using silver nanoparticles in regioselective O-alkylation reaction [119]. Furthermore, they have used Fe nanoparticle for the regioselective N-alkylation of 4(3*H*)-pyrimidone, **2.23.17**, (Scheme 37) with various quinoline containing alkyl halides in an ecofriendly way. The possible mechanism is described in Figure 5. [120].

3. Conclusions

This review is the first attempt to compile the literature on the subject of nanomaterials application in organic synthesis. It should be noted that a correct and update citation and literature survey is very important for researchers to find relevant information, pioneer ideas, and progress of any subject. On the other hand, published data using nanomaterials indicate a wide synthetic potential of the described catalysts and a great interest of researchers in this field. The use of green nanocatalyst for the synthesis of various heterocycles has advantages such as short reaction time, high yield, inexpensive chemicals usage, easy work-up procedure, and very specific reaction [2]. The use of nanocatalyst can also be applied on the synthesis of various heterocycles which



SCHEME 37: Regioselective alkylation reaction towards pyrimidone molecule, 2.23.17.

are very difficult to prepare by conventional methods. Also more and transition metals can be checked for its catalytic activity and surface modifications of the existing catalyst can also be performed. In most of the reactions the spent catalyst can be easily separated from the reaction mixture, also it can be reused without noticeable change in its catalytic activity. A wide range of original procedures for synthesizing various classes of organic compounds, including organic functional group transformation, have been developed on the basis of nanoparticles. We assume that the present review article may be bringing a basis to advance information to this very important subject and to encourage active researchers in this field for the synthesis of organic compounds using nanoparticles.

Acknowledgments

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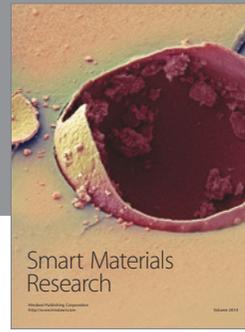
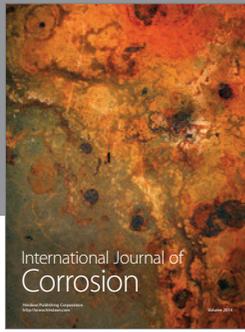
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