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Synthesis of Quinazolines Scaffold Using Green and Sustainable Heterogenous Nano-catalysts

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Abstract

Six-membered nitrogen containing heterocyclic bioactive molecules like quinazolines, and its derivatives have gained significant interest in recent years. Quinazolines have found to be an important base moiety in many of drugs used in the treatment of cancer and other diseases. Syntheses of quinazolines using green and sustainable methods have grabbed reasonable attention of researchers, to limit the use hazardous catalysts and reaction conditions. Among various methods, use of heterogenous eco-friendly nano-catalysts have emerged as an excellent approach for preparation of quinazolines. Nanoparticle based catalysts possess many advantages over conventional catalytic system since nano-catalyst provides high surface area for the rapid conversion into product even under mild reaction conditions and easily separated out from reaction mixture without following tedious work-up procedures. In this review we documented various nano-catalysts used for synthesis of biologically active quinazolines and their derivatives.

Keywords

Multi-component reactions, 2,3-dihydroquinazolin-4(1H)-ones, Heterogenous nano-catalyst, Green and sustainable catalyst

Introduction

In the field of organic synthesis and drug discovery research, there is growing interest in designing and development of energy-efficient and sustainable synthetic strategies, particularly for the synthesis of heterocyclic molecules with molecular complexity and structural diversity [1] via multi-component reactions. Multi-component reaction involves reaction between two or more components reactions in one step thus preferred to the conventional multi-step synthetic technique because they offer desired advantages lie structural variety and chemical complexity, easy experimental methodology and cost effectiveness. Development of nitrogen containing heterocyclic moieties exhibiting wide spectrum of biological property including anticancer, antibacterial, anti-inflammatory, antimalarial, and antihypertensive effects generally synthesize via multi-component reactions and have gained much attention in recent years. One such nitrogen-containing heterocyclic scaffold is quinazolines having chemical formula $C_8H_6N_2$. Quinazolines are fused, planar six-membered aromatic rings, possessing a benzene ring and a pyrimidine ring. Quinazolines and its derivatives exhibit versatile applications in medicinal chemistry such as antimalarial, anticancer agents and used as analgesics as depicted in figure 1. Quinazoline scaffold linked with various moieties, or bicyclic residue were

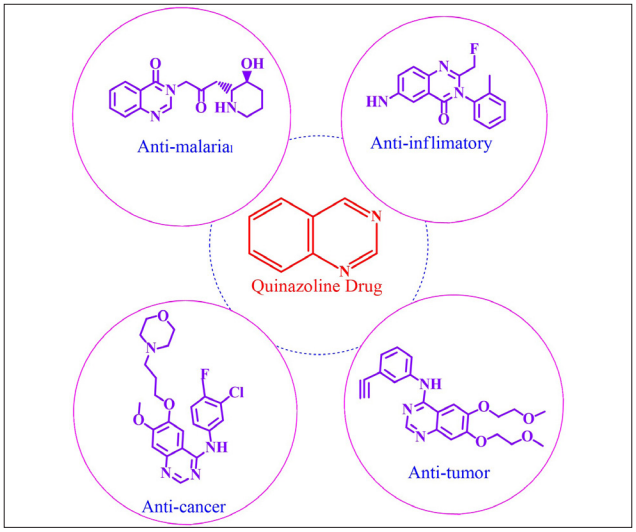


Figure 1: Derivatives of quinazolines.

found to possess potent anticancer [2] antimalarial activities [3], and as an inhibitor moiety for treatment of Alzheimer's disease [4]. Apart from widespread use of quinazolines in various bioactivities, quinazolines have also been used as serine/threonine kinase inhibitors or as selective inhibitors of Aurora B kinase [5] which have ability to control embryonic development, cell differentiation, programmed apoptosis, and cell proliferation. In literature, various synthetic protocols have been developed by research scientists from all over the world for synthesis of six-membered quinazolines using different reaction conditions and in presence of different catalysts like deep eutectic mixture of maltose-dimethyl-urea [6], magnetic ionic liquid bmim ($FeCl_4$) [7], penta-fluoro-phenyl ammonium triflate [8], 1-methyl-imidazoliumtriflouroacetate [9], and ceric ammonium nitrate-tert-butyl hydroperoxide [10]. It has been noticed that reaction conditions used in presence of these catalyst are not environment friendly and harsh like high temperature, hazardous volatile organic solvents, tedious work-up procedure. Therefore, to synthesize structurally diverse heterocyclic molecules with ecofriendly conditions, green and efficient catalysts especially magnetic nanoparticles are excellent choice under mild reaction conditions.

In view of recent advancement in field of various catalysts like homogeneous, heterogeneous, nano-catalyst, endo selective, organo-catalyst and inorganic catalyst, use of heterogenous nano-catalysts have received extraordinary growth due to high surface area, easy separation, excellent yield, and selectivity. Most nanoparticles are synthesized from different metals, metal oxides, polymers, or composite materials and can be used in synthesis of various bioactive heterocyclic moieties. It was noticed that from last decade, researchers have more focused on modification of nanoparticle based catalysts like formation of magnetic nanoparticles [11], graphene based nanoparticles [12], Schiff-based nanoparticles [13], core-shell nano-catalyst [14], and thin-film based nano-catalyst [15] which not only decrease the reaction time with improved product yield but also increases the selectivity and catalytic activity of the catalyst. Reports suggests that the magnetic nano-catalyst can be used as strong support for immobilization of organic and inorganic nano-catalyst [16] and they can easily be recovered which makes them more environment-friendly and increase effectiveness for the use as a catalyst in catalyzing organic transformation. Minimum waste generation, low cost, availability of active sites for conversion of reactants into products make them the best candidate in catalytic chemistry as compared to homogenous and heterogeneous catalyst. The present review article updates about the recent advancements in synthesis of six membered fused heterocyclic quinazolines by using green and sustainable nano-catalysts which have allowed scientists to explore the various types of nano-catalyst which are reusable and exhibit high catalytic efficiency. A green protocol for preparation of functionalized quinazoline was developed by Santra and co-workers [17] using nano In_2O_3 catalytic system. The catalytic efficiency of nano-In_2O_3 was evaluated and optimized from reaction between isatoic anhydride, aniline, and benzaldehyde in equimolar amount to yield substituted quinazolines. The reaction was optimized by using 5.0 mol% of nano-In_2O_3 catalyst at 80 °C in water /ethanol (2:1) as reaction medium to provide highest yield in 4 h. Nano-In_2O_3 catalyst was found to be an efficient catalyst as it not only provides high yield as compared to other catalyst but also can be recycled and used for up to six times under similar reaction conditions without much loss in its activity. Different spectroscopic methods like scanning electron microscopy (SEM), X-ray diffraction (XRD), transmission electron microscope (TEM) confirmed the synthesis of In_2O_3 nanoparticles.

In view of continuous demand of less toxic, cheap, and environmentally sustainable heterogenous catalysts, a green synthetic protocol to synthesize biologically active quinazolines using 2,3-hydroxyapatite nanoparticles. The magnetic hydroxyapatite nanoparticles were synthesized by the reaction of $Ca(NO_3)_2.4H_2O$ and $(NH_4)_2HPO_4$ at 90 °C within 5 h as shown in figure 2. The efficiency of catalyst was monitored through condensation reaction of isatoic anhydride, aniline, and aldehyde in equimolar ratio in presence of magnetic hydroxyapatite nanoparticles. It was found that scope of reaction was expanded to different electron-withdrawing and electron-donating groups and latter ones found to give better results. However, substituents attached to para and meta position give results in less time as compared to ortho-substituted aldehydes. The use of aqueous medium, ambient reaction condition, high product yield are some advantages of reported protocol [18].

A Similar method for synthesis of 2,3-dihydroquinazolin-4(1H)-ones was reported by Mohammadi and Ghorbani-Choghamarani where they used sulfuric acid based hercynite

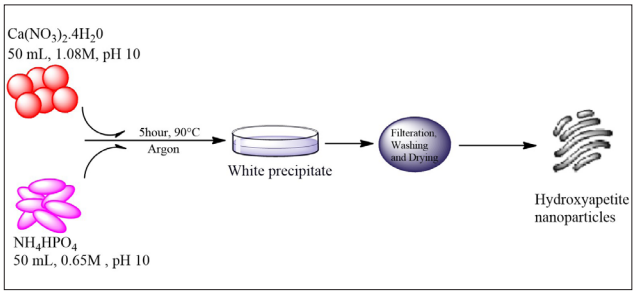


Figure 2: Preparation of hydroxyapetite nanoparticles.

magnetic nanoparticles. The novel hercynite@sulfuric acid was synthesized through co-precipitation reaction between $FeCl_2.4H_2O$ and $Al(NO_3)_2.9H_2O$ in deionized water to yield hercynite magnetic nanoparticles followed by treatment of chloro sulfuric acid as shown in figure 3.

The $FeAl_2O_4$ spinel ferrite hercynite magnetic nanoparticles provide the surface for immobilization of sulfuric acid which enhances the rate of reaction by lowering the reaction time. The catalytic efficiency of magnetic nanoparticles was studied for the reaction of p-chlorobenzaldehyde and anthranilamide. It was observed that reaction give excellent yield when catalyst was refluxed at 75 °C in ethanol for 15 min (Figure 4). It was found that using various electron-withdrawing and releasing substituents under optimized reaction condition give almost similar results as that of model reaction. Hercynite@sulfuric acid was also investigated for synthesis of polyhydroquinoline, and the reaction afforded good to excellent yield. Further, catalyst studies suggest that catalysts can easily be separated by using external magnet and can be recycled and reused five times for similar reactions [19].

In another report Singh and coworkers used nickel nanoparticles for efficient synthesis of triazoloquinazolinones. The nickel nanoparticles were easily prepared by reducing nickel chloride hexahydrate with hydrazine hydrate and further characterized by SEM, TEM, EDX (Energy dispersive X-ray spectroscopy) and powder XRD. The catalytic activity of reported catalyst was investigated for one-pot reaction between substrates like dimidone, 3-amino-1H-1,2,4-triazole/3-amino-5-methyl-1-H-pyrazole and aldehyde in equimolar ratio to yield triazoloquinazolinones. The reported methodology is associated with various advantages like recyclability, reusability, easy preparation of nanoparticles, lower reaction time, high yield, etc. [20].

In this context, a naturally occurring saponin coordinated with copper(II) metal was immobilized on a solid support of iron oxide (Fe_2O_3) nanoparticles to prepare an environmentally benign Fe_3O_4@Sap/Cu(II) catalyst as depicted in figure 5. The synthesized catalyst was found to be less toxic, recoverable and phase transfer catalyst and used for the synthesis of quinazolines and acridines by Kazemnejadi et al. The efficiency of catalyst was investigated under optimal conditions which include reaction between aromatic aldehyde, amines/phenyl hydrazine/ammonium acetate and isatoic anhydride in water at room temperature to produce up to 96% yield. Further, scope of reaction was also explored with different electron donating and withdrawing substituents present on aldehyde and amine and there was no significant change observed in yield of different derivatives of 2,3-Dihydroquinazolin-4(1H)-ones. The high activity of Fe_3O_4@Sap/Cu(II) was anticipated due to the synergistic effects of Fe_3O_4 nanoparticles and saponin, which not only increase the active surface of the catalyst, sites for attachment of copper metal, but also reduce the agglomeration of magnetic nanoparticles. The reported protocol exhibited advantages such as environmentally friendly, high thermal stability, cheap, recyclability and reusability of catalyst up to six cycles with without loss in its activity [21].

Similarly, selenium nanoparticle catalyzed synthesis of imidazo-quinazoline were reported by Abdelghany and co-workers. In their work, the catalytic efficiency of selenium nanoparticles was studied for the reaction between aldehyde, β-naphthol and 2-aminobenzoimidazole in distilled water at 60 - 70 °C (Figure 6). Detailed studies revealed that rate of reaction and yield was increased when electron-withdrawing groups are attached on aldehyde. The reported selenium nanoparticles were biologically prepared by sodium selenate and albumin and formation of selenium-nanoparticles which was further

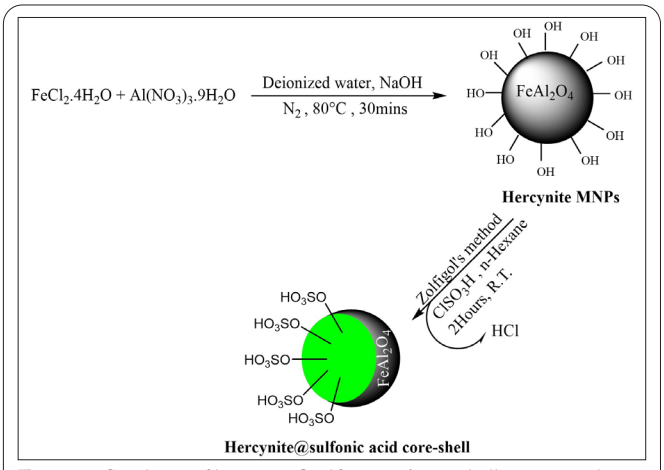


Figure 3: Synthesis of hercynite@sulfuric acid core shell nanoparticles.

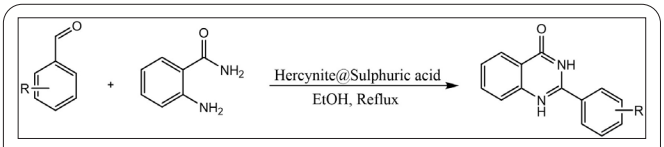


Figure 4: Synthesis of 2,3-dihydroquinazolin-4(1H)-ones in presence of hercynite@sulfuric acid.

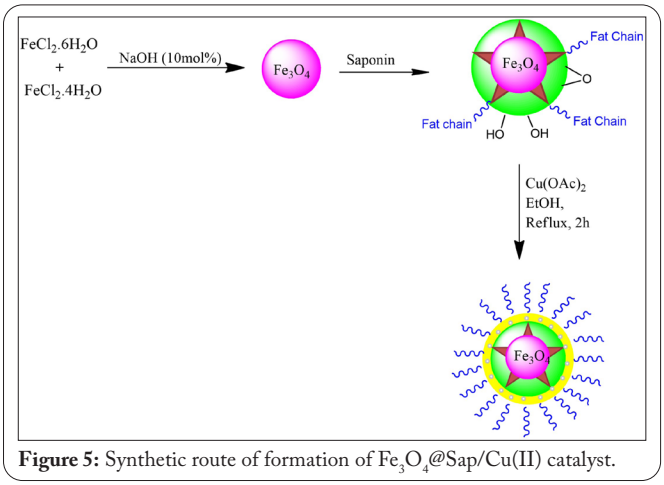


Figure 5: Synthetic route of formation of Fe_3O_4@Sap/Cu(II) catalyst.

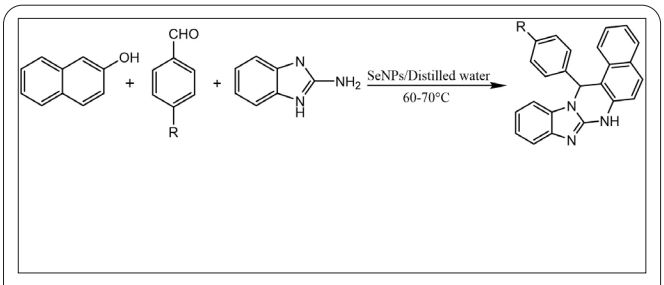


Figure 6: Synthesis of imidazo-quinazoline using selenium nanoparticles.

confirmed by Fourier transform infrared (FTIR) spectroscopy, ultraviolet-visible spectroscopy, TEM, and EDX [22].

Another method was reported to synthesize quinazoline-2,4(1H,3H)-diones using dendritic fibrous nano-silica supported zinc oxide (ZnO) nanoparticles. Shamsa and co-workers exploited ZnO nanoparticles and embedded them on nanosilica (DFNS) blended with cellulose to form DFNS/cellulose-ZnO and used in preparation of quinazoline-2,4(1H,3H)-diones (Figure 7). In the synthesis of reported catalyst, condensation of 3-CPTES and hydroxyl group present on the surface of DFNS to form DFNS@N_3 which on further reaction with alkyne-functionalized cellulose leads DFNS/cellulose-ZnO as depicted in figure 8. It was found that presence of ZnO nanoparticles increases surface area of catalyst which results improvement in catalytic activity. Additionally, the influence of particle size and surface area was studied by SEM, XPS, EDX, ICP-MS, TEM, BET, and TGA. Assessment of reusability and recyclability confirmed that there is not much loss in the product yield and catalyst can be used for up to 10 cycles [23].

The reaction was optimized in presence of carbon dioxide when substrates like 2-iodoaniline, tertbutyl isocyanides, 1,8-diazabicyclo[5.4.0]undec7-ene, at various temperatures and reaction medium. The best yield was obtained when anhydrous DMF was used as a solvent and temperature kept at 70 °C for the reaction medium. Use of various bases was also explored in the reaction and Cs_2CO_3 used as base afforded the higher yield as compared to when no bases were used.

Eidi and co-workers reported green synthesis of Fe_3O_4@SiO_2-$PrNH_2$ magnetic nanoparticles as shown in figure 9. In the synthesis of reported nanoparticles, Fe_3O_4 was coated with silica then functionalized with amino group and finally grafted with Schiff base. The synthesized Fe_3O_4@SiO_2-$PrNH_2$ was used in reaction between ammonium acetate, aldehyde, 2-amino aryl ketones at 60 °C. It was observed that no change in the yield and the catalytic activity was found when the catalyst was used for four consecutive cycles and in case of electron withdrawing and donating groups. The synthesized magnetic nanoparticles were confirmed and characterized with various analytical techniques [24].

In another report, synthesis of quinazoline was commenced using fibrous nano-silica based metal nano-catalyst functionalized with hyperbranched polyglycerol groups (HPG) These fibers provide a high surface area with active sites for catalysis as the metal nanoparticles of Au, Pd, and Cu can be dispersed easily without causing aggregation. The catalytic activity of fiber-based nano-catalyst was investigated on reaction between CO_2 and 2-amino benzonitriles to yield quinazoline-2,4(1H,3H)-diones. The reaction was optimized using KCC-1/HPG/X and KCC-1/X with different metals and it was observed that KCC-1/HPG/X yield better results in 40 min at 100 °C when water was used as solvent. The catalytic study reveals that KCC-1/HPG/X can be used for up to 10 cycles without any significant loss. Apart from other attributes like eco-friendliness, reusability, and cost effective KCC-1/HPG/X catalyst also possess twice the stationary nanoparticles as compared to KCC-1/X which increases the

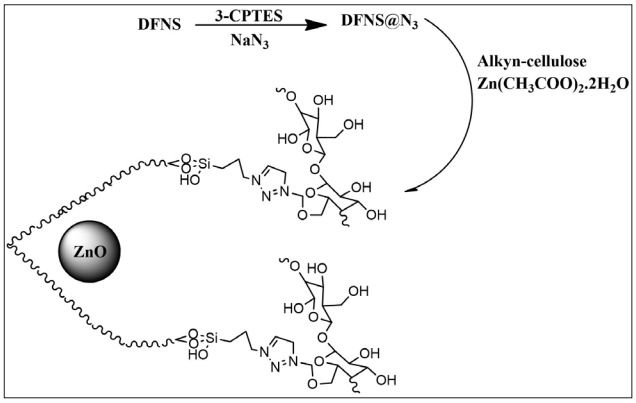


Figure 7: Synthesis of quinazoline-2,4(1H,3H)-diones.

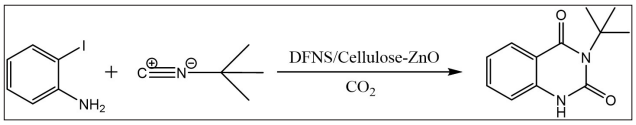


Figure 8: The synthetic pathway for synthesis of DFNS/cellulose-ZnO.

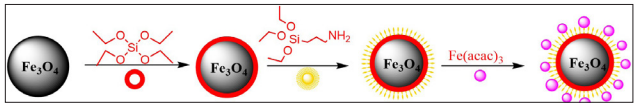


Figure 9: Preparation of magnetic nanocatalyst Fe_3O_4@SiO_2-$PrNH_2$- Fe^{3+}.

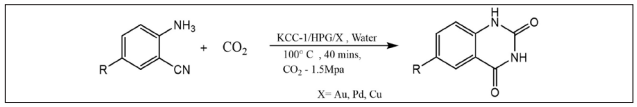


Figure 10: Synthesis of quinazoline-2,4(1H,3H)-diones derivatives.

rate of completion of reaction. Further study was confirmed by FTIR, TEM, powder XRD [25].

Baghbanian and Farhang synthesized magnetic $CuFe_2O_4$ nanoparticles by decomposing $Cu(NO_3)_2$ and $Fe(NO_3)_3$ in presence of sodium hydroxide using water as medium. The catalytic efficiency of magnetic reusable $CuFe_2O_4$ nanoparticles was investigated on synthesis of quinazoline and it was observed that the reaction completes in one hour

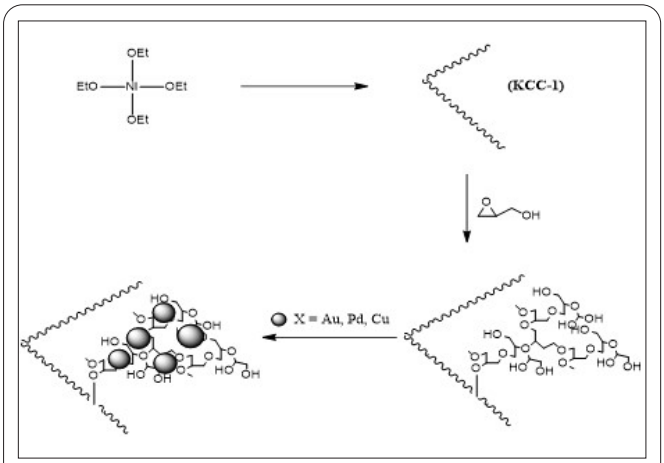


Figure 11: Schematic representation of synthesis KCC-1/HPG/X.

using 5.0 mol.% of catalyst at 80 °C in water yielding 95% of the product. The efficiency of catalyst was confirmed as the catalyst can be used up to 5 times without any significant loss [26]. General reaction for synthesis of quinazolines involves the reaction between isatoic anhydride, substituted aldehyde and substituted amine under different reaction conditions (Figure 12).

Similarly, in another report copper oxide (CuO) nanoparticles catalyzed synthesis of quinazoline was reported. The reported catalyst was initially optimized with aldehydes and N-arylamidines which further extended to the use of alcohols in place of aldehyde. It was interesting to fond that effect of catalyst was significantly improved CuO NPs was used with DMEDA, Ph_3P, TMEDA, and 1,10-phenanthroline when refluxed for 24 h at 110 °C in toluene (Figure 13) [27].

An effective and environmentally friendly synthetic method to synthesize substituted quinazolines from Fe-FeO_x nanoparticles bonded with N-doped carbon layers via strong anti-poisonous bond. It was found that one pot synthesis was carried out in simple reaction conditions like uses of water as a solvent and air as an oxidant at 130 °C for 24 h. The catalyst can be used up to 5 times [28].

Similarly, Kouser and coworkers reported environmentally benign methods for synthesizing quinazolines using Co(II)-NH_2-Si@Fe_2O_3 nanoparticles. In this report, authors used waste toner powder and transformed into iron oxide nanoparticles which further coated with silica and immobilized along Co(II) to produce Co(II)-NH_2-Si@Fe_2O_3 as depicted in figure 14. The catalyst was used prepare quinazoline derivatives by aldehyde, urea and dimidone in equimolar ratio in water/ethanol as solvent at 60 °C to provide more than 92% yield [29].

In another report, Ghorbani-Choghamarani and co-workers reported Fe_2O_3 nanoparticles supported on magnetic ionic liquid as shown in figure 15. Further, synthesis of catalyst was confirmed by various characterization techniques. The catalyst was used to generate nitrogen containing quinazolines

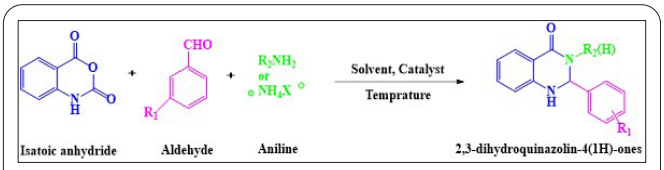


Figure 12: General reaction for synthesis of quinazolines and derivatives.

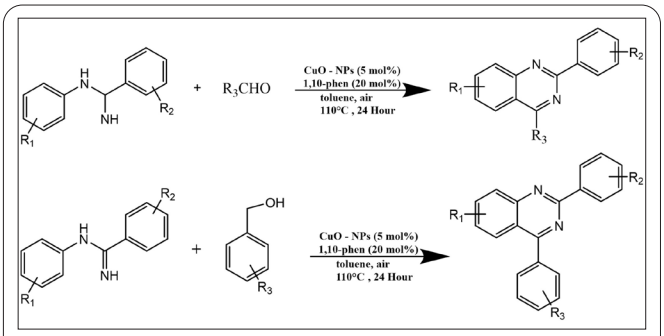


Figure 13: Synthesis of quinazoline from N-arylamidines and aldehydes or benzyl alcohol in presence of CuO NPs.

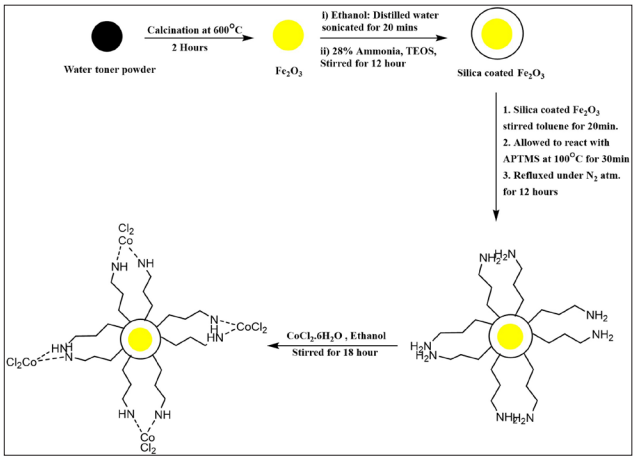


Figure 14: Schematic representation of preparation of Co(II)-NH_2-Si@Fe_2O_3.

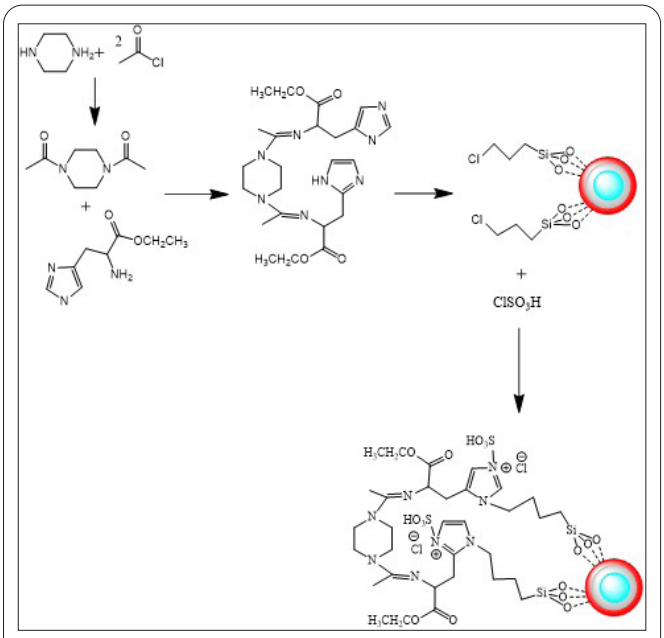


Figure 15: Schematic pathway for synthesis of nano-Fe_2O_3, supported ionic liquid.

molecules by using ninhydrin, benzyl amine and isatoic anhydride in PEG at 80 °C [30].

A plausible mechanism in synthesis of quinazolines and its derivatives, suggests that the role of catalyst was to activate the carbonyl carbon of isatoic anhydride which was attacked by nucleophillic amine to form amide as an intermediate, which further reacts with electrophilic aldehydic carbonyl carbon followed by cyclization of the ring as depicted in figure 16 (Table 1).

Conclusion

The present review comprises the recent development in field of catalysis for the synthesis of six-membered fused heterocyclic quinazolines rings. The review focuses on synthesis of nano-catalyst whose catalytic activity can be examined on synthesis of privileged class of heterocyclic scaffolds known as quinazolines, which affords wide range of applications in

Table 1: Different nano-catalysts used for synthesis of quinazolines scaffold.

S. No.	Nano-catalyst	Solvent	Temp. (°C)	Time	Yield (%)	Ref.
1	In_2O_3	Water:Ethanol (2:1)	80	4 h	72	[17]
2	2,3-hydroxyapatite nanoparticles	Water	110	1 h	95	[18]
3	Hercynite@sulfuric acid	Solvent free	100	20 min	99	[19]
4	Nickel nanopartilces	CH_3CN	80	10 min	97	[20]
5	Fe_3O_4@Sap/Cu(II)	Water	RT	<1 h	96	[21]
6	Selenium nanoparticles	Distilled water	60 - 70	30 min	88	[22]
7	DFNS/Cellulose-ZnO	Anhyd. DMF	70	3 h	98	[23]
8	Fe_3O_4@SiO_2-PrNH_2-Fe^{+3}	Solvent free	60	4 h	95	[24]
9	KCC-1/HPG/X X = Au, Pd, and Cu	Water	100	25 min	70 - 94	[25]
10	$CuFe_2O_4$ nanoparticles	Water	80	32 min	95	[26]

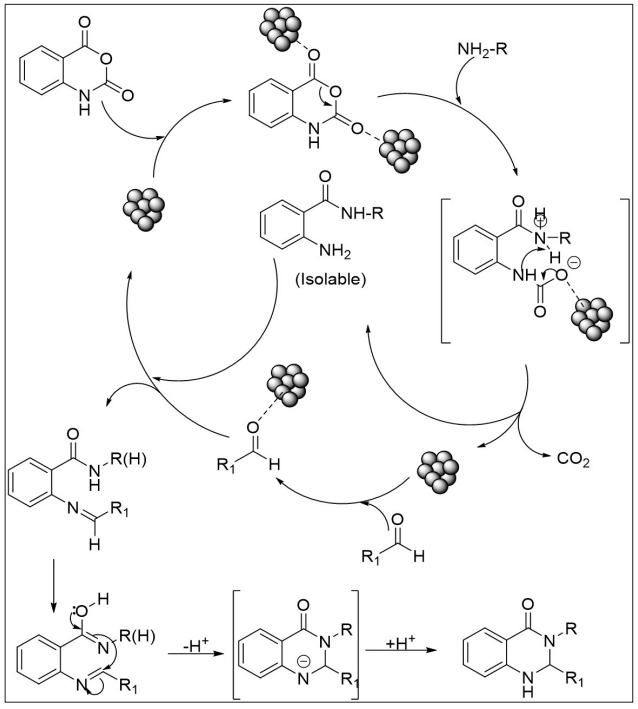


Figure 16: The proposed mechanism for synthesis of 2,3-dihydroquinazolin-4(1H)-ones in presence of nano-catalyst.

field of medical and pharmaceutical industries. To avoid harsh conditions involved in various multi-component reactions, use of nano-catalyst have helped scientist to design clean and efficient method for synthesis of fused six- membered heterocyclic compound like 2,3-dihydroquinazolin-4(1H)-ones and its derivatives. In this literature we have tried to bring up the detailed synthetic and mechanistic approach of green and sustainable nano-catalyst that would eventually be used in various organic transformations. We hope that the design and synthesis of nano-structured catalyst will provide an aid in synthetic route of heterocyclic medicinal compounds that will benefit society.

Acknowledgements

None.

Conflict of Interest

None.

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