

Chemistry & Biology Interface

An official Journal of ISCB, Journal homepage; www.cbijournal.com

Review Paper

Recent advancements of copper as an inexpensive and mild catalyst in heterocyclic synthesis

Anshu Dandia^{*a}, Jyoti Joshi^{*b}, Sukhbeer Kumari^b and Shyam L. Gupta^a

^aDepartment of Chemistry, University of Rajasthan, Jaipur-302004

^bDepartment of Chemistry, Malaviya National Institute of Technology, Jaipur-302017

Received 2 February 2013; Accepted 30 March 2013

Keywords: Catalysis, Cu(I) and Cu(II) compounds, Copper Nanomaterials, Multicomponent reactions, Heterocyclic synthesis

Abstract: Recent studies showed that Cu-catalysts proved to be a boon for heterocyclic synthesis. The Cu-catalysts made oxidative cross-coupling reactions, hydrogen transfer, carbon-carbon, carbon-heteroatom and heteroatom-heteroatom bond formations. It is environmentally benign and inexpensive than other transition-metal catalysts in organic synthesis. This review summarizes the recent advancements in Cu (I), Cu (II), and Copper nanoparticles catalyzed coupling reactions and synthesis of various complex heterocyclic compounds in mild conditions.

Introduction

There has been considerable interest in copper-mediated reactions for organic synthesis over other transition metal catalysts, as it is environmentally benign and economically viable [1-3]. A large number of copper compounds with variable oxidation states readily available [4-6]. Thus, due to their higher reactivity, efficient selectivity, high tolerance, low cost, non-toxicity, easy availability, easier operations and variable oxidation states, copper compounds are best suited for organic synthesis [7-9]. Moreover, copper is apparently more versatile and productive catalyst which leads to high-yielding

reactions and are found in numerous industrial and academic applications [10, 11]. Further, with reference to their high capacity in dioxygen activation and capability to use oxygen or air (as the green oxidant), make them superior catalysts or co-catalysts in cross-dehydrogenative coupling (CDC), C-H activation and aerobic alcohol oxidation [12-14]. Copper catalyzed reactions are also vital tools for constructing carbon-carbon, carbon-heteroatom and heteroatom-heteroatom bonds in organic synthesis [15]. Copper has a remarkable quality for facilitating hydrogen transfer from different set of donor molecules to specific acceptors [16].

Corresponding Author* email: dranshudandia@yahoo.co.in

A large number of useful reactions in organic synthesis have been catalyzed by copper catalysts (such as CuI, CuBr, CuCl, Cu₂O, CuCN etc.) in which copper is in +1 oxidation state. But, Copper (II) forms the most stable complexes and has been widely used and analyzed. The coordination geometry of Cu (II) complexes is also predictable. This makes the copper compounds with +2 oxidation state as most promising catalysts for organic synthesis with regards to both reactivity and selectivity [17]. There are many copper(II) compounds such as CuCl₂, CuBr₂, Cu(OTf)₂, Cu(NO₃)₂, Cu(OAc)₂, Cu(BF₄)₂·6H₂O, and Cu(ClO₄)₂·6H₂O etc. with +2 oxidation state [18].

Considerable attention has been paid to copper nanoparticles in the past two decades due to their unusual properties and potential applications in many fields [19-22]. Non-agglomerated, spherical, uniform copper nanoparticles have also been used in various other fields, such as catalysis, sensors, conductive films, lubrication, nanofluids and so on [23-29]. Recently a review on “dehydrogenative functionalization using copper as a catalyst” is published by Jiao *et al.* [30].

From the earlier days of development of organic chemistry to present state, heterocyclic compounds have held centered stage in the development of molecules to enhance quality of human life. For example, more than seventy percent of drugs used today are heterocyclic compounds [31]. They are widely distributed in nature and are key intermediates in many biological processes [32, 33]. Generally, heterocyclic compounds isolated from natural sources act as lead compounds for the development of new molecules of biological interest [34-36]. Heterocyclic compounds are well known for their multifaceted pharmacological and

biochemical behavior and as far as their relationship to medicinal chemistry is concerned, the two areas are almost inseparable [37, 38]. In recent years a vast number of pharmacodynamic heterocyclic have been developed which are in regular clinical use. Some of these are natural products, e.g. the antibiotic penicillin and the antibiotic erythromycin. Besides antibiotics, some other noteworthy natural heterocyclic alkaloids likes morphine and reserpine etc are well known [39]. However, the large majority are synthetic compounds, incorporating unusual systems and skeletal patterns [40, 41].

Heterocyclic compounds occupy a prominent position amongst the chemicals used for crop protection and pest control. The recent decades have witnessed the increasing use of heterocycles in the development of agrochemicals [42].

Reactions catalyzed by copper (I):

The copper(I) compounds have occupied a major place in organic synthesis due to their relevant properties such as low cost, ready availability, rate enhancement and regioselectivity of the reaction and their importance in biological chemistry [43]. Many useful reactions in organic synthesis have been catalyzed by copper (I) compounds such as CuI, CuBr, CuCl, CuCN etc.

Benzoxazoles are important heterocyclic molecules, which exhibit interesting biological and pharmaceutical activities [44]. Hu *et al.* [45] carried out an efficient copper (I) catalyzed direct alkylation of benzoxazole with secondary alkyl halide to synthesize alkylated product (**Scheme 1**).

4*H*-chromenes and naphthalene play a significant role in the natural bioactive

molecules and in the field of material sciences [46]. An efficient and convenient method has been developed by Beifuss *et al.* [47] for the synthesis of 4*H*-chromenes and naphthalene derivatives by the reaction of 2-bromobenzyl bromides and β -ketoester catalyzed copper (I) iodide (**Scheme 2**).

Xi *et al.* [48] developed a novel and concise method for the synthesis of benzisothiazol-3(2*H*)-ones by the reaction of O-halobenzamides with potassium thiocyanate. The reaction proceeds via tandem reaction with S–C bond and S–N bond formation. The reaction was carried out by using 1,10-phenanthroline as a ligand, DABCO as a base, Bu₄NI as an additive and water as a solvent (**Scheme 3**).

Isocoumarin derivatives are potent biologically active molecules [49, 50] and can be used as synthetic intermediates in heterocyclic synthesis [51]. Copper-catalyzed cyclization of 2-halo-N-phenyl benzamides and acyclic 1,3-diketones to synthesize isocoumarin derivatives has been developed by Yao *et al.* [52]. The reaction proceeds via a tandem sequential cyclization with C-C/C-O coupling transformation (**Scheme 4**).

An efficient and novel copper-mediated cross dehydrogenation coupling (CDC) was developed for the synthesis of β -arylamino ketones from N,N-dimethylanilines and methyl ketones. Moreover, the alkylated inoles synthesized from the N,N-dimethylanilines and free (NH) indoles in presence of TBHP (tert-butyl hydroperoxide) as an oxidant and catalyzed by copper has been reported by Huang *et al.* [53]. This reaction involves C-H bond activation and subsequent C-C bond formation (**Scheme 5**).

Aza-heteocycles are important organic molecules because they exhibit interesting pharmaceutical activities and are found in various alkaloids [54, 55]. Chiba *et al.* [56] carried out the C-C bond formation using CuBr.SMe₂ as a catalyst. It involves an aerobic intramolecular carbo- and amino-oxygenation of N-(2-alkynylaryl)enamine for the synthesis of aza-heterocycles (**Scheme 6**).

A mild, versatile and convenient method for the synthesis of 4-oxo-indeno[1,2-*b*]pyrroles from 1-(2-indoaryl)-2-yn-1-ones and isocyanides has been developed by Cai *et al.* [57]. This reaction proceeds via tandem reaction with [3+2] cycloaddition / coupling process (**Scheme 7**).

Benzimidazo[1,5-*a*]imidazoles, which incorporates benzimidazole and imidazole framework exhibits a number of biological properties and used in pharmaceutical preparations [58]. Wu *et al.* [59] developed a copper-catalyzed tandem [3+2] cycloaddition C–N coupling of carbodiimides and isocyanoacetates, leading to benzimidazo[1,5-*a*]imidazoles in good yields. This is a pioneer step for generation of other N-heterocycles, which are important key constituents of biologically active natural products and synthetic materials (**Scheme 8**).

N-aryl acridones are important structural motif due to their biological importance and drug discovery process [60, 61]. Zhou *et al.* [62] reported a copper catalyzed intramolecular direct amination of C–H bonds for the synthesis of N-aryl acridones using air as an oxidant under neutral conditions. This reaction provided an alternative method for constructing medicinally important acridones and also offers a new strategy for C–H bond amination (**Scheme 9**).

N-containing heterocyclic moieties have improved solubility and can facilitate salt formation, proving to be important in drug development. The N-fused heterocycles of pyrazoles and isoquinoline derivatives, 1*H*-pyrazolo[5,1-*a*]isoquinolines, exhibits interesting biological activities for the inhibition of CDC25B, TC-PTP, and PTP1B [63]. Fu *et al.* [64] developed a one-pot copper-catalyzed methodology for the synthesis of 1*H*-pyrazolo[5,1-*a*]isoquinolines, containing various functional groups like, halo, amino, ester, cyano and carbonyl. These groups provide opportunity for the construction of diverse biologically active molecules (**Scheme 10**).

Benzimidazoles exhibit a large spectrum of biological properties such as antiviral, antifungal, antibacterial, anti-tumor etc. [65, 66]. They are widely used as important synthetic intermediates in synthetic organic chemistry [67]. Wu *et al.* [68] achieved the preparation of 2-fluoro-alkylbenzimidazoles from N-aryltrifluoroacetimidoyl chlorides and primary amines via copper (I) catalyzed tandem reaction (**Scheme 11**).

Tetrahydroisoquinoline derivatives are found in various natural products, which exhibit various biological properties such as antitumor and antimicrobial activities [69, 70]. Wu *et al.* [71] described a diversity-oriented approach for the synthesis of 1,2-dihydroisoquinolin-3(4*H*)-imines using copper(I)chloride as a catalyst. The reaction proceeds via three component reaction of (E)-2-ethynylphenylchalcone, sulfonyl azide and amine under mild reaction conditions (**Scheme 12**).

2*H*-1,4-benzoxazin-3(4*H*)-ones are important compounds as biologically active natural and synthetic products. These are also used in pharmaceuticals, herbicides, and fungicides industries [72, 73]. A facile

and efficient approach for the synthesis of such compounds has been reported by Lv *et al.* [74] via a CuI-catalyzed cascade condensation process between 2-(*o*-haloaryloxy)acyl chlorides and primary amines (**Scheme 13**).

N-fused heterocycles constitute the core structure of the heterocyclic compounds. They display a broad spectrum of promising biological properties. For example, inhibition of acetylcholinesterase, calcium channels antagonistic activity, antifungal properties, anti-inflammatory properties, CDC25 phosphatase inhibitor activity, mGluRs antagonist properties, anti-neurodegenerative and anti-tumor activities [75-82]. A novel and synthetically efficient Cu(I) catalyzed Csp-s coupling and a sequence of 5-endo-dig cyclization reaction has been developed for synthesis of biologically important N-fused heterocycles by Li *et al.* [83] (**Scheme 14**).

The indoline structural motifs have shown promising biological activities [84]. An efficient and mild method for the synthesis of indoline derivatives from reaction of 2-ethynylarylmethylenecyclopropane with sulfonyl azide catalyzed by copper (I) iodide has been developed by Wu *et al.* [85] (**Scheme 15**).

The carbazole framework is present in a wide range of natural products and synthetic compounds with varied biological activities such as cytotoxic, antitumor, antibiotic, antiviral and anti-oxidative activities [86-89]. A novel cascade Ullmann N-arylation and aerobic oxidative C-H amidation reactions of substituted 2-bromo-9*H*-carbazole-3-carboxamides and substituted benzylamine using CuI as a catalyst has been developed for the synthesis of pyrimido[4,5-*b*]-carbazolone derivatives by Nagarajan *et al.* [90] (**Scheme 16**).

Phenothiazines are interesting structural scaffolds and extensively used as drugs (such as the promazine series), insecticides, inhibitors of polymerization, optoelectronic materials, antioxidants, paints, etc. [91, 92]. Zeng *et al.* [93] developed cascade coupling reaction of the aryl ortho-dihalides and ortho-aminobenzenethiols for the synthesis of Phenothiazines using CuI as a catalyst without any additives or ligands (**Scheme 17**).

Nitrogen-containing heterocyclic compounds, e.g., polysubstituted pyrroles are one of the most prevalent components found in numerous natural products, potent pharmaceutical drugs, and various kinds of functional materials [94-97]. Huang *et al.* [98] developed synthetic method for polysubstituted pyrroles from readily available β -enamino ketones or esters and alkynoates using CuI as a catalyst, O₂ as a oxidant and DMF as a solvent (**Scheme 18**).

Indole and its derivatives are an iconic component of numerous bioactive and natural products, and are of a potent structure in drug discovery [99, 100]. Lu *et al.* [101] developed a simple and efficient method for the synthesis of 3-functionalized indoles from the three-component reaction of indoles, sulfonyl azides and terminal alkynes. This reaction proceeds via copper-cascade catalysis (**Scheme 19**).

Reactions catalyzed by copper(II):

Copper (II) compounds play a significant role as catalyst in the synthetic organic chemistry from the point of view of both reactivity and selectivity. Copper (II) forms the most stable compounds and also possess predictable coordination geometries. Irving and Williams noted in 1953 that of all bivalent ions of the first transition period,

Cu (II) forms the most thermodynamically stable complexes [102, 103].

Oxazoles is an important class of heterocyclic compounds which are found in a wide variety of biologically active molecules [104]. Cu (II) catalyzed direct oxidative cyclization reaction to synthesize oxazoles from enamides has been developed by Buchwald *et al.* [105]. The reaction was carried out using ethyl nicotinate as a ligand, tetrabutylammonium bromide as an additive, K₂S₂O₈ as a base and acetonitrile as a solvent (**Scheme 20**).

Cu (II) triflate as an efficient and sustainable catalyst in C-H functionalization for direct mannich reaction. The reaction of 2,4-lutidine with the imine for direct α - and γ -addition of 2- and 4-alkyl azarenes to aldimines has been reported by Rueping *et al.* [106] (**Scheme 21**).

β -amino acids and aziridines are important class of nitrogen-containing compounds which are building blocks in various organic reactions and they are also found in numerous bioactive natural products and medicinal chemistry [107, 108]. A suitable method for the synthesis of β -amino acids and aziridine derivatives has been described by Chan *et al.* [109]. It involves a copper (II) catalyzed amination and aziridination of a common and readily available 2-alkyl substituted 1,3-dicarbonyl compounds with PhINTs using dichloromethane as a solvent (**Scheme 22**).

Azide-alkyne 1,3-dipolarcycloaddition reaction plays a significant role in the field of chemistry [110]. A facile and rapid synthesis of 1,4-disubstituted-1,2,3-triazoles involving a copper-catalyzed azide-alkyne 1,3-dipolarcycloaddition reaction has been described by Limand *et al.* [111]. The reaction of a wide range of an alkyl

bromide, sodium azide and terminal alkyne as starting materials and β -cyclodextrin as a phase-transfer catalyst were investigated by the group (**Scheme 23**).

The presence of fluoroalkyl groups and particularly the trifluoromethyl (CF_3) group in pharmaceutically and agrochemically important molecules change their physical and biological properties significantly [112, 113]. Buchwald *et al.* [114] developed a copper-catalyzed oxidative difunctionalization strategy for the efficient oxytrifluoromethylation of unactivated alkenes that allows rapid access to a variety of synthetically useful building blocks such as CF_3 -containing lactones, cyclic ethers, and epoxides from simple starting materials (**Scheme 24**).

Indole-2,3-diones are very important structural motifs of numerous biologically active natural compounds and pharmaceuticals. Indole-2,3-diones are also important synthetic blocks in organic synthesis [115, 116]. Li *et al.* [117] developed copper (I) chloride catalyzed intramolecular cyclization of formyl-*N*-arylformamides to synthesise substituted indoline-2,3-dione derivatives (**Scheme 25**).

Enantioselective cycloisomerisation reactions play a significant role in the asymmetric synthesis of heterocyclic structural motifs [118, 119]. Toste *et al.* [120] described copper (II) phosphate catalysed asymmetric cycloisomerisation reaction for the enantioselective synthesis of substituted furans (**Scheme 26**).

Much attention has been paid for the preparation of benzoxazoles due to their utility in medicinal chemistry. They are characterized as estrogen receptor agonists, 5-HT₃ receptor agonists, melatonin receptor agonists, HIV-1 reverse transcriptase

inhibitors and antitumor agents [121-125]. Nagasawa *et al.* [126] described a mild and efficient method involving Cu (II) catalyst for the regioselective C-H functionalisation/C-O coupling of anilides under an air atmosphere (**Scheme 27**).

Hu *et al.* [127] developed a novel, efficient and highly diastereoselective three-component reaction of aryldiazoacetate, alcohol and α,β -unsaturated carbonyl compounds (chalcone) catalysed by copper (II) complexes to synthesize furan derivatives. The reaction proceeds via Michael type addition (**Scheme 28**).

Substituted oxindoles are important heterocyclic compound, which exhibits wide variety of biological activities [128]. An efficient and simple procedure has been developed for the synthesis of substituted oxindoles using Cu(II) acetate as a catalyst by Taylor *et al.* [129] (**Scheme 29**).

Pyrrolo[2,1-*a*]isoquinolines and their derivatives are very important functional moieties present in a wide variety of biologically and chemically significant molecules [130]. A novel and efficient method has been developed for the synthesis of pyrrolo[2,1-*a*]isoquinolines by the reaction of maleimides and tetrahydroisoquinolines in presence of catalytic amount of CuBr_2 in toluene by Wang *et al.* [131] (**Scheme 30**).

A novel and efficient method has been developed for the synthesis of indolo[3,2-*b*]carbazole by the reaction of *N*-substituted amidobiphenyls using copper (II) triflate as a catalyst, $\text{PhI}(\text{OAc})_2$ as oxidant and CF_3COOH as an additive by Chang *et al.* [132]. This reaction proceeds via intramolecular oxidative C – N bond formation (**Scheme 31**).

Copper (II) acetate catalyzed amidation reaction of 2-phenylpyridine with substituted amide/amine for the synthesis of N-(2-pyridylphenyl)benzenesulfonamide/amine derivatives has been developed by Nicholas *et al.* [133]. The reaction proceed via C–H bond activation using anisole as solvent and DMSO as an additive (**Scheme 32**).

Nitrogen-containing heterocyclic compounds play an important role in the pharmaceuticals and synthons for material-based applications [134]. Chiba *et al.* [135] developed a simple and efficient copper-catalyzed procedure for synthesis of 3-azabicyclo[3.1.0]hex-2-enes and 4-carbonylpyrroles via reactions of N-allyl/propargyl enamine carboxylates under aerobic oxidation conditions (**Scheme 33**).

Propargylamines or β -amino alkynes are important class of synthetic chemistry that exhibit promising biological activities [136, 137]. Sharghi *et al.* [138] developed an efficient, one-step and one-pot three-component method for the synthesis of propargyl amines. This method involves the reaction of aldehydes, alkynes, and amines in the presence of 1,4-dihydroxyanthraquinone-copper(II) under solvent-free conditions (**Scheme 34**).

Reactions catalyzed by Copper nanoparticles:

Recently, research has been directed towards the synthesis and application of metal nanoparticles in view of their unique properties compared to the bulk metals. Among various metal nanoparticles, copper nanoparticles have received considerable attention because of their unusual properties and potential applications in diverse fields. Copper nanoparticles, in particular, being cheap, require only mild reaction conditions

to produce high yields of products in short reaction times compared to traditional catalysts and can also be recycled [139].

Tetrazoles are N-containing heterocyclic molecules widely used as building blocks in organic synthesis, material science and medicinal chemistry [140]. A novel and efficient method has been developed for the synthesis of 5-substituted 1*H*-tetrazoles using substituted benzonitriles and sodium azides in the presence of CuFe₂O₄ nanoparticles by Sreedhar *et al.* [141] (**Scheme 35**).

1,3-dipolar cycloaddition reaction continues to be one of the most studied reactions in organic synthesis under different reaction conditions. Alonso *et al.* [142] used the readily prepared copper nanoparticles to generate the substituted triazoles using various azides and alkynes in shorter reaction period (**Scheme 36**).

Since past few years, cross coupling reactions have become a major interest in the chemistry community. Panda *et al.* [143] explored the synthetic utility of copper ferrite nanoparticles for the N-arylation of nitrogen containing heterocyclic compounds (**Scheme 37**).

2-aminobenzothiazole represent an important class of heterocyclic compounds which exhibit promising biological activities, such as anti-inflammatory, anti-microbial, anti-tumour, neuroprotective and anti-convulsant [144-147]. Patel *et al.* [148] developed a novel and efficient method for the synthesis of 2-aminobenzothiazoles. This procedure has been demonstrated to be economical, simple and facile for the preparation of 2-aminobenzothiazoles derivatives from the “in situ” generated 2-halothiouras in the presence of CuO nanoparticles (**Scheme 38**).

Oxazinone derivatives are one of the most prevalent components in numerous natural products and potent pharmaceutical drugs [149]. Kumar *et al.* [150] developed an efficient and green methodology for the synthesis of naphthalene condensed oxazinone derivatives from aldehydes and urea in presence of copper nanoparticles as catalyst (**Scheme 39**).

Polyhydroquinolines have received considerable attention due to the diverse biological and physiological activities [151]. A simple, eco-friendly, green and efficient procedure has been developed for the synthesis of polyhydroquinolines from aldehydes, dimedone, ethyl acetoacetate and ammonium acetate by Safaei-Ghomi *et al.* [152]. This reaction proceeds via one pot multicomponent methodology using CuO nanoparticles under solvent-free conditions (**Scheme 40**).

Conclusion

The results sum up in this review underscore several imperative progresses that have been attained in the development of copper-catalyzed synthesis of structurally diversified heterocyclic identities. The copper catalyzed processes have accomplished a significant success in carrying out complex reactions in mild and eco-friendly conditions. The strength of copper salts with regards to reactivity, high-

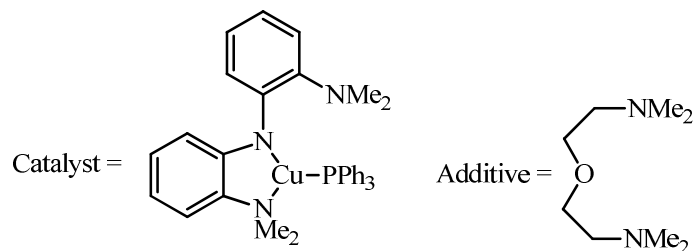
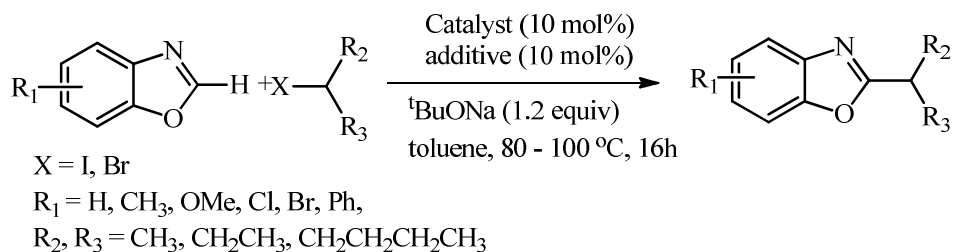
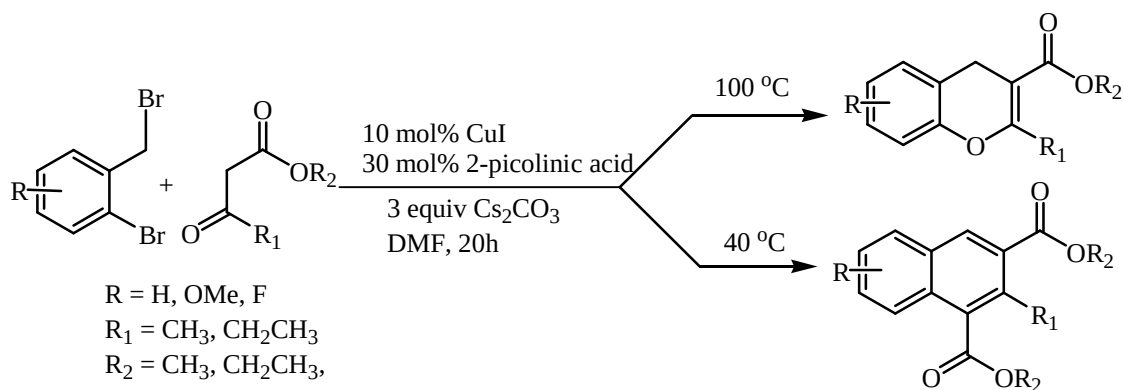
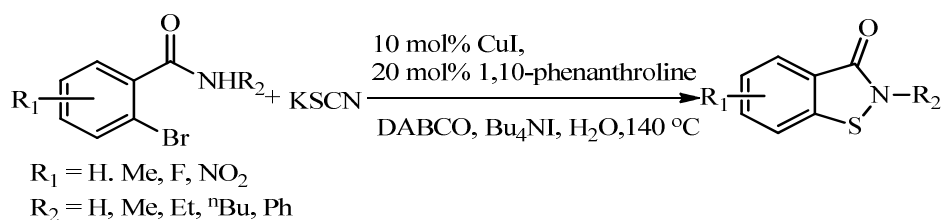
yields and selectivity in synthesis of various complex heterocyclic compounds and particularly cross-coupling reactions has been discussed. Beyond doubt, the Cu-catalyzed coupling reaction is still going to play a vital role throughout synthetic development due to the low cost and low toxicity, as well as many other merits.

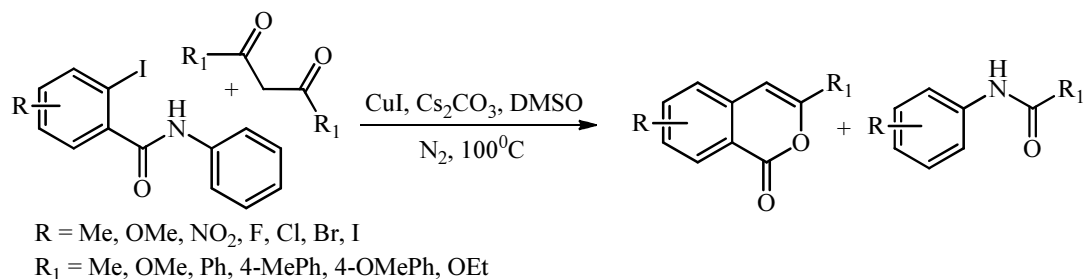
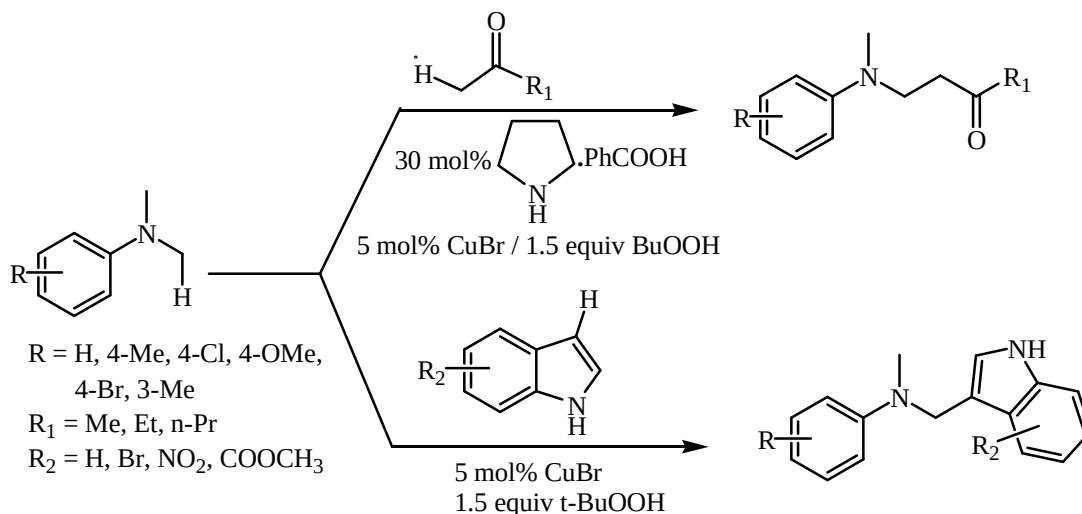
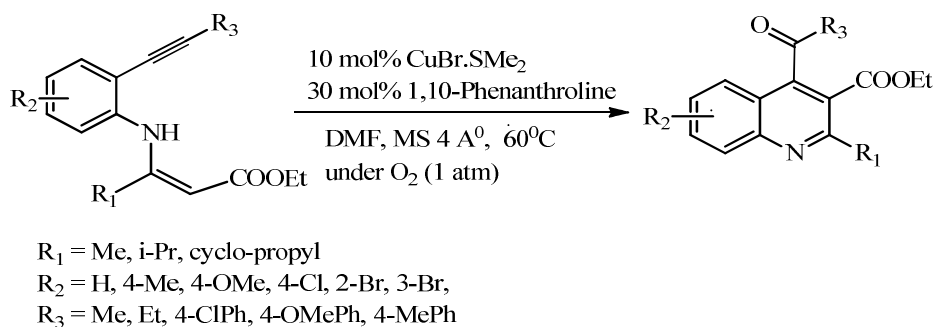
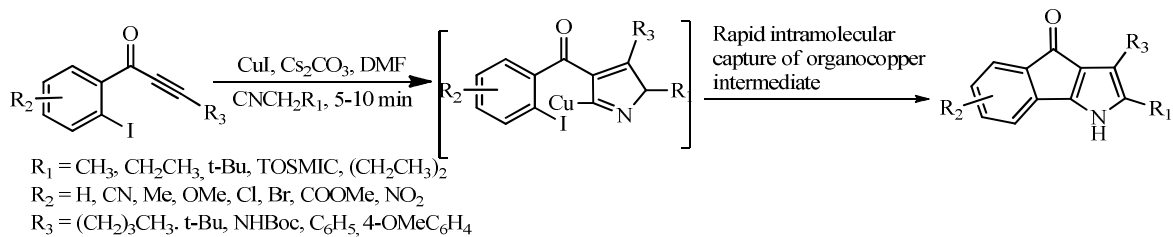
A remarkable amount of progress has been reported during the last few years by the use of stoichiometric oxidants like copper metal salts. Therefore, further developments are to be sought in copper-catalyzed aerobic oxidative dehydrogenative coupling processes. Another important opportunity is the development of stable catalytic systems and innovative catalyst recycling units from an economical and ecological point of view.

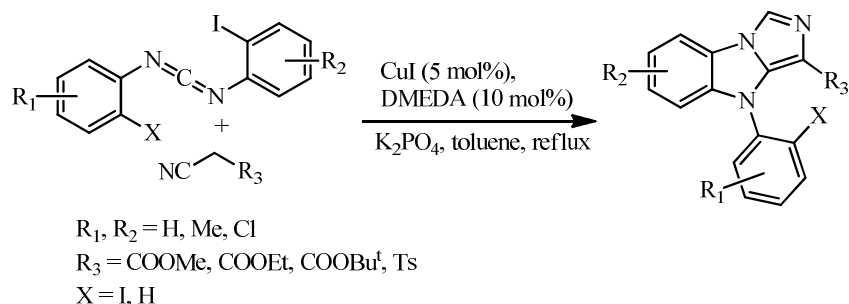
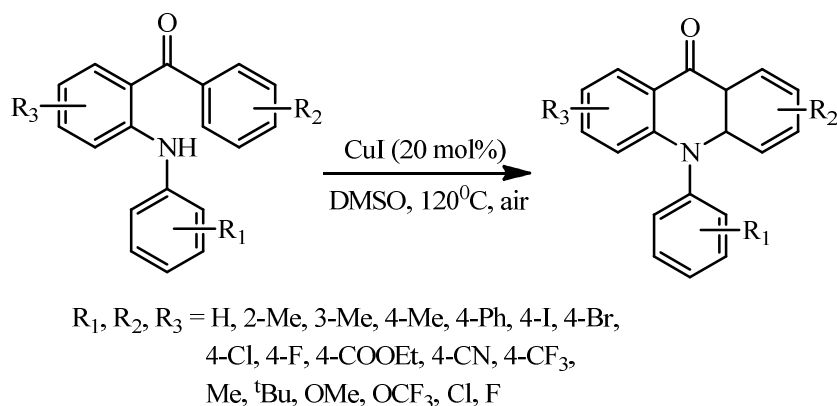
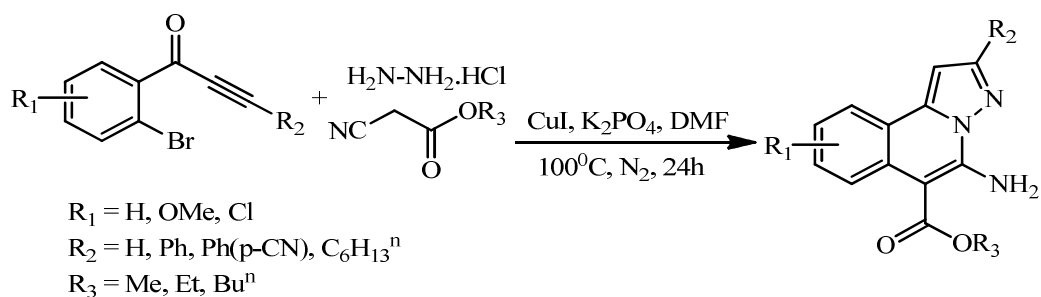
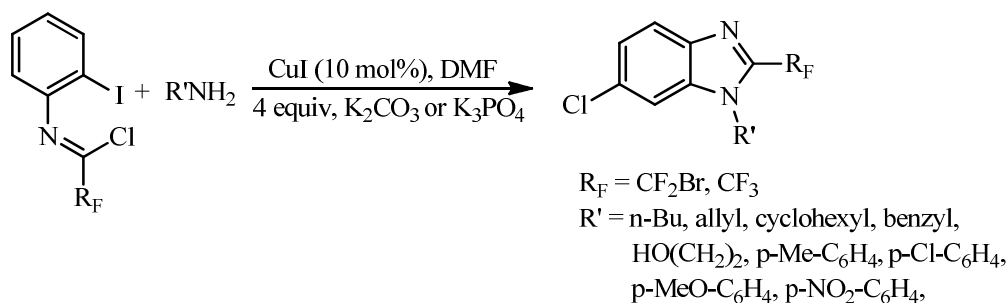
The copper salts especially copper nanoparticles can prove harbinger for many complex organic synthetic processes, under mild conditions and high efficiency achieved. The recent studies of nanoparticles indicate improvement in design and discovery of new copper catalyzed processes for heterocyclic synthesis.

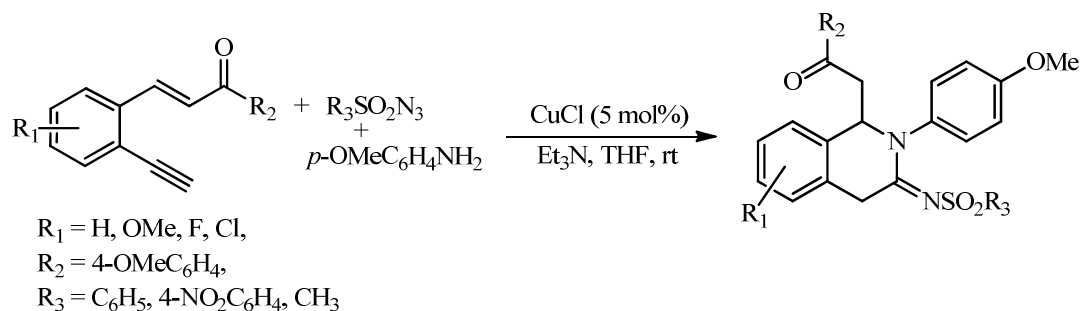
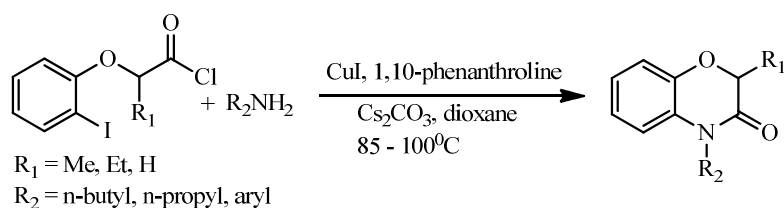
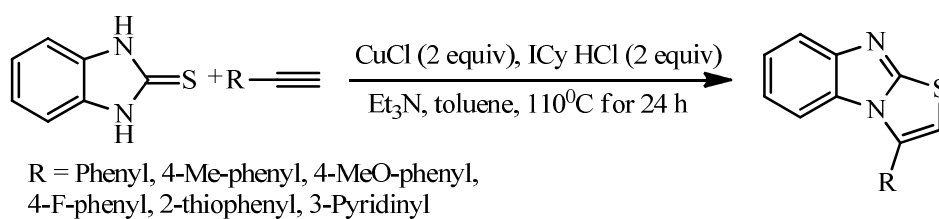
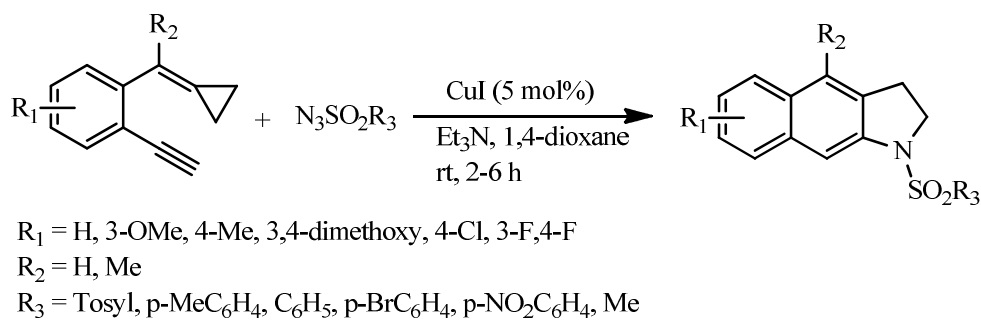
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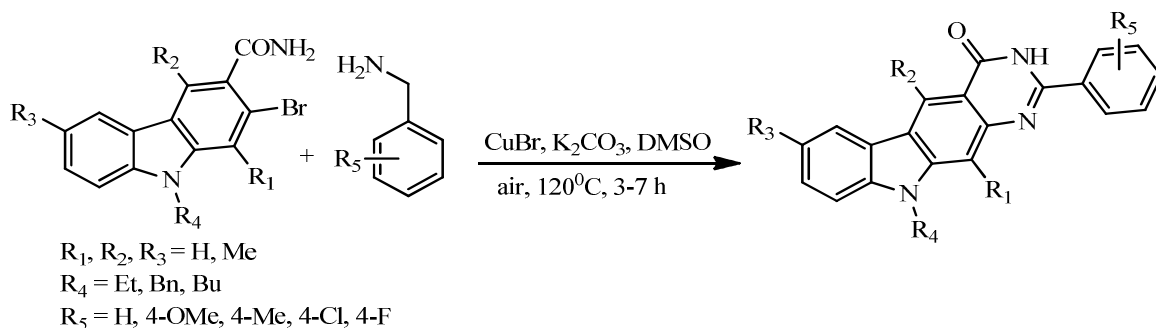
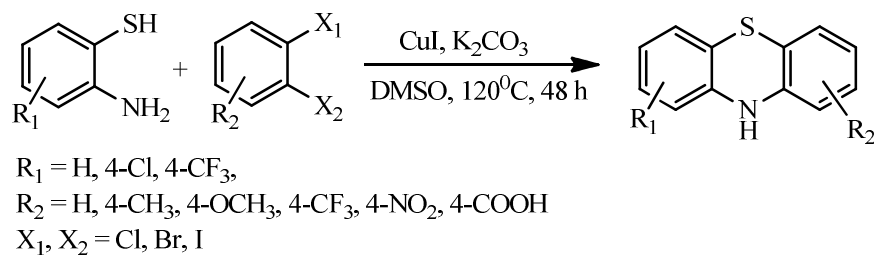
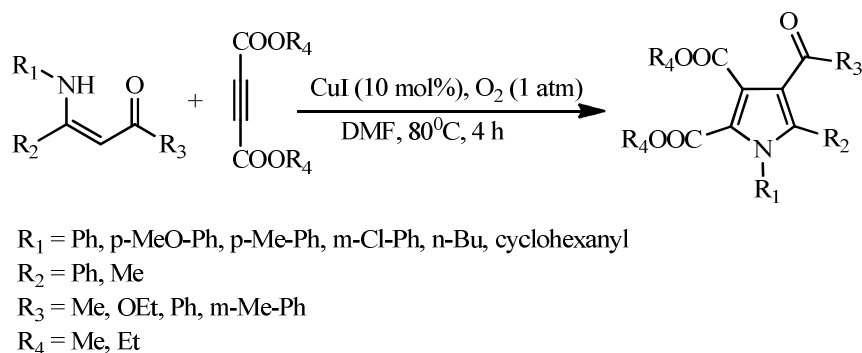
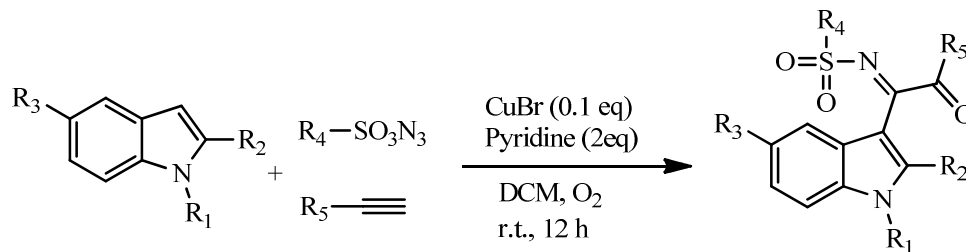
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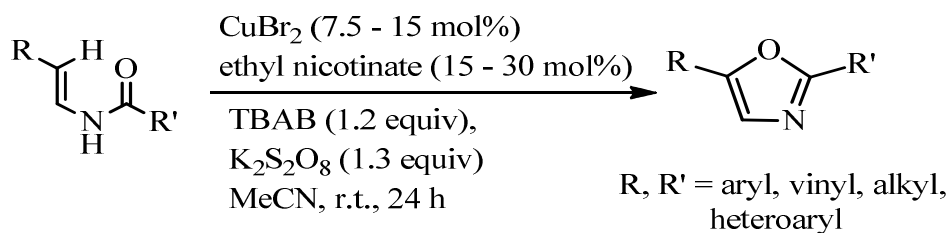
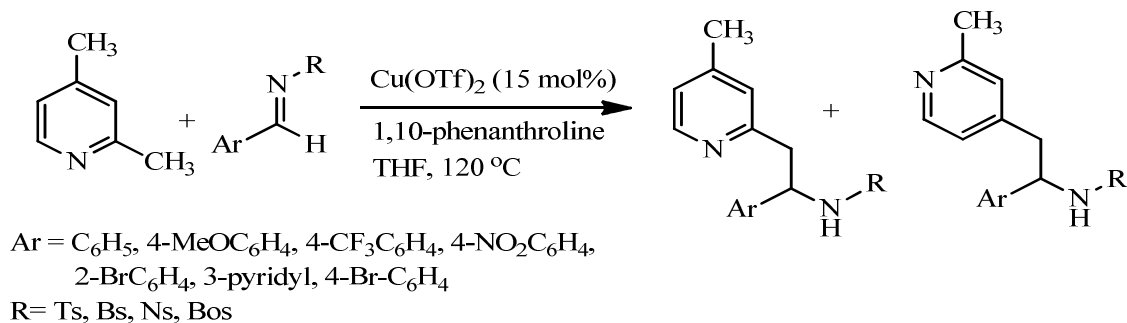
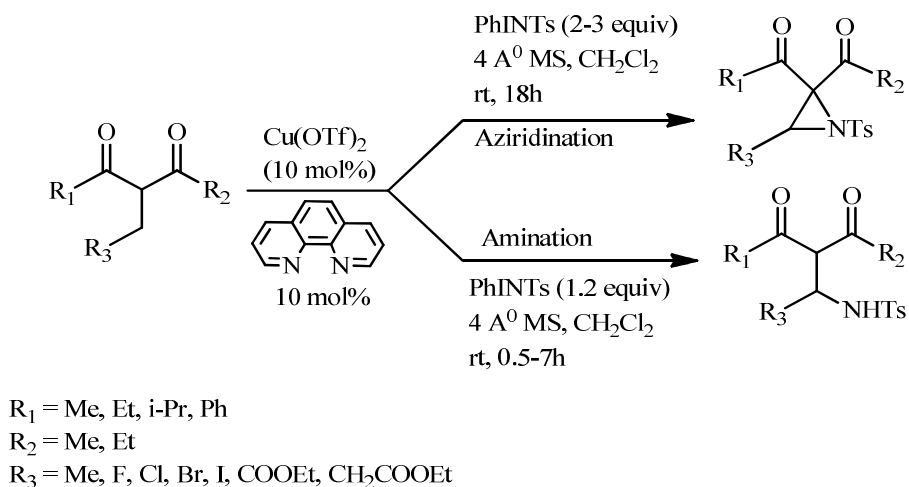
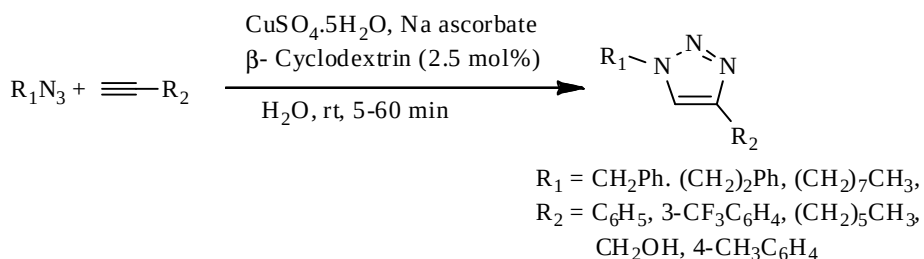
**Scheme 1.** Alkylation of benzoxazole derivatives**Scheme 2.** Synthesis of 4*H*-chromenes and naphthalene derivatives**Scheme 3.** Synthesis of benzisothiazol-3(2*H*)-one derivatives

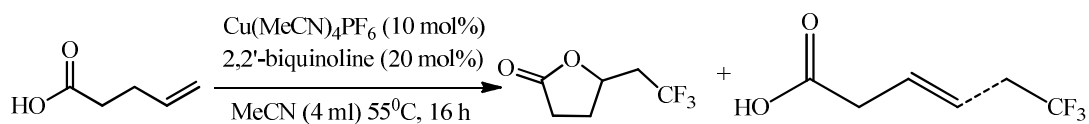
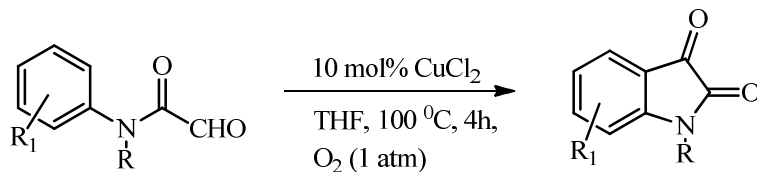
**Scheme 4.** Synthesis of isocoumarin derivatives**Scheme 5.** Synthesis of β -arylamino ketones and alkylation of indole**Scheme 6.** Synthesis of azaheterocycles**Scheme 7.** Synthesis of 4-oxo-indeno[1,2-*b*]pyrrole derivatives

**Scheme 8.** Synthesis of benzimidazo[1,5-*a*]imidazole derivatives**Scheme 9.** Synthesis of N-aryl acridone derivatives**Scheme 10.** Synthesis of 1*H*-pyrazolo[5,1-*a*]isoquinoline derivatives**Scheme 11.** Synthesis of 2-fluoro-alkylbenzimidazole derivatives

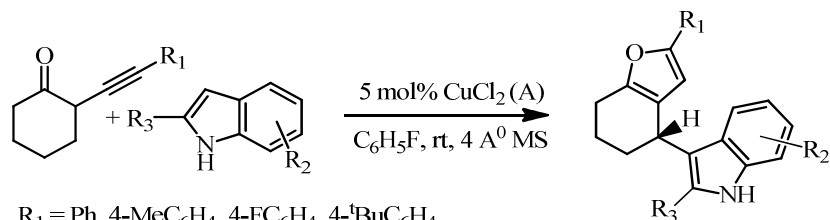
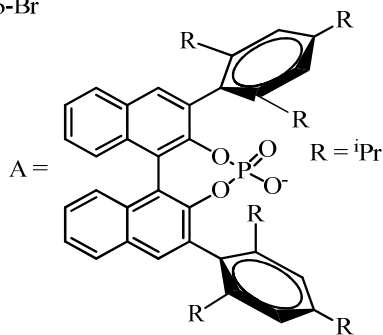
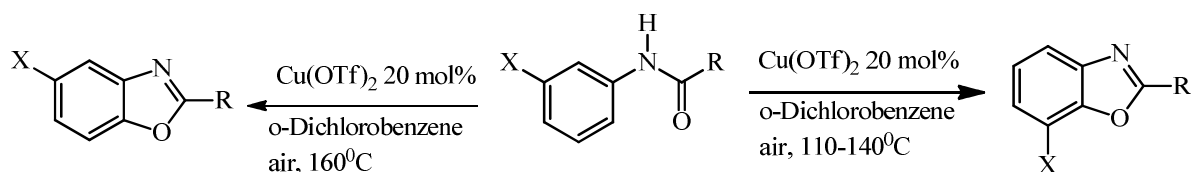
**Scheme 12.** Synthesis of 1,2-dihydroisoquinolin-3(4*H*)-imine derivatives**Scheme 13.** Synthesis of 2*H*-1,4-benzoxazin-3(4*H*)-one derivatives**Scheme 14.** Synthesis of N-fused heterocycles**Scheme 15.** Synthesis of indoline derivatives

**Scheme 16.** Synthesis of pyrimido[4,5-*b*]-carbazolone derivatives**Scheme 17.** Synthesis of phenothiazine derivatives**Scheme 18.** Synthesis of polysubstituted pyrrole derivatives**Scheme 19.** Synthesis of 3-functionalized indole derivatives

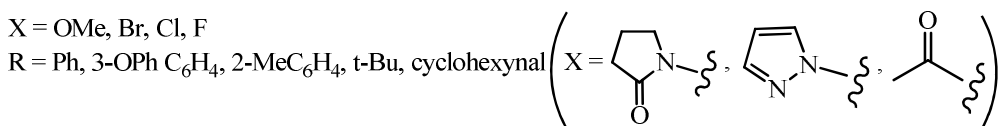
**Scheme 20.** Synthesis of oxazoles derivatives**Scheme 21.** Synthesis of aldimine derivatives**Scheme 22.** Synthesis of aziridines and β -amino acid derivatives**Scheme 23.** Synthesis of 1,4-disubstituted-1,2,3-triazole derivatives

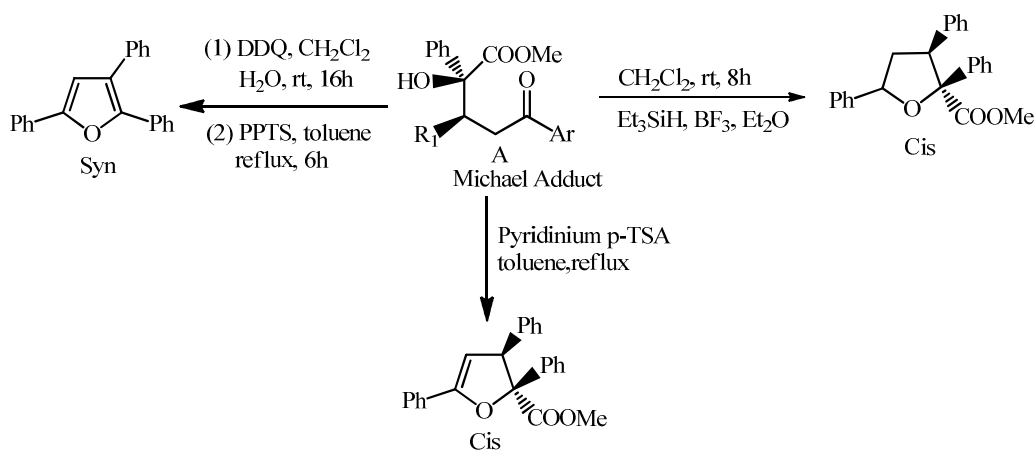
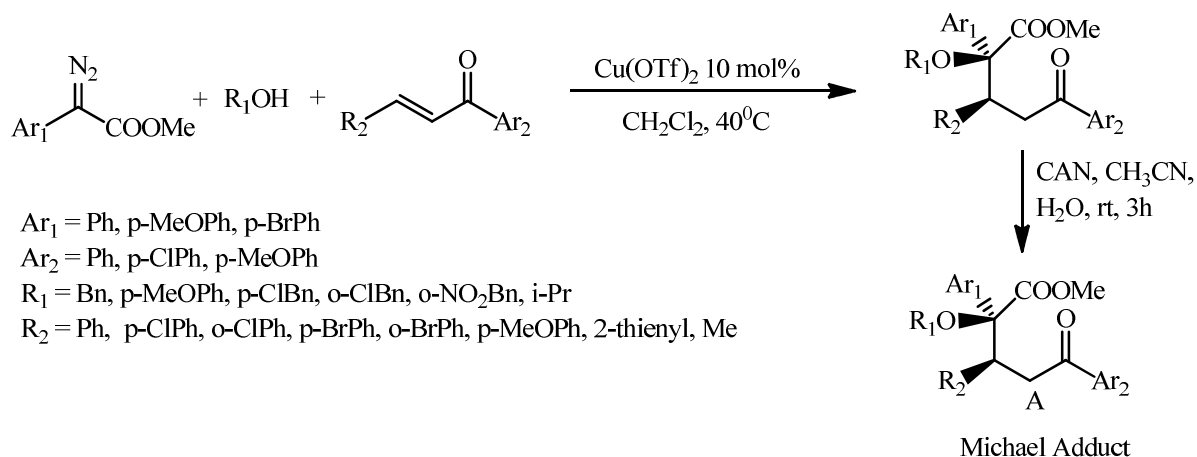
**Scheme 24.** Synthesis of CF₃-containing lactones

R = H, benzyl, allyl, methyl

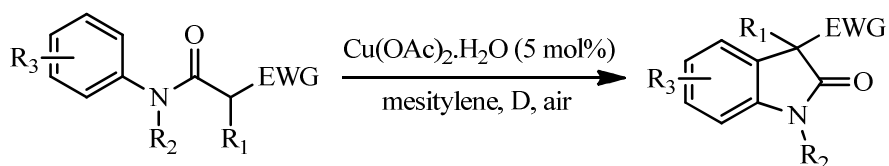
R₁ = 5-OMe, 5-F, 5-Cl, 5-OMe, 5-CF₃, 7-Me, 6-Ph**Scheme 25.** Synthesis of substituted indoline-2,3-dione derivativesR₁ = Ph, 4-MeC₆H₄, 4-FC₆H₄, 4-^tBuC₆H₄,3-MeC₆H₄, cyclopentyl, cyclohexyl, Benzyl,R₂ = H, 5-OMe, 6-Me, 5-Cl, 5-F, 6-BrR₃ = H, Me**Scheme 26.** Enantioselective synthesis of substituted furan derivatives

X = OMe, Br, Cl, F

R = Ph, 3-OPh C₆H₄, 2-MeC₆H₄, t-Bu, cyclohexynal**Scheme 27.** Synthesis of benzoxazole derivatives

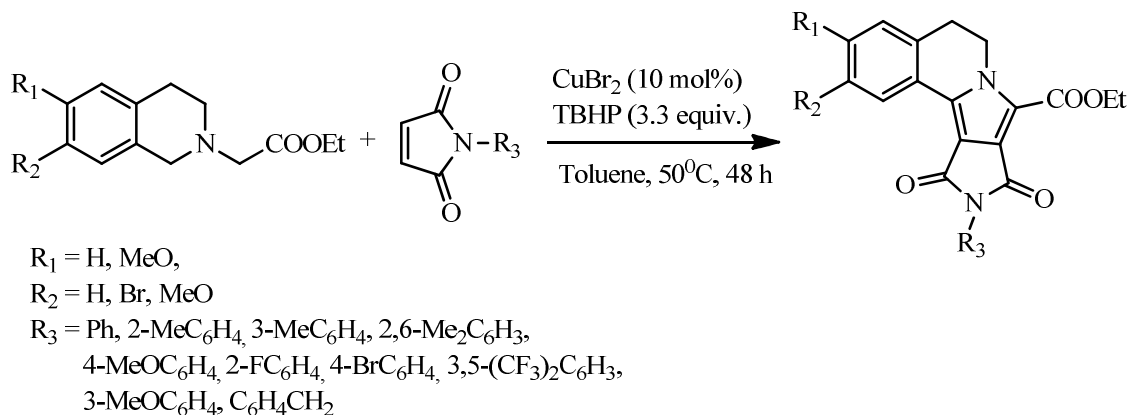
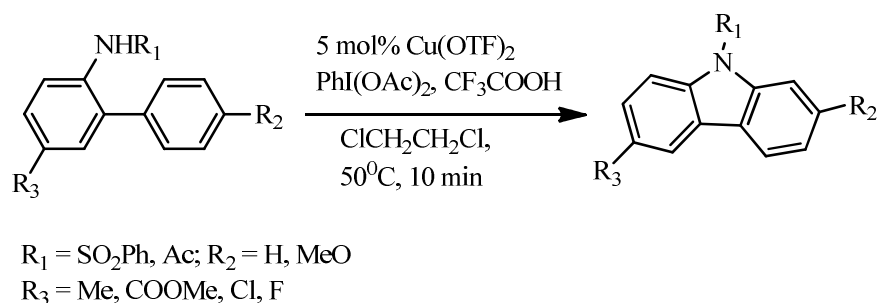
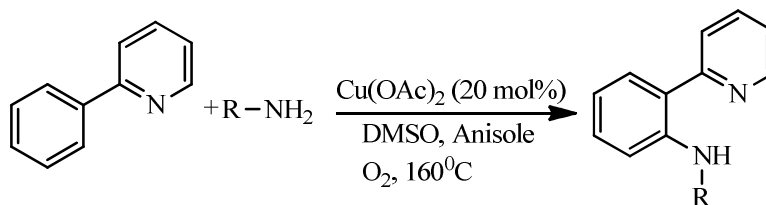
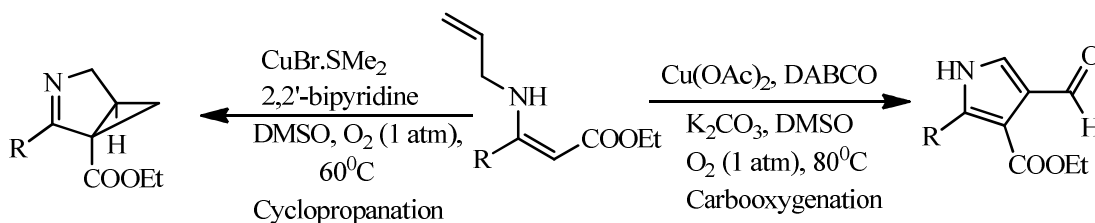


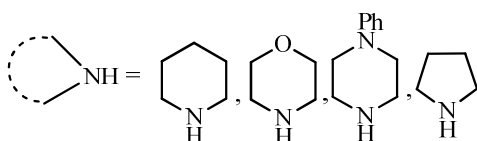
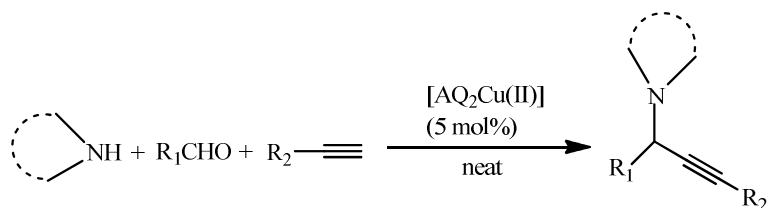
Scheme 28. Synthesis of furan derivatives



R₁ = Me, allyl, benzyl
 R₂ = Me, Bn
 R₃ = H, 5-OMe, 7-OMe, 5-CF₃, 5-COOEt,
 EWG= COOEt, COO*i*Pr, COO*t*Bu, CN

Scheme 29. Synthesis of oxindole derivatives

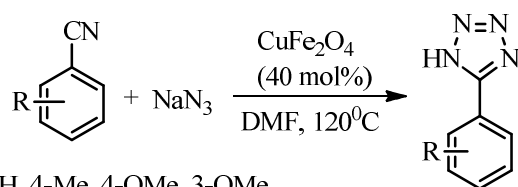
**Scheme 30.** Synthesis of pyrrolo[2, 1-a]isoquinoline derivatives**Scheme 31.** Synthesis of indolo[3,2-*b*]carbazole derivatives**Scheme 32.** Synthesis of N-(2- pyridylphenyl)benzenesulfonamide/amine derivatives**Scheme 33.** Synthesis of 3-azabicyclo[3.1.0]hex-2-enes & 4-carboxylpyrrole derivatives



R_1 = Aryl, R_2 = Alkyl

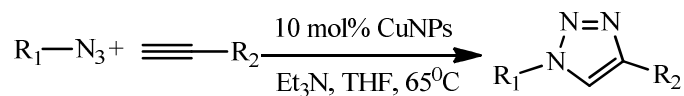
1,4-dihydroxyanthraquinone (AQ)

Scheme 34. Synthesis of propargyl amine derivatives



R = H, 4-Me, 4-OMe, 3-OMe,
4-Cl, 3-Cl, 4-Br, 3-Br, 4-CF₃

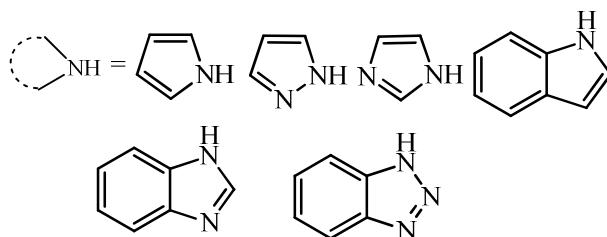
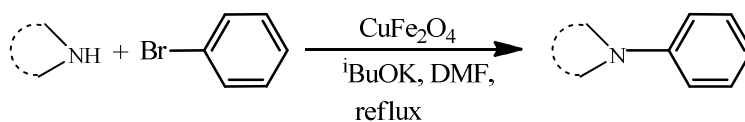
Scheme 35. Synthesis of 5-substituted 1*H*-tetrazole derivatives



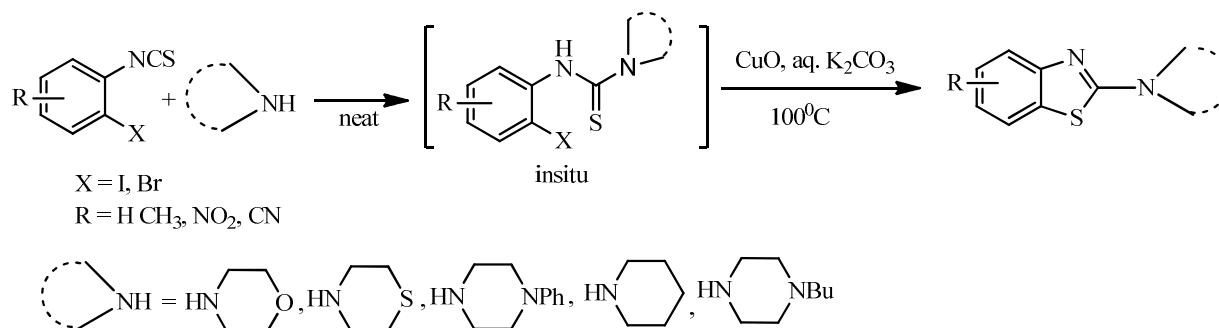
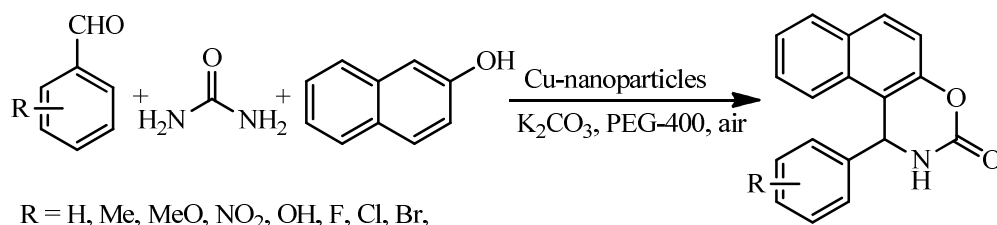
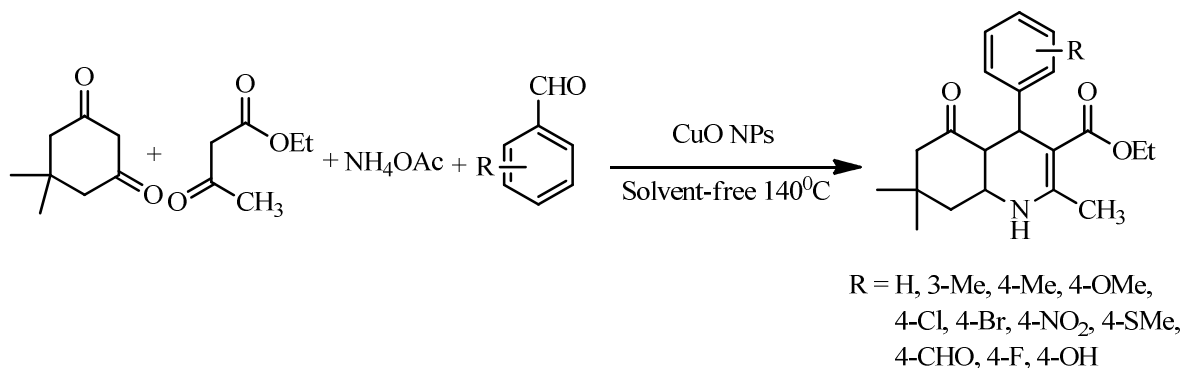
R_1 = CH₂Ph, Ph, -C₆H₁₀, CH₂COOEt,
CH₂CH=CHPh

R_2 = cyclohexanyl, CH₂Ph, CH₂OH

Scheme 36. Synthesis of substituted triazole derivatives



Scheme 37. N-arylation reaction of nitrogen containing heterocycles

**Scheme 38.** Synthesis of 2-aminobenzothiazole derivatives**Scheme 39.** Synthesis of naphthalene condensed oxazinone derivatives**Scheme 40.** Synthesis of polyhydroquinoline derivatives

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