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ADVANCES IN CHEMICAL SYNTHESIS OF QUINAZOLINE AND QUINAZOLINONES

Pratyoosh Kumar, Vishwa Deepak Tripathi*

Department of Chemistry, C. M. Science College (A Constituent Unit of L. N. Mithila University Darbhanga), Bihar
Email: vdtmkclnmu@gmail.com

Abstract: Quinazolinones and Quinazolines are considered to be most important heterocyclic molecules in the pool of biologically active heterocyclic molecules synthesized in literature. The importance is reflected by the large number of biological potential related with the same molecule. The present review is focused on the compilation of the results reported by researchers in the area of synthesis of biologically active quinazoline based heterocyclic molecules. This review is compiled by authors with the intention of summarizing the various synthetic process for the preparation of quinazolines in the order of their catalysts, reaction conditions and solvents. The ultrasound methods, microwave techniques, Aqueous medium reactions, metal based catalytic systems have been incorporated. The level of biological activity and the active functional moiety or pharmacophores are pointed out and the best active compound of each series is discussed sequentially. Authors believe that this review will be very beneficial for readers and covers the important study related to the quinazolines based heterocyclic molecules.

Introduction:

Nitrogen based hererocyclic molecules are most common nucleus present in wide range of natural and synthetic biologically important molecules. Among all the nitrogen based heterocyclic systems Quinazolines and Quinazolinones are very significant from medicinal and biological perspective.[I-III] They are well known to have anti-bacterial, anticancer, antifungal, anti-HIV, anti-inflammatory and analgesic potential. Therefore, this chemical moiety had remained the attractive choice for synthetic and medicinal chemists since last 2 decades. The number of publications related to synthesis and application of these molecules increases exponentially. A number of synthetic methodologies and protocols has been adopted by synthetic chemists to construct these heterocycles. The present chapter mainly focused on the collection of various synthetic protocols for the preparation of quinazolinones. The synthetic protocols selected to incorporate in this review chapter includes the multicomponent synthesis, use of Cu, Ru, Co and other transition metal compounds as catalysts to prepare the quinazpolinones.[IV-X]

Quinazoline heterocycle come under the class of fused heterocyclic system in which two six membered aromatic rings are condensed one is aromatic ring and other is pyrimidine ring. The physical appearance of quinazoline is mostly yellow and amorphous in nature with

molecular mass of 130-gram mol. Structurally this heterocycle is very similar to the quinoxaline, cinnoline and pthalazine depending on the position of nitrogen atom in the ring and the pattern of hetroatom. Quinazoline was first prepared by Gabriel in chemical laboratory in 1903 and the name for this hetrocycle was first given by Widdege because of it's similar appearance with the quinoxaline.[XI]

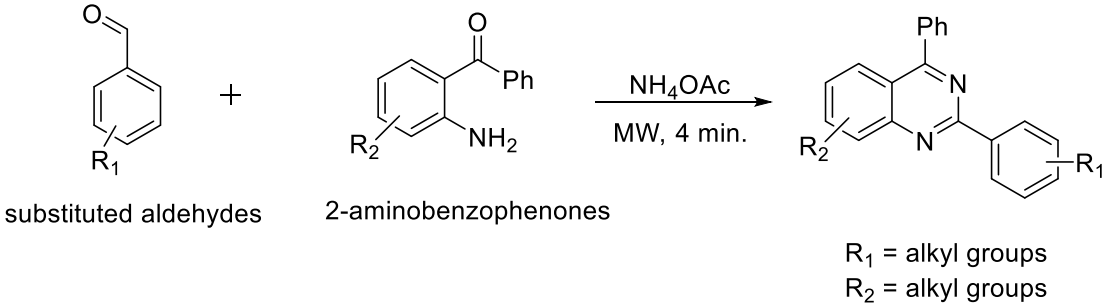
The biological potential associated with this nucleus covers a broad dimension. They are active for virus, HIV, Malarial, Inflammation, fungus, Bacteria, Spasm, Pain reliever, Cancer, Hypertension, Depression, Psychosis, Diabetes and Tuberculosis which make this nucleus very special for synthetic medicinal chemists.[XII-VIII] Several approved drug molecules contain this heterocyclic nucleus as an active moiety like Prazosin hydrochloride, doxazosin mesylate and tetrazosin hydrochloride as shown in figure **2**. In few of the reports quinazoline molecules are reported for their DNA potential and adrenergic blockers properties. In addition to this more than 150 natural alkaloids are known to possess quianzolines in their skeleton. The Fenquizone, idelalisib are drug molecules which are quinazolinone mediated compound and known to display excellent pharmacological property. [XIX-XXII]

Despite the beneficial effects of these molecules, the quinazolinones nucleus itself is a part of research for scientists in order to improve the biological and physical properties associated with the quinazolinones. To modify the original quinazoline and quinazolinone nucleus in desired way synthetic chemists continuously try link different pharmacophoric groups and moieties with the parent nucleus. Additionally, due to growing environmental constraints the need for development of new synthetic protocols is necessary. [XXIII-XXV]

A careful literature survey reveals that the 2nd 6th and 8th positions in the parent quinazoline nucleus are very crucial to affect the biological activity of this molecule. Whereas 2,3-difunctionalized quinazoline are well reported for their anti-viral and anti-hypertensive and anti-bacterial activities. Attachment of aryl and heteroaryl groups at N-3 and C-2 positions generates the analgesic and anti-inflammatory potential of the moleclules.[XXVI-XXVIII] It is observed that the presence of bridged phenyl ring, phenyl ring and heterocyclic ring as substitution enhances the antibacterial potential. The lipophilic groups at C-4 position is very crucial for inhibitory potential of the molecule. The thiadiazole rings attached with the quinazolines enhances the Potential of compounds against HIV, Bacterial infection and fungal infection.[XXIX-XXXI]

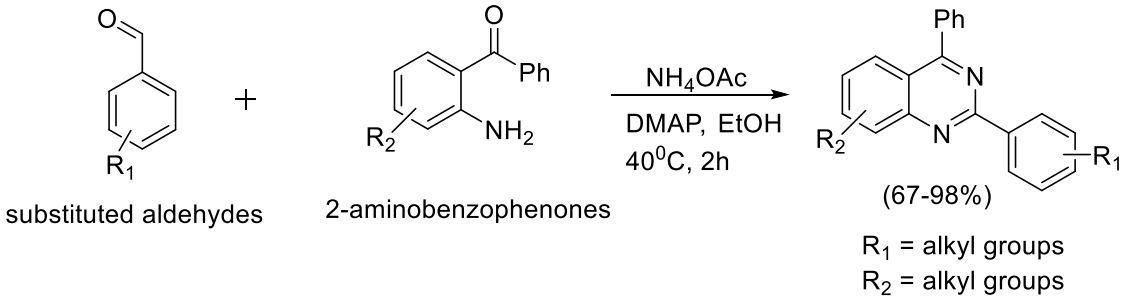
The present chapter summarizes the various ways adopted by synthetic chemists for the synthesis of quinazoline and quinazolinone frameworks and further will provide an insight regarding reaction condition and catalysts involved in the process.

A synthetic process without using any catalyst and also in absence of any solvent leading to formation of quinazoline analogues were reported by Sarma and Prajapati involving 2-aminobenzophenones, substituted aldehydes and NH_4Ac using microwave reaction conditions (Scheme **1**).[XXXII] The efficiency of their reported procedure were further supported by employing various electron donating and electron withdrawing groups. The yields reported by them were ranging from 70 to 91%.



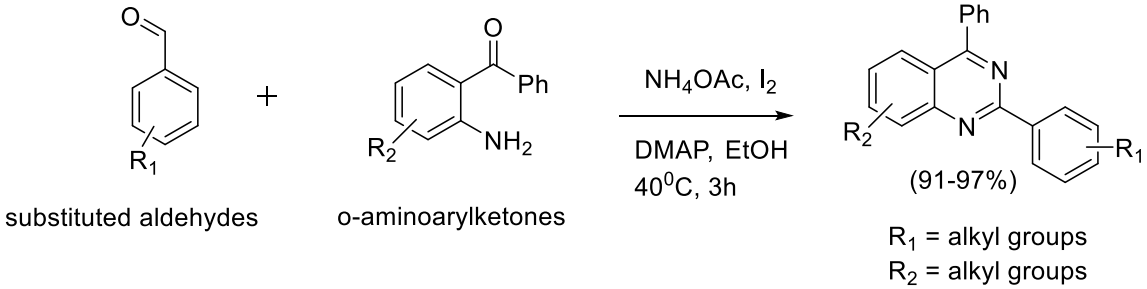
Scheme 1: Synthesis of Quinazolines under microwave irradiation

A one pot methodology has been developed by Boulcina and coworkers leading to the preparation of quinazoline analogues. The ([N,N-dimethylamino]pyridine) was used as catalyst in the suggested process involving aromatic aldehydes and 2-aminobenzophenones. The substrate scope and easy product isolation made this process very useful (Scheme **2**).[XXXIII]



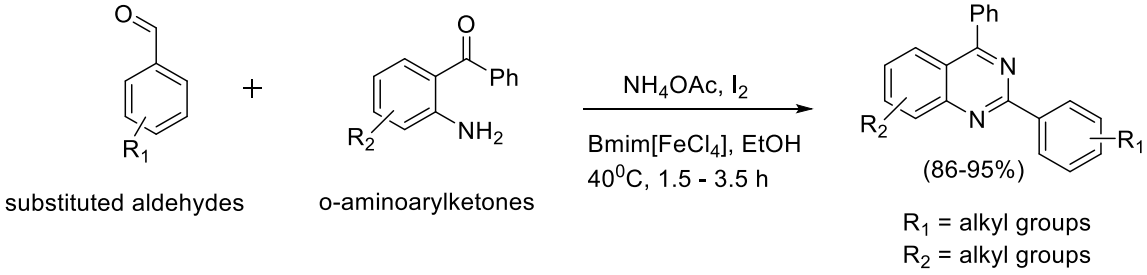
Scheme 2: DMAP mediated synthesis of Quinazolines.

An easy multicomponent approach was developed by Panja and coworkers involving three component reaction benzaldehydes and o-aminoarylketones. This synthesis was Catalyzed by Iodine and occurs in presence of ammomium acetate. The product formation was observed in EtOH or even in neat conditions. The isolated yields of synthesized quinazolinones were from 91-97% (Scheme **3**) The oxidizing behaviour and Lewis acidic nature of I_2 was reported to be one of the crucial factor behind the catalytic efficiency of the Iodine in this protocol. [XXXIV]



Scheme 3: Iodine catalysed synthesis of quinazolines.

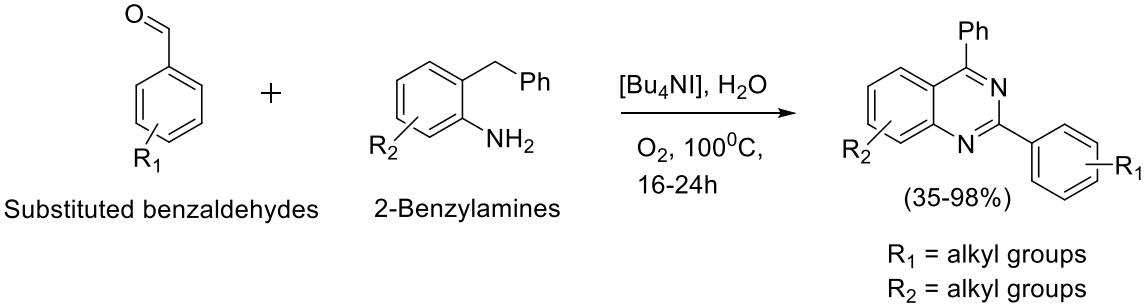
In an attempt to develop green synthetic methodology Ionic Liquid (IL) Bmim[$FeCl_4$] (butylmethylimidazolium tetrachloroferrate) was employed forth preparation of quinazoline derivatives by Panja and co-workers.[XXXV] The authors have developed an efficient, multicomponent and sustainable synthetic protocol to yield quinazolines in good yields. The aminobenzophenones and aldehydes were reacted in ionic liquid in presence of ammonium acetate at average temperatures. The recyclability of ionic liquids and stress-free isolation associated with the process make it very valuable and ecofriendly (Scheme **4**).



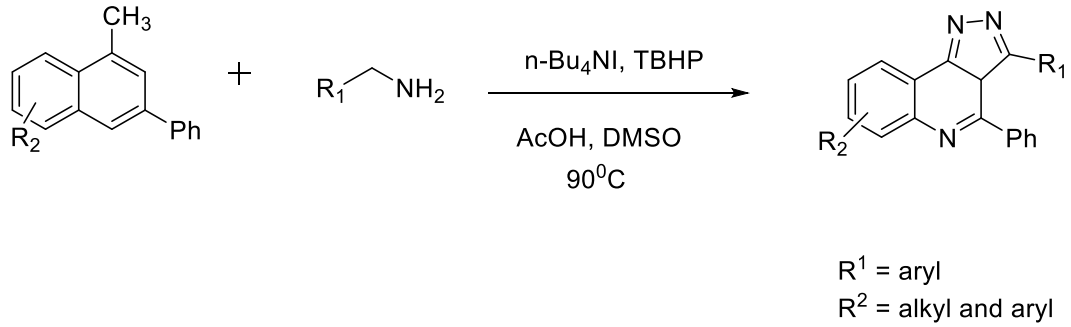
Scheme 4: Synthesis of quinazolines in Ionic Liquids.

In one of the methodology reported n-Bu_4NI was employed as catalyst resulting in the synthesis of imidazolo quinazolines. Different 2-phenylquinazolines were used as reactants, which were mixed with benzylamines to form the quinazoline frameworks in 35-98% yields.[XXXVI] The dual amination at the sp^3 hybridized carbon is the speciality associated with

this protocol. The reported methodology proves to be a valuable access the imidazo-N-heterocycle derivatives and functionalized quinazolines (Scheme **5**).



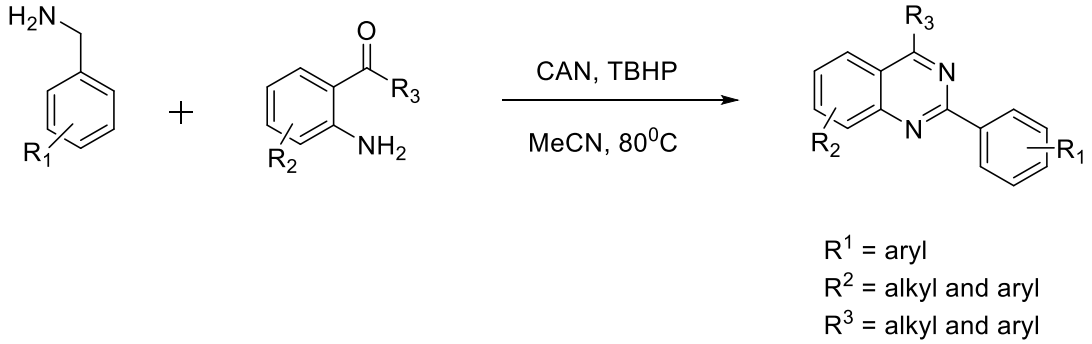
Scheme 5: Tetrabutylammonium Iodide catalyzed Synthesis of quinazolines.



Scheme 6a: Synthesis of Quinazoline derivatives.

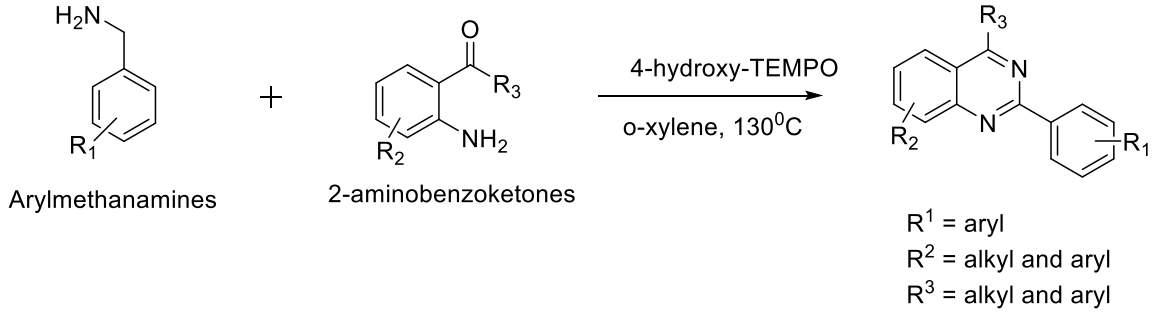
The construction of the preparation of quinazoline scaffolds was reported by Nageshwar et. al. in 75-93% yield via Ceric Ammonium Nitrate catalysed reaction of benzylamine and 2-aminobenzophenones using TBHP (tetrabutylhydroperoxide) in acetonitrile as solvent. Applying CAN/TBHP system for this synthesis proved be very efficient and new for synthesis of these scaffolds (Scheme 6a). [XXXVII]

During the synthesis it was observed by authors that the decreasing the electron density in phenyl ring by attaching the electron attracting group at the 4th point of benzylamines increased the conversion rate, but electron-donating group exerts the opposite effect. (Scheme **7a**)



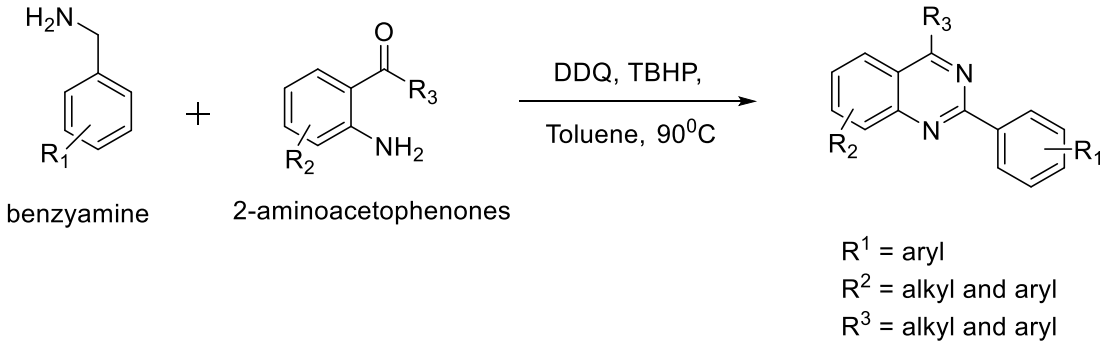
Scheme 7a: CAN/TBHP mediated synthesis of Quinazoline derivatives.

An oxidative aerobic condition was employed resulting in preparation of aryl analogues of quinazoline was reported by Han and co-workers was proved to be very efficient. [XXXVIII] The philosophy of one pot chemistry has been applied for this work. The amination was done at Benzyl C-H Bond involving radical catalysis. The reaction sequence in the suggested work involves the reaction of amino aldehydes and amino ketones with methanamines (Scheme 6b). The chemistry in absence of any catalyst make this process more valuable.



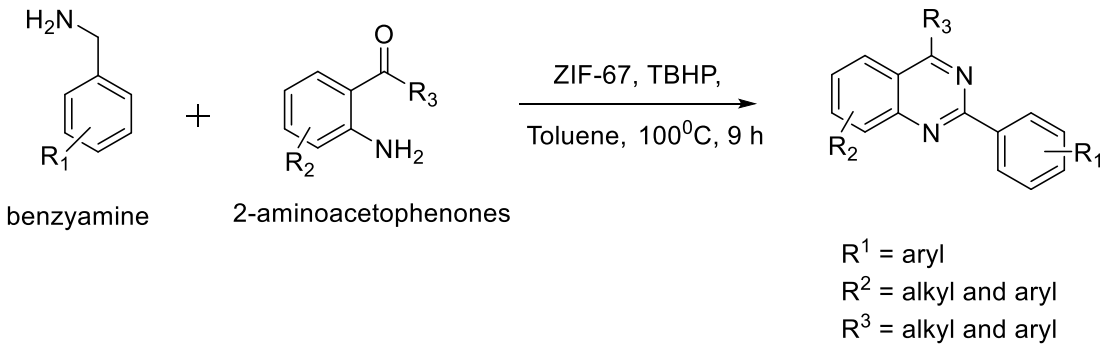
Scheme 6b: TEMPO mediated synthesis of quinazolines.

Rachakonda and his group has proposed an efficient method of preparing 2-arylquinazolines in metal free conditions using benzyamine derivatives and 2-aminoacetophenones as reactant using reagents for aromatization such as Dichloro dicyano benzoquinone as reagent. The condensation followed by cyclisation's one of the plausible mechanistic pathway for this type of reaction. The 2-arylquinazolines synthesized via this process were prepared in 71-92% yield (Scheme **7b**)[XXXIX]



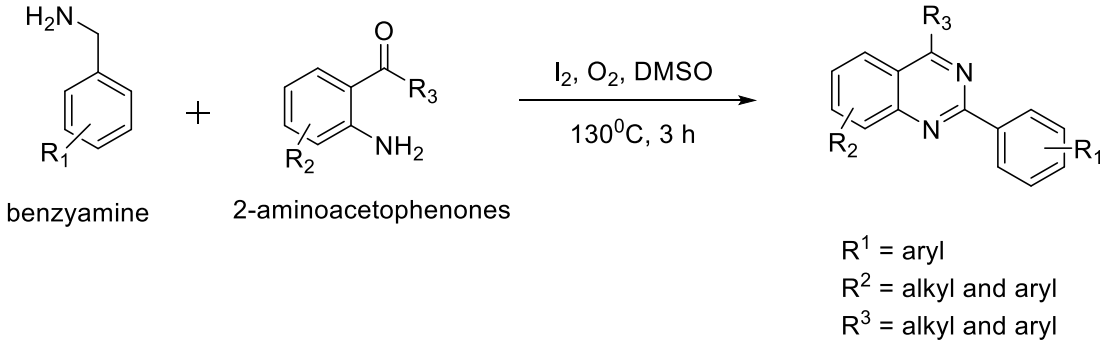
Scheme 7b: DDQ mediated synthesis of quinazoline derivatives.

In one of synthesis reported by Truong et. al. the Cobalt Zeolite Imidazole Framework (ZIF-67) was investigated as catalyst for the synthesis of quinazoline derivatives. This hetrogenous catalytic system was used in the cyclisation of benzylamines with aminobenzophenones.[XL] The TBHP was used as oxidant with toluene as solvent at 100^0C. The catalyst recovery after reaction made this protocol more cost efficient also.



Scheme 8: ZIF-67/TBHP mediated synthesis of quinazoline analogues.

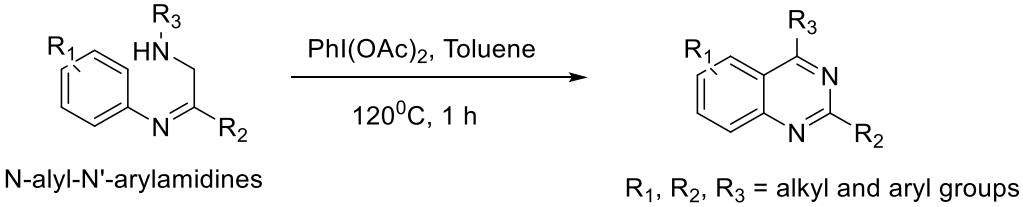
In one of the attempt for the synthesis of quinazolines involves the use of molecular iodine in amination reaction at the C-H centre. The process was conducted by using air oxygen as oxidant.



Scheme 9: Molecular iodine mediated synthesis of quinazolines.

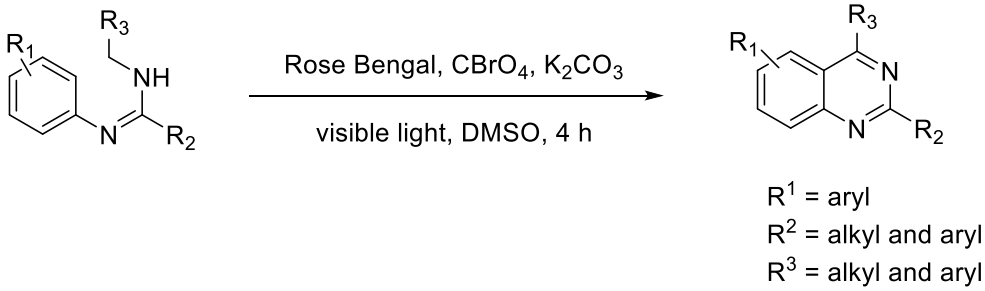
This synthesis involves reaction of amino substituted benzaldehydes or amino substituted benzophenones with benzylamines to afford the quinazoline derivatives in 50-93% yield. The oxygen gas works as natural oxidant in this process to make it more eco-friendly (Scheme **9**).[XLI]

In a work involving the formation of C-N bond and also C-C bond with the help of iodine based catalyst was proposed by Lin and co-workers results in formation of quinazoline based heterocycles. They proposed the formation of various substituted quinazoline at different positions via mixing of N-alyl-N'-arylamidines involving Iodine (III) mediated oxidative coupling in a base and metal free conditions. The substrate variation has been introduced in the protocol by introducing substrates with all types of substitutions like electron rich and electronically poor. It was observed that electron deficient groups tend to decrease the product yield in this process (Scheme **10**).[XLII]



Scheme 10: Iodine (III) mediated synthesis of quinazoline derivatives.

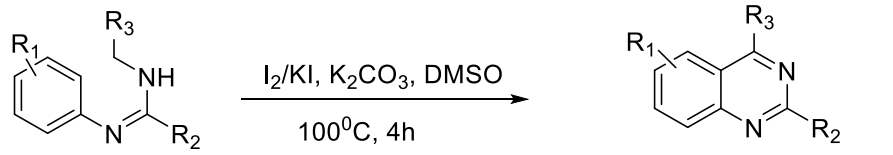
In one of the report visible light was employed in a photo redox oxidative annulation reaction leading to the synthesis of quinazoline analogues. The quinazolines derivatives were formed by the visible light catalysed oxidative coupling sp^2-sp^3. This route provides an easy and metal free access towards photo-redox synthesis of quinazolines (Scheme **11**).[XLIII]



Schme 11: Visible light promoted synthesis of quinazolines.

Lv and coworkers described a new methodology for synthesis of quinazoline analogues involving potassium Iodide mediated process through the formation C-C linkage leading to quinazoline nucleus. [XLIV] The synthesis was based on the ring closure reaction of N,N'-difunctionalized amidines. Under reaction conditions N,N'-difunctionalized amidines converted into corresponding quinzoline analogues in good to excellent yields (38-95%) as

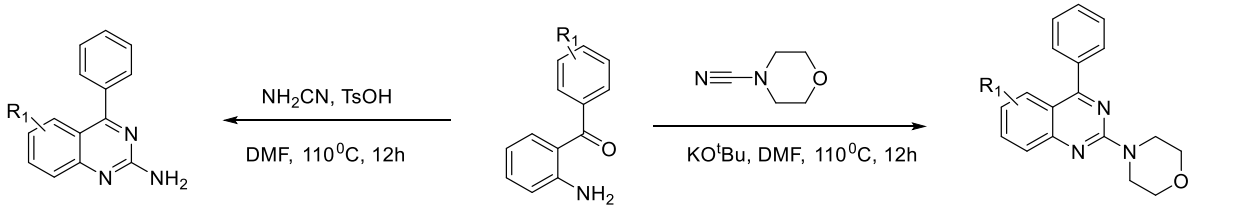
shown in (Scheme **12**). This synthetic protocol presents an environmentally benign and easy process for the synthesis of reported heterocycles.



R^1 = aryl
R^2 = alkyl and aryl
R^3 = alkyl and aryl

Scheme 12: I_2/KI promoted oxidative synthesis of quinazolines.

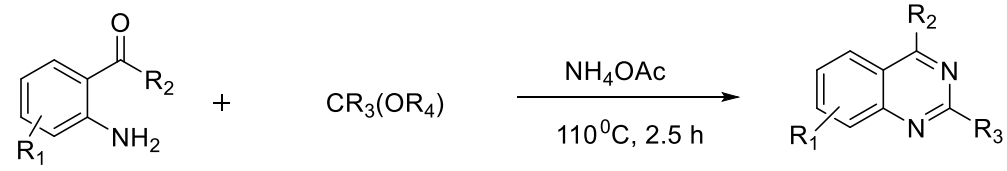
The annulation reaction using cyanamide or Carbonitrile was proposed from the group of North which showed the generation of quinazolines in moderate to excellent yields. Their reported protocol involves the mixing of amino substituted benzophenones and morpholine analogue of carbonitrile. Their methodology is a metal free and easy route for the construction of quinazoline nucleus. [XLV] The synthesized 2-aminoquinazolines are also very important (Scheme **13**).



2-aminoquinazolines — 2-aminobenzophenones — 2-morpholino quinazolines

Scheme 13: Cyanamide mediated synthesis of 2-aminoquinazolines

An Orthoester-catalyzed process was reported from the group of Bhat and coworkers as an environment friendly alternative for synthesis of quinazolines. The synthetic process involves one pot reaction of aminoarylketones and trialkyl orthoesters with ammonium acetate. This reported process benefitted with the high product yield and easy product isolation method. The yield of isolated products was 79-93%. The less reaction time and room temperature for the reaction made it more valuable from the environmental perspective. Authors proposed and important route for the construction of biological significant heterocycles with high product yield and eco-friendly pathway. This methodology does not involve ant hazardous metal catalysts (Scheme **14**).[XLVI]



aminoarylketones — trialkyl orthoesters

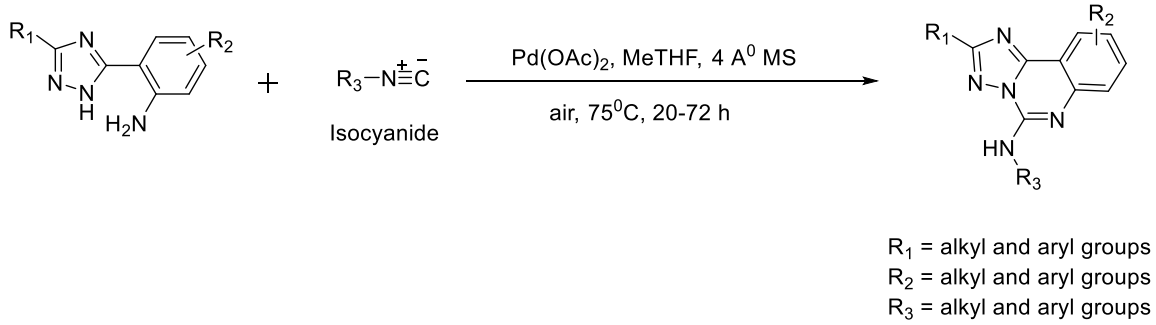
Yield (79-93%)

R_1 = alkyl and aryl groups
R_2 = alkyl and aryl groups
R_3 = alkyl and aryl groups

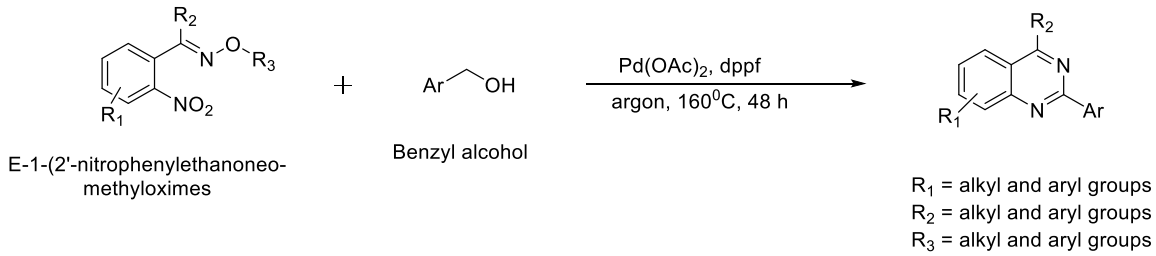
Scheme 14: Orthoester mediated synthesis of Quinazolines.

During last two decades the C-H activation based synthesis has attracted the attention among synthetic organic chemists.[XLVII-LI] Floods of research articles has been published since 2005 on the C-H activation involving synthesis of heterocyclic molecules. Among the various heterocycles synthesized via C-H activation quinazolines and quinazolinones important one.

The C-H activation synthetic routes involve the use of transition metal and organometallic catalysts. The first synthesis was reported in 1960 and was under the organometallic chemistry. The advantages associated with C-H activation synthetic methodology are the short reaction time, easy construction fused heterocycles and functional group tolerance.



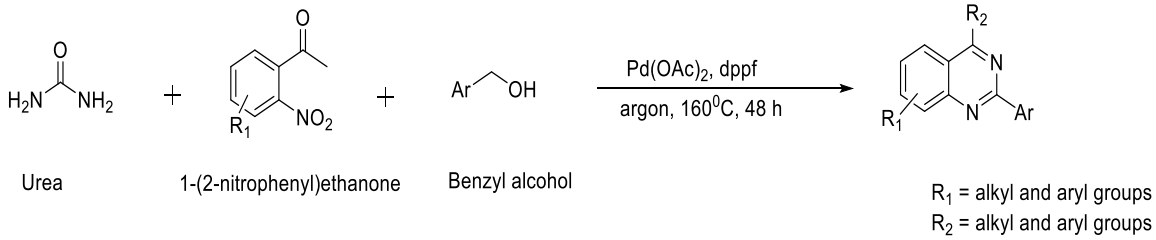
Scheme 15: $Pd(OAc)_2$ mediated synthesis of fused quinazolines.



Scheme 16: Palladium(II) mediated synthesis of quinazolines.

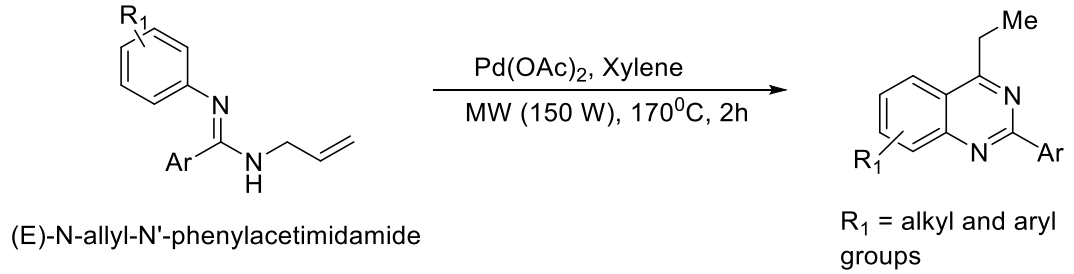
In this direction a palladium(II) acetate mediated synthesis of fused quinazolines was proposed by Vlaar and co workers in 2014. This synthetic route was comprised of aerobic oxidative coupling of aminophenyl azole derivatives and isocyanides. The product azole fused quinazolines were found to biologically active (Scheme **15**).[LII] The isolated yield of the synthesized derivatives was ranging between 12-37%. Wang et. al. also suggested palladium mediated synthesis of 2-arylquinazolines. They achieved the quinazoline nucleus by the reaction of E-1-(2'-nitrophenylethanoneo-methyloximes and benzyl alcohols. [LIII] The reaction was believed to be proceed through the hydrogen transfer approach (Scheme **16**).

Another synthesis of in efficient way was reported involving benzyl alcohol, urea and 1-(2-nitrophenyl)ethanone using palladium catalysis under the argon atmosphere. The products formed in this synthesis was in 25-90% yields (Scheme **17**).[LIV]



Scheme 17: Synthesis involving use of urea.

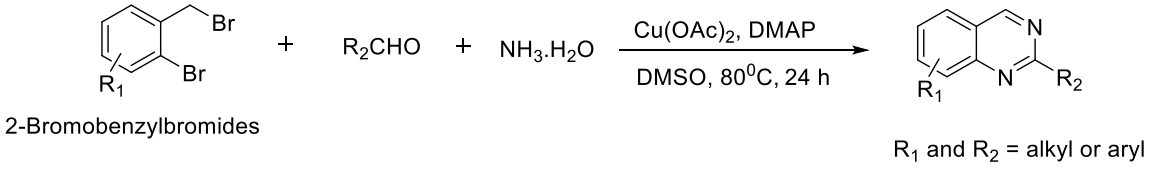
Chen and coworkers in an interesting work reported the formation of disubstituted series of quinazoline analogues. Their process involves the application of palladium based catalyst by reacting phenylacetimidamide in the influence of microwave irradiation at 170^0C using Xylene solvent. This process afforded the respective quinazoline derivatives in fair yields (Scheme 18).[LV]



Scheme 18: Palladium catalyzed synthesis of quinazolines under microwave irradiation.

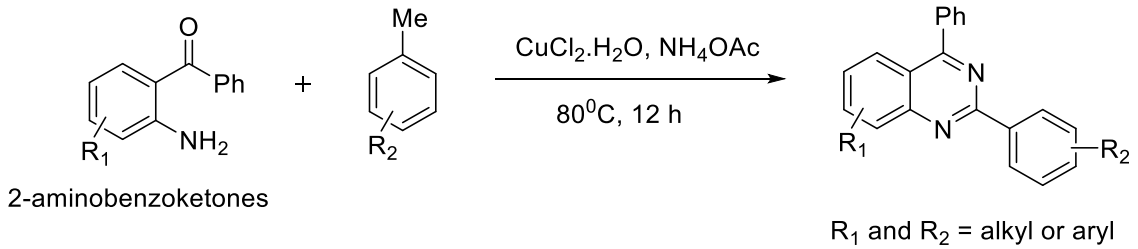
The use of copper based molecules is very popular as metal catalysts among the synthetic chemists for the construction of various fused heterocycles. Application of copper based metal catalysts are explained by researchers in the formation of nitrogen based heterocyclic molecules. During last decade the tremendous enhancement is observed in the publication related to the use of copper based molecules as metal catalysts. Employment of copper based salts in the cross coupling reactions for the synthesis of bioactive heterocycles is very popular among the scientists. The functional group tolerance, less reaction time, cheap costs and easy work up procedure make theses catalysts even more favourable for the construction of nitrogen based heterocyclic molecules. Cross coupling reaction with copper catalysts has been utilized for construction of new synthetic routes to access fused heterocycles.[LVI]

An efficient and fast synthetic methodology of diversely functionalized quinazolines was reported by Fan and co-workers in 2014. Their reported synthetic process involves copper catalysed reaction of 2-bromobenzyl bromide analogues and aldehydes in presence of ammonium hydroxide. The explained protocol is said to occur via Cu(II) acetate mediated amination of 2-bromobenzyl bromides and subsequent condensation with aldehydes followed by cyclisation and aromatization (Scheme **19**). The utility of this protocol were also explored using the ammonium as nitrogen source and gave excellent results.[LVII]



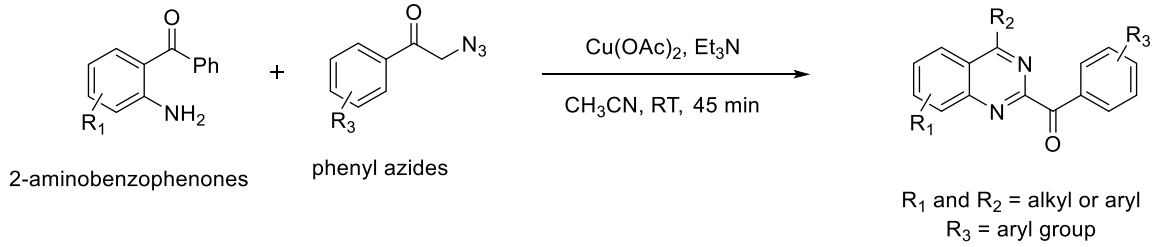
Scheme 19: Cu(II) mediated synthesis of quinazolines.

Similarly, one synthetic approach to prepare quinazoline derivatives were proposed by Liu et. al. They reported synthesis of substituted quinazoline by reacting 2-aminobenzoketones and toluene, ammonium acetate via a copper chloride catalysis. The reaction sequence involved in process are the amination of carbon hydrogen bond at benzylic position of in methylarenes with amino benzoketones in presence of air. The cyclisation leads to formation of substituted quinazoline analogues (Scheme **20**).[LVIII]



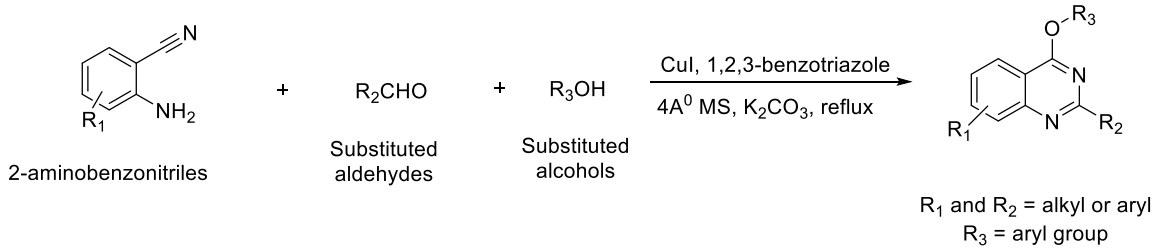
Scheme 20: $CuCl_2$ mediated synthesis of substituted quinazoline analogues.

In another report Kamal and coworkers have shown that the quinazolines can be prepared by using reacting 2-aminobenzophenones and phenyl azides. The reported synthetic procedure is cupric acetae catalysed in presence of triethyl amine in acetonitrile as solvent. The absence of oxidant and any external source of nitrogen this procedure presents an easy and better alternative for the synthesis of quinazolines as depicted in (Scheme **21**).[LIX]



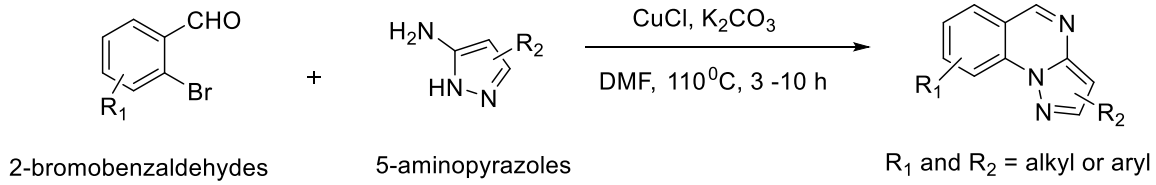
Scheme 21: Synthesis of substituted quinazolines with phenyl azides.

Similarly, an effective synthetic methodology to prepare o-protected-4-hydroxyquinazolines have been reported by Vishwakarma and co workers. They proposed a copper catalysed reaction of amino substitution at benzonitriles, aldehyde analogues and alcohol functionality. Reaction is believed to occur via formation of N-functionalized bicyclic precursors and nucleophilic attack of alkoxy group. The wide range of substrate variation have been incorporated with incorporating three variation as R_1, R_2 and R_3 which are alkyl and aryl groups. The isolated yields of products were among 43-89% [LX]



Scheme 22: Formation of o-protected quinazoline derivatives via Copper based catalysts.

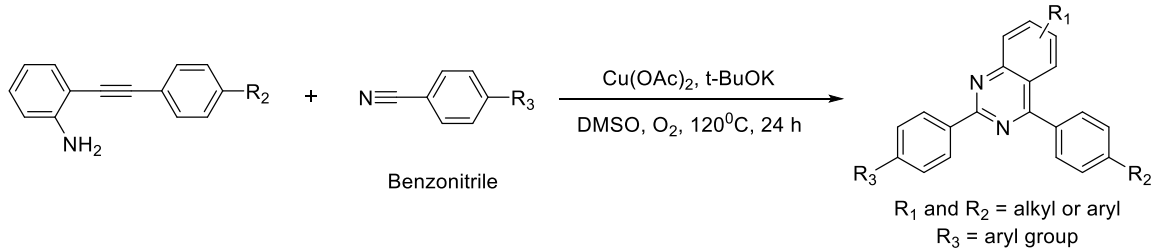
In one of the report Gao and coworkers have shown an efficient and direct methodology leading to generation of pyrazolo substituted quinazolines in single step fashion. They reported the synthetic procedure involving reaction of 2-bromobenzaldehydes and 5-aminopyrazoles by using base like potassium carbonate via Cu mediated imine formation. Later the Ullmann coupling leads to the formation of desired quinazoline derivatives. The substrate variation in 5-aminopyrazoles and 2-bromobenzaldehydes have been incorporated to check efficiency of the reported protocols (38 compounds prepared). The mild reaction conditions and easy product isolation are main advantages associated with this synthetic protocol. [LXI]



Scheme 23: CuCl mediated generation of pyrazole quinazoline library.

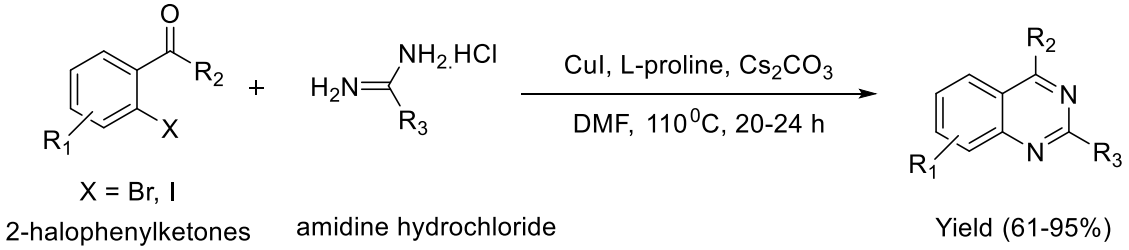
Substituted quinazolines were synthesized by Wang and co workers via Copper catalysed synthesis involving reaction of 2-ethylanilines and benzonitriles.[LXII] The molecular oxygen O_2 was used as sole oxidant in above reported protocol. The above mentioned reaction

was proceeded by cleaving the triple bond which forms the C-C bond and C-N bond effectively. Moreover, this protocol exhibited a wide range of substrate solvent in ethynylanilines and benzonitriles. The final quinazolines were isolated in 52-86% yields (Scheme 24).



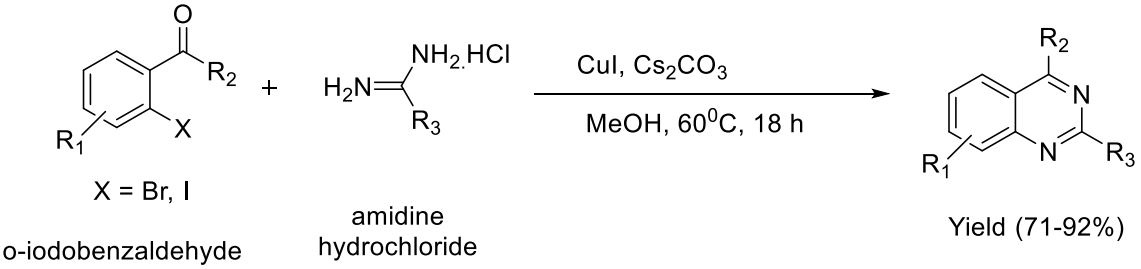
Scheme 24: Synthesis of highly functionalized quinazoline derivatives.

The coupling reaction of 2-halophenylketones and amidine hydrochloride was utilized for the preparation of quinazoline derivatives by the research group of Fu and co-workers. [LXIII] The methodology reported is a Cu catalysed coupling reaction which produces the quinazoline derivatives in high yield upto 95%. The yield of isolated product was found to be very similar with aromatic as well as aliphatic substrates. The cost effective and simple nature of this synthetic protocol made it more valuable to access the highly functionalized quinazoline analogues (Schem **25**).



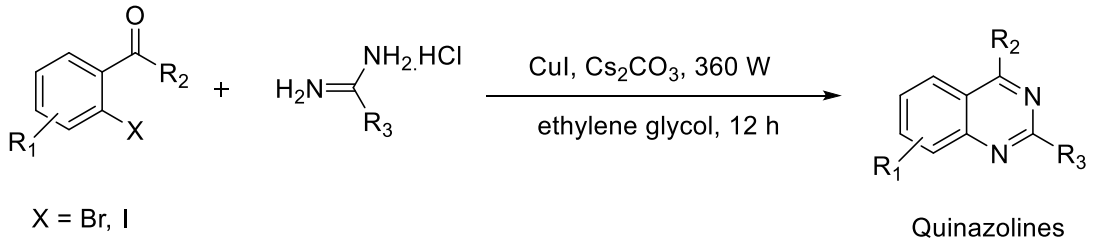
Scheme 25: Synthesis of quinazolines from amidine hydrochloride.

Another Ullmann cross coupling method described for the generation of quinazolines applying copper based catalytic environment was reported by Truong and co workers. This one pot and ligand free synthesis was proved be an important method for the synthesis functionalized quinazolines. In this process the desired heterocycles were acheived by reacting iodo benzaldehyde derivatives with substituted amidine analogues using base Cs_2CO_3 in methanol as solvent. This synthetic protocol is an alternative method to access the quianzoline derivatives in an easy and efficient manner (Scheme **26**).[LXIV]



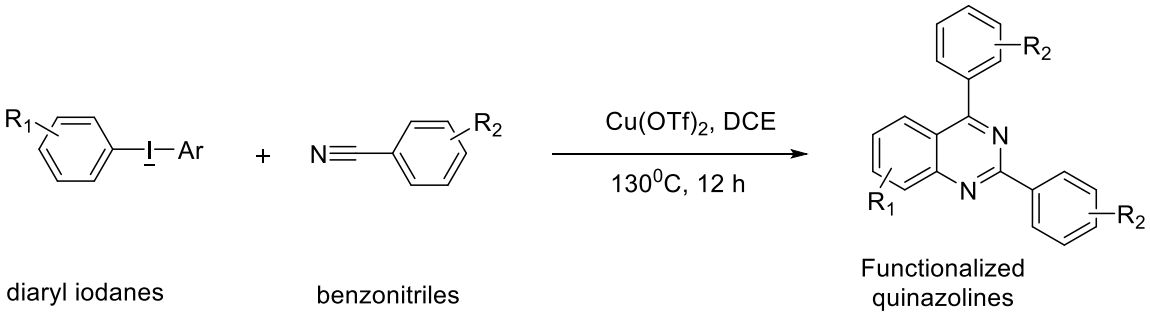
Scheme 26: Synthesis of quinazolines using cesium carbonate.

Ultrasonic irradiation is also used for the construction of quinazoline nucleus. In one of the research published by Raut et. al. they have shown the synthesis of quinazoline by using cuprous oxides nano tubes as heterogenous catalytic system. The catalyst was prepared by then in ultrasonic assisted chemistry, which was further utilized for the construction of quinazolines via one pot cyclisation of 2-bromobenzaldehyde derivatives with amidine hydrochloride as shown the Scheme **27**. The recyclability of the cuprous oxide nano tubes in the suggested process is very significant from green chemistry perspective. [LXV]



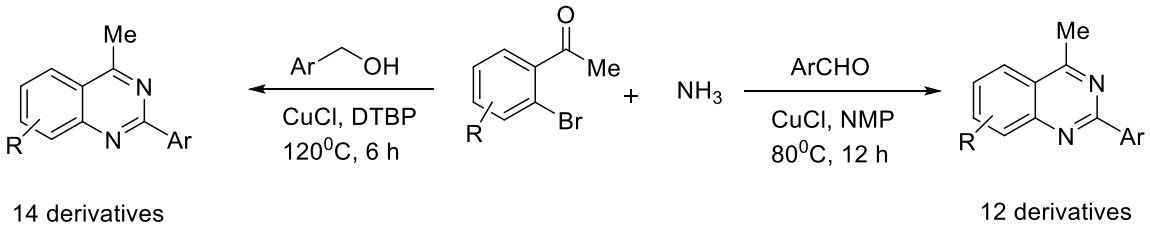
Scheme 27: Copper mediated synthesis in ethylene glycol.

In another report the regioselective synthesis of quinazolines were reported by Wang et. al. in one pot manner. They used diaryl iodanes and nitriles with potassium tertiary butoxide in DMSO. The cupric acetate was employed as heterogeneous catalyst in the process. The rection was believed to procced by electrophilic annulation of reactants leading to regioselective formation of highly functionalized quianazolines (Scheme 28). The respective quinazolines were achieved in 55 to 90% yield. [LXVI]



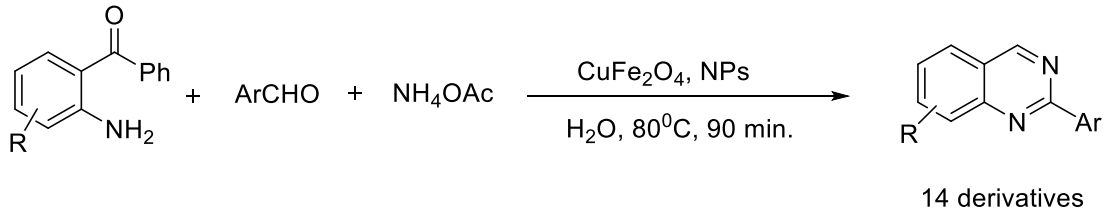
Scheme 28: $Cu(Otf)_2$ mediated synthesis of functionalized quinazolines.

Another copper chloride mediated multicomponent approach has been developed by the research group of Hua which proved very good for the generation of quinazoline heterocycles. The process involves mixing of bromo derivatives of ketones and aromatic aldehydes with ammonia as nitrogen source. Reaction was performed in aqueous medium using water as solvent and air or DTBP as oxidant. The isolated yield of products was upto 91% (Scheme 29).[LXVII]



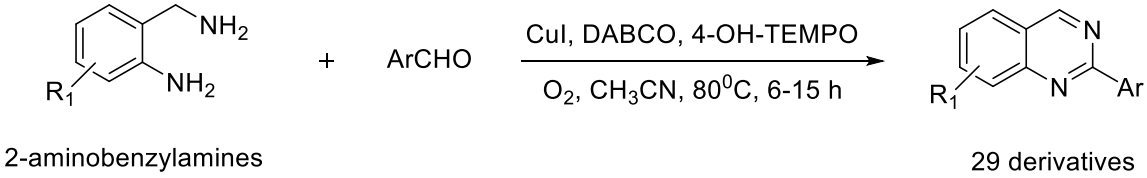
Scheme 29: CuCl mediated synthesis of quinazolines.

Farhang and Baghbanian have proposed an environmentally benign one pot synthetic protocol to access the quinazoline analogues. The recyclable and magnetically recoverable $CuFe_2O_4$ was used as heterogenous catalyst, which leads to cyclisation of aryl aldehydes and 2-amino benzophenones in presence of ammonium acetate.[LXVIII] The copper catalyst $CuFe_2O_4$ was also prepared by them by using Iron and copper nitrate based catalyst in water and NaOH. Catalytic efficiency in nanoparticle was investigated by using differently substitute reactant molecules. Authors found that the catalyst showed a wide range of substrate tolerance (Scheme **30**).



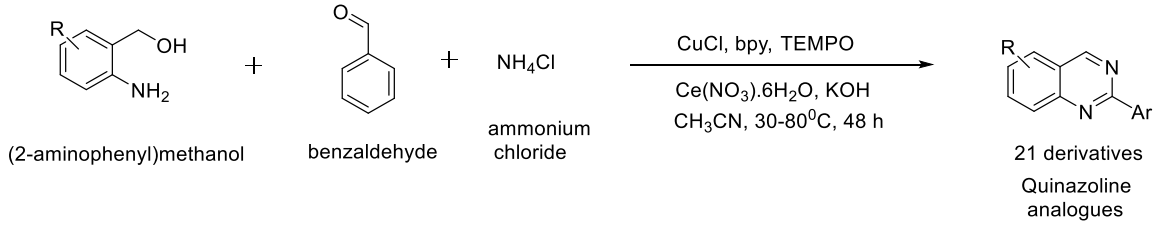
Scheme 30: $CuFe_2O_4$ nanoparticles mediated synthesis of quinazolines.

Han and co-workers have published a single step synthetic fashion leading to generation of quinazoline frameworks involving reaction of 2-aminobenzylamine analogues with the aromatic aldehyde analogues by employing CuCl based catalyst.[LXIX] The air is used as an oxidant for the synthesis. A small library of substituted aryl aldehydes and various substituted 2-benzylamines were used to include the substrate variation and to investigate the catalytic efficiency of the reported catalytic system (Scheme **31**).



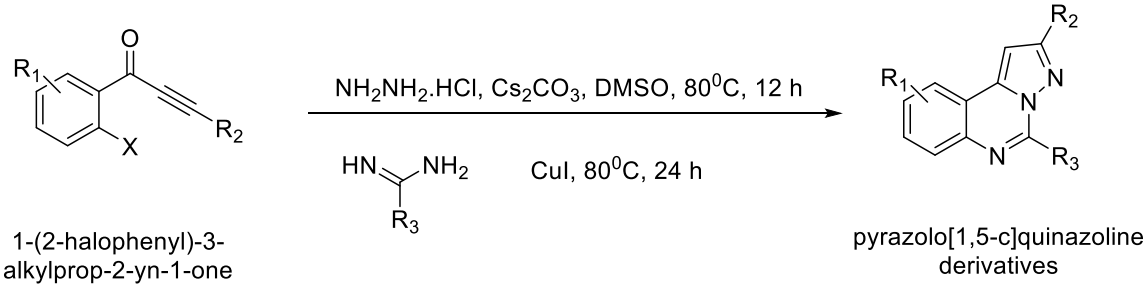
Scheme 31: CuI/DABCO/TEMPO mediated synthesis of quinazoline derivatives.

A single step tandem process leads to formation of quinazolines based compounds was reported by Wu and coworkers. They achieved the formation of desired heterocycles by the reacting substituted aldehydes, amino phenyl analogue with NH_4Cl and TEMPO with CAN (ceric ammonium nitrate). The reported procedure was very practical in order to prepare quinazolines and their analogues as shown in the scheme 32.[LXX]



Scheme 32: TEMPO mediated synthesis of quinazolines.

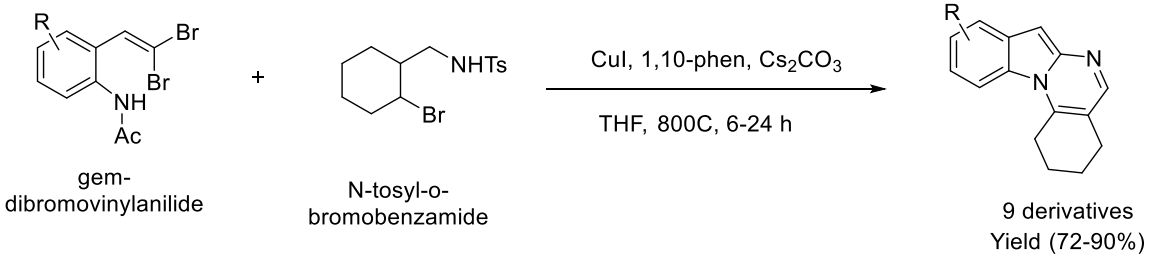
Another synthesis related to pyrazoloquinazolines frameworks reported by Yang and coworkers reveals that the substituted halophenyl alkylpropyn-1-one scaffolds can be used for pyrazoloquinazolines via Cu(I) catalysed reaction sequence.[LXXI] The synthesis was achieved by the reacting halophenyl alkylpropyn-1-one, amidine chloride slat and hydrazine leading to formation of pyrazole derivative of quinazoline in fair quantity. The reported method was claimed to very crucial in the preparation fused heterocycles to generate new quinazoline based libraries (Scheme **33**).



Scheme 33: Preparation of pyrazolo based quinazolines.

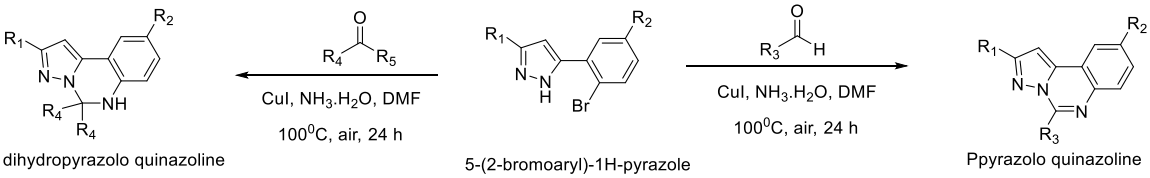
The Kiruthika and Perumal described the construction of indoloquinazolines in one pot reaction by using dibromo vinylanilide and tosyl bromo benzamide analogues using cesium

carbonate in THF under relaxing conditions. Various substituted N-tosyl-o-bromobenzamides were allowed to react in suitable conditions to afford desired heterocyclic system in average yields. Practicality, cost effectiveness and wide scope associated with this synthesis made it a valuable alternative for synthesis of fused quinazolines (Scheme **34**).[LXXII]



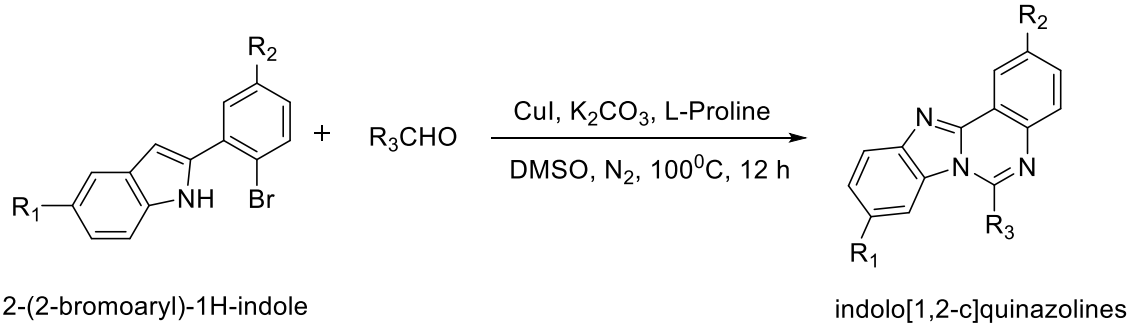
Scheme 34: Synthesis of quinazoline analogues using *gem*-dibromovinylanilide.

In another report published by Gaou and coworkers the one pot copper catalysed reaction is utilized for the synthesis of dihydropyrazolo quinazoline derivatives in good yield. The reported synthetic route involves mixing of bromo substituted pyrazoles with carbonyl analogues in presence of NaOH alkali and air.[LXXIII] The heteroaryl, alkenyl and aryl analogues of aldehydes or ketones were also incorporate to include the substrate variation the proposed synthetic methodology. Various functionalized quinazolines were obtained by this process shows that this synthetic protocol is an excellent alternative for the synthesis of functionalized quinazoline analogues (Scheme **35**).

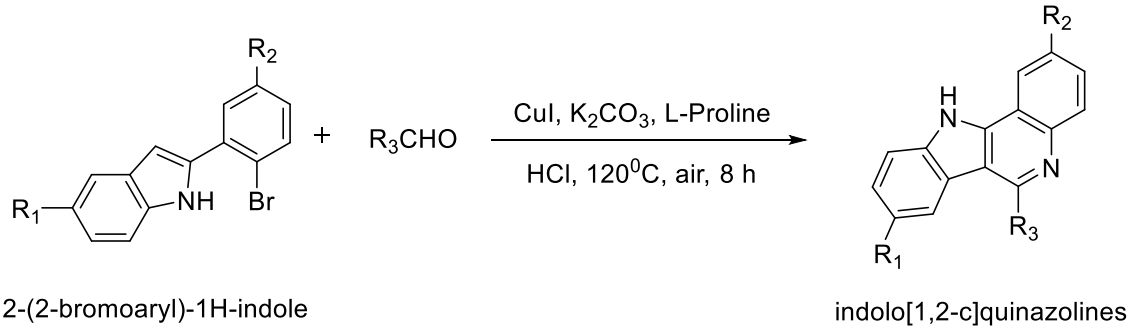


Scheme 35: Synthesis of pyrazoloquinazolines from 5-(2-bromoaryl)1-H-pyrazole

Fan and coworkers in another publication explained the preparation of indolo quinazolines by performing copper catalysed synthesis in one pot sequential manner. They have shown that the indolo substituted quinazolines compounds can be prepared by the reacting bromoaryl indole with aldehydes in presence of ammonium hydroxide. The suggested method provides a selective route for synthesis of indolo quinazolines. They claimed that on controlling the reaction condition one can maintain the regioselectivity of the reaction. Synthetic process reveals that under the acidic medium C-C coupling leads to formation of indolo quinazolines (Scheme 36a and 36b).[LXXIV]



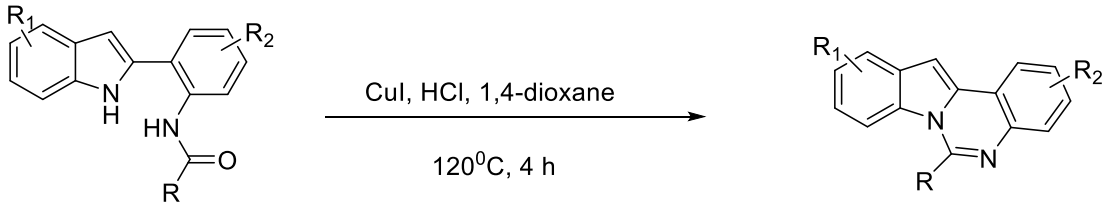
Scheme 36a: Synthesis of indoloquinazolines.



2-(2-bromoaryl)-1H-indole indolo[1,2-c]quinazolines

Scheme 36b: Synthesis of indolo quinazolines in acidic medium.

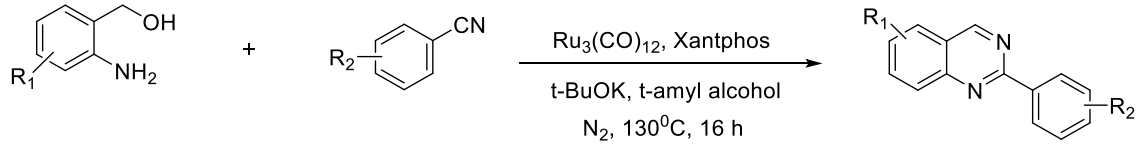
An improved process suggested from the group of Fan generates quinazoline based derivatives have been explored oxygenation methodology by mixing amidoaryl indoles, which further undergone intramolecular cyclisation reaction in acidic medium affords quinazoline molecules. The isolated yield of the synthesized quinazolines were found to be 45 – 70% as depicted in (Scheme **37**).[LXXV]



2-(2-amidoaryl)-1H-indoles indoloquinazoline analogues

Scheme 37: Synthesis of Indole analogues of quinazoline under acidic medium.

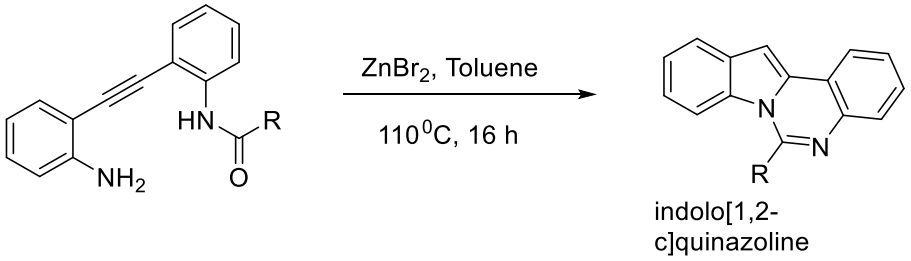
Chen and coworkers published an excellent synthesis of quinazolines using Ruthenium catalysed dehydrogenative coupling reaction. They achieved the quinazoline nucleus by reacting 2-aminomethanol analogues and benzonitrile derivatives using triruthenium dodecacarbonyl as metal catalyst and Potassium t-butoxide base. BY applying this synthetic protocol, they have generated a 22 membered library of quinazoline derivatives. Avoiding the use of halogenated reactants make this synthetic process more valuable for the synthesis of quinazoline analogues (Scheme **38**).[LXXVI]



2-aminomethanol benzonitrile 26 derivatives

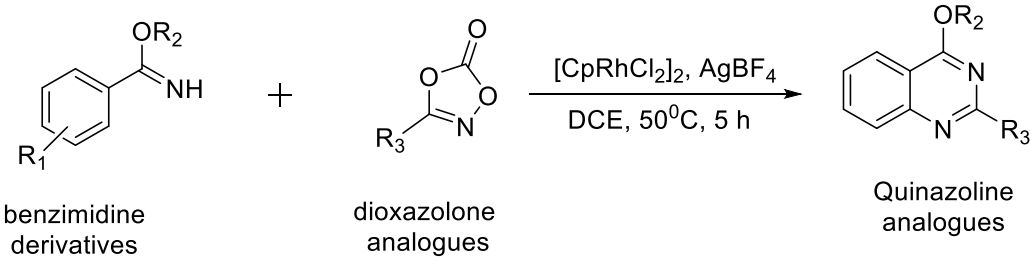
Scheme 38: Ruthenium mediated synthesis of quinazoline derivatives.

Synthetic methodology involving hydro-amination cyclisation sequence leading to formation of indole derivative of quinazoline skeleton was reported by Wang and coworkers. Their reported methodology proceeds by reaction of acyclic alkynes through $ZnBr_2$ catalysis. Product formation goes through zinc bromide mediated amination and cyclisation of amide and indole nitrogen. The product forms in fair yields (31 – 92%) as shown in the (Scheme **39**).[LXXVII]



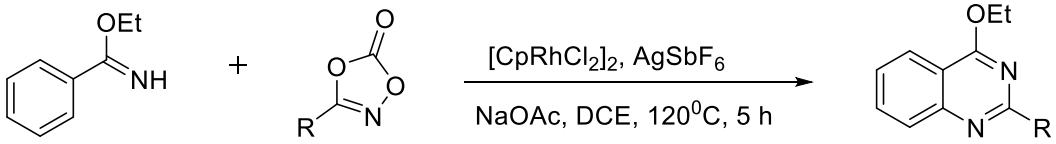
Scheme 39: Zinc-catalyzed cyclisation to access indoloquinazolines.

Rhodium based catalysts are also proved to be significant for the synthesis of quinazoline analogues.[LXXVIII-LXXXII] In this sequence an important synthesis has been reported by Zhu and coworkers in which they have used $[CpRhCl_2]_2/AgBF_4$ as catalyst. They utilized the catalytic efficiency of rhodium based catalyst in double C-N linkage generation sequence for the making different quinazolines. The synthetic sequence involves the reaction of benzimidine derivatives with dioxazolone analogues leading to formation of functionalized quinazolines.[LXXXIII] The dioxazolone derivatives are believed to be used for oxidation which is crucial to maintain the catalytic property of system used. Various substituted reactants were used as reactant to prove the efficiency of developed protocol (Scheme **40**).



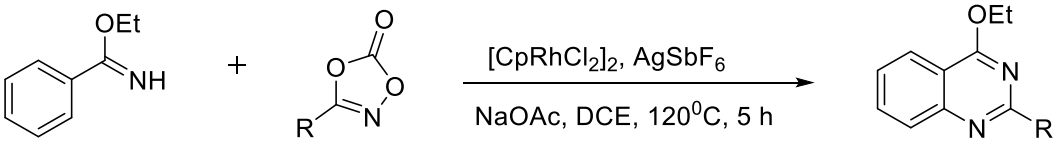
Scheme 40: Rhodium catalysed synthesis of Quinazoline analogus.

Similarly, and effective synthesis of quinazoline N-oxides was reported by Li et. al. using oximes of ketone and oxazolon analogues. The reaction proceeds by the Zinc and Rhodium catalysts in +2 state to promoting reactivity in C-H functionality in ketoxime analogues. The oxidant free reaction sequence results the formation of differently switched heterocyclic system in fair yields (48-90%) as shown in (Scheme **41**).[LXXXIV]



Scheme 41: Rhodium mediated synthesis of highly functionalized quinazolines.

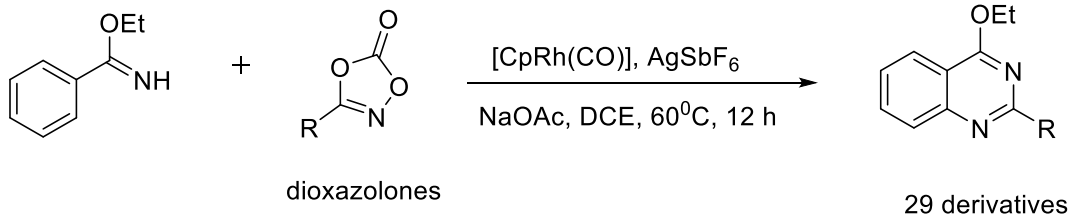
An interesting synthesis of biologically active quinazolines were proposed by Wang and coworkers which exploited the catalytic potential of copper and Rhodium complexes to promote C-H activation. They achieved the desired synthesis by the reaction of imidate derivatives and alkyl azide analogues. This reported methodology an important other process to get N based heterocycles from alkyl azides as depicted in (Scheme **42**).[LXXXV]



Scheme 42: Rhodium mediated synthesis of functionalized quinazolines.

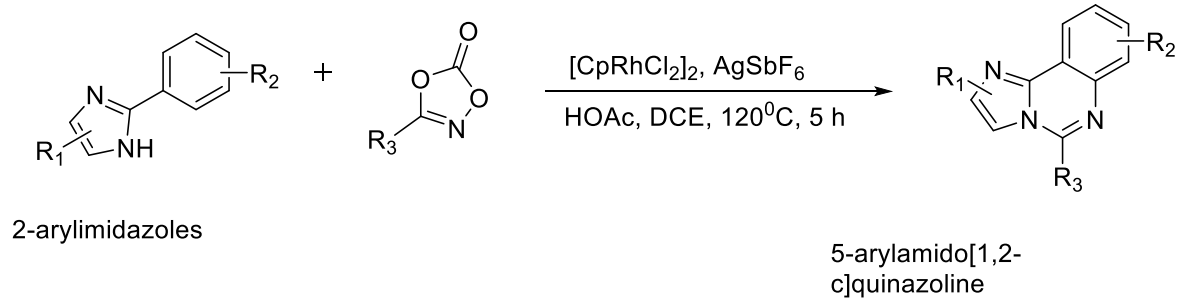
Cobalt based catalytic system are also explored to achieve the generation of quinazoline based compounds. In another report published by Wang and coworkers the

cyclopentadienylcobalt-dicarbonyl catalyst was used for synthesis of multifunctional quinazolines.[LXXXVI] The reaction proceeds via [4+2] cycloaddition process of dioxazolones with imine analogues leading to the formation of quinazolines. The cobalt mediated carbon-hydrogen amidation reaction followed by cyclisation provides the functional quinazolines in good to excellent yields (50-97%). Authors claimed that the cobalt based catalytic system is very selective for the synthetic methodology (Scheme 43).



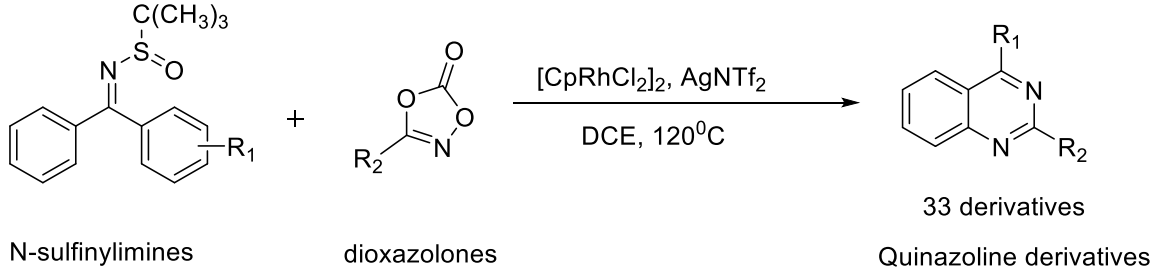
Scheme 43: Cobalt based catalyst mediated synthesis.

Another rhodium catalysed synthetic methodology was proposed by Wang and coworkers leading to formation of 5-arylamido[1,2-c]quinazoline derivatives. The formation of quinazoline analogue was occurred by the reaction of ketone derivatives and 2-arylimidazoles. The halogen free reactants and cheap starting material made this process more valuable to access the quinazoline derivatives (Scheme **44**).[LXXXVII]



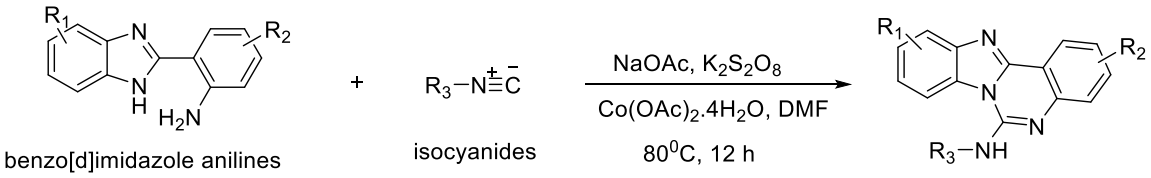
Scheme 44: Synthesis of rhodium catalysed quinazolines.

An interesting synthesis of quinazolines was proposed by Wang et. al. involves reaction of N-sulfinylimines and benzamidates with dioxazolones. This synthesis employed cobalt based catalytic system for rapid conversion of various dioxazolones into substituted quinazolines. The regioselectivity observed in this synthetic sequence is one of the prime speciality of this procedure (Scheme 45).[LXXXVIII]



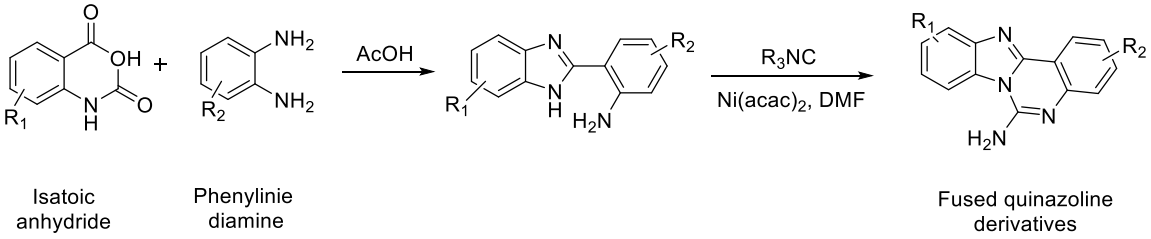
Scheme 45: Synthesis of quinazolines from N-sulfinylimines.

Ahmadi and coworkers in an example have shown the preparation of Quinazoline skeleton using cobalt mediated isonitrile insertion cyclisation reaction sequence. The reaction of isocyanides and benzo imidazoles.

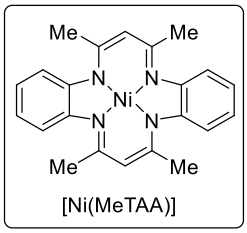


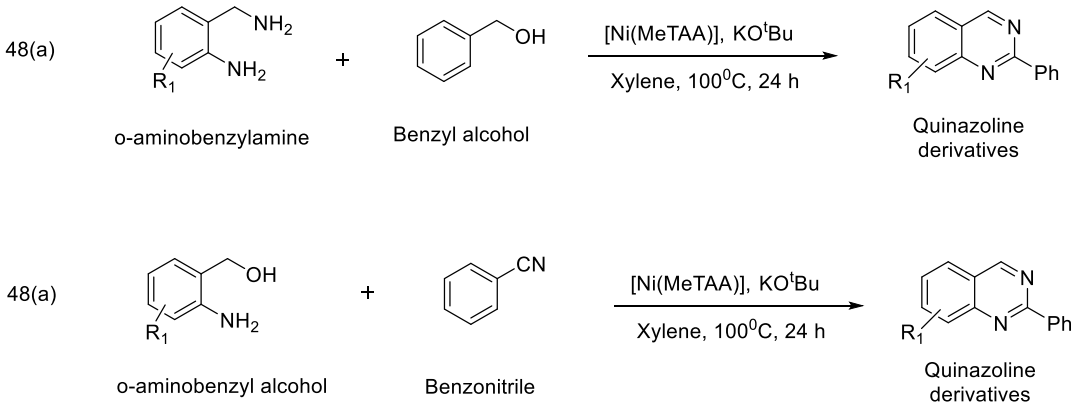
Scheme 46: Synthesis of fused quinazolines from isocyanides.

The simplicity in the product isolation and a direct methodology made this reaction as an important alternative for the synthesis of the functionalized quinazolines (Scheme **46**).[LXXXIX] Nickel based catalytic systems have also been explored for the construction of quinazolines. Nickel based molecules are well known for their catalytic potential in the synthesis of various heterocyclic molecules. [XC-XCII] In the same direction Sharda and co workers proposed an interesting way to prepare quinazolines by one pot approach involves insertion process of isonitrile in presence of air followed by cyclisation.[93] During the reaction the Isatoic anhydride ring opens first and then it gets annulated with the benzimidazole to form the product. The Ni(II) is reported to promote intramolecular insertion of isocyanides (Scheme **47**). This synthetic procedure has benefits of ligand and base free reaction conditions in a metal catalysed synthesis. Cheap starting chemicals and ease of work up are the features of this synthesis which make it a better alternative over traditional way to achieve the quinazoline heterocycles. It was also investigated that the quinazoline derivative prepared via this synthetic protocols were exhibiting the fluorescence properties.



Scheme 47: Ni mediate synthesis of fused quinazoline derivatives.

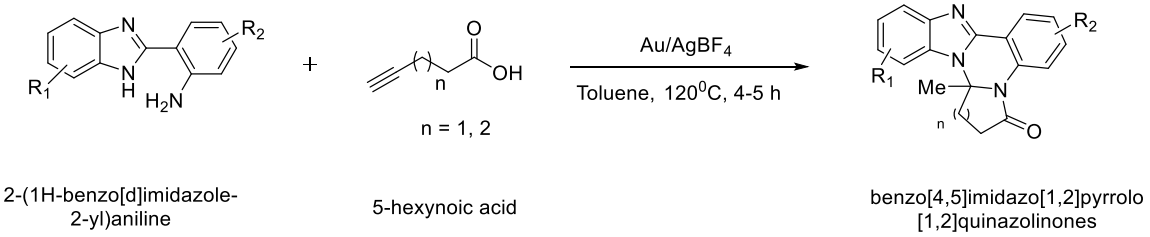




Scheme 48: [Ni(MeTAA)] mediated synthesis of quinazoline derivatives.

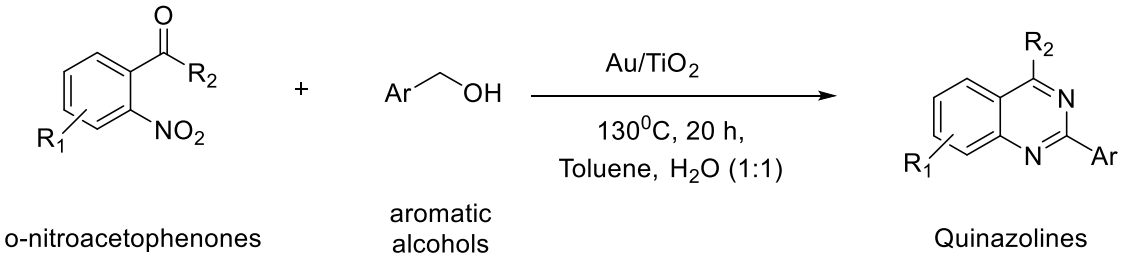
Another nickel mediated synthesis involving use of Ni(MeTAA) catalysis was given by Parua and coworkers to generate the quinazolinebased heterocycles. This was achieved by dehydrogenation and coupling reaction sequence of amino benzylamine and alcohol functionality and amino benzyl alcohols with nitriles.[XCIV] The reaction was performed with the help of tertiary butoxides at 110^0C, which took 24 hrs for completion. The proposed synthetic route is an environment friendly alternative to achieve the quinazoline hetrocycles (Scheme **48a** and **48b**).

Gold based catalytic systems are also reported used for the construction of quinazoline heterocycles in literature. This catalytic system was used by Liu and coworkers in facile and easy synthetic procedure leading to prepare benzo fused quinazolinones. They achieved molecule mentioned by reacting functionalized benzo fused anilines and hexynoic as well as pentynoic acid. The published reaction sequence was catalyzing by silver / gold based maediaters. The three new C-N bond formed in this reaction in a single step (Scheme **49**).[XCV]



Scheme 49: Gold mediated synthesis of fused quinazolines.

In another gold catalysed reaction Wang and coworkers have shown a new procedure for the synthesis of 2,4-difunctional quinazolines. Their process involves the hydrogen transfer with mixing of alcohols and nitro acetophenones. The reaction was catalysed by silver / titanium oxide as catalytic system. The designed catalytic system has demonstrated a wide range of substrate scope by using different aromatic alcohols or nitroacetophenones. The recyclability of catalyst presents a better alternative for the preparation of quinazolines via gold catalysed reaction (Scheme **50**).[XCVI]



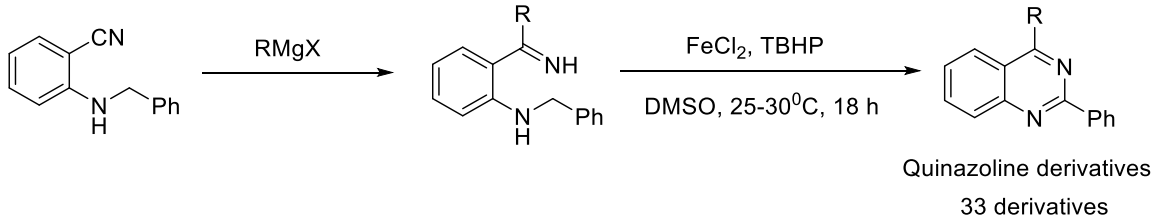
Scheme 50: Au/TiO_2 mediated synthesis of quinazolines.

Tang and coworkers have reported the formation of dihydropyrazole forms of quinazolines using gold based catalytic systems. They proposed chemo selective protocol involving cyclisation of propargyl hydrazones.

Scheme 50: Synthesis of dihydropyrazolo quinazoline analogues using propargyl hydrazones.

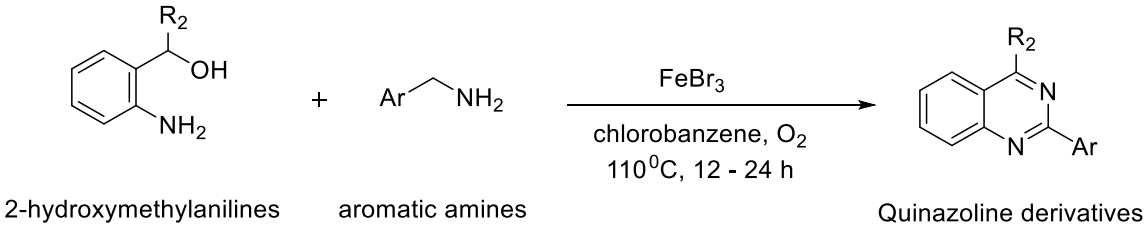
Their process was performed in DMSO as a solvent. To explore the substrate tolerance of the the catalyst various N-propargylic sulfonyl hydrazones were employed for the synthesis of pyrazolo quinazoline derivatives (Scheme **50**).[XCVII]

Iron based catalytic systems have also been proved to be very efficient for the synthesis quinazoline derivatives.[XCVIII-C] Towards this direction the research group of Chen et. al. proposed an easy and facile synthesis using ferric chloride as catalyst. Their process involves C-H oxidation leads to formation of intramolecular carbon nitrogen bond formation for the construction of quinazolines (Scheme **51**). t-butyl hydroperoxide as terminal oxidant.[CI]

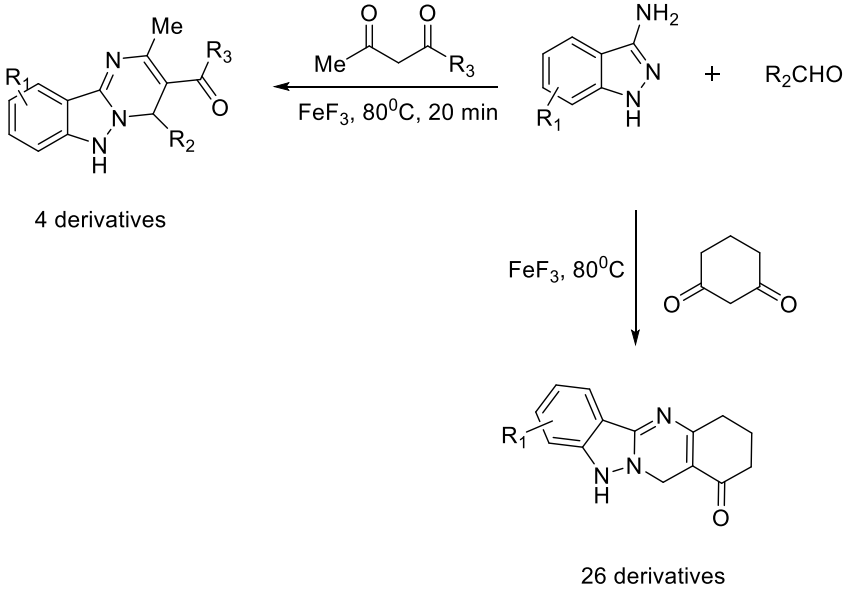


Scheme 51: Iron mediated synthesis of quinazoline derivatives.

In another report published by Gopalaiah and coworkers in 2017, illustrating the preparation of quinazoline analogues by the reaction of 2-hydroxymethylanilines and aromatic amines. The reaction was performed under aerobic conditions using benzene as solvent. The intermediate formation of N-benzylidenebenzylamine leads to intramolecular cyclisation which ultimately form corresponding quinazoline analogues in good yields (Scheme **52**).[CII]



Scheme 52: Synthesis of quinazolines from 2-hydroxymethylanilines.



Scheme 53: Synthesis of quinazoline analogues via iron based catalysts.

Jeong and Shinde have published an important process which leads to generation of functionalized quinazolines using indazole amine derivatives, aldehyde in a ferric chloride catalyzed reaction sequence. Reported process was performed under ultrasonication and in absence of solvent. The easy work up procedure and ultrasonic reaction condition lake the

reported protocol an alternative over the other synthetic schemes for the synthesis of quinazoline molecules (Scheme 53).[CIII]

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