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REVIEW

Recent advances in the application of acetophenone in heterocyclic compounds synthesis

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Abstract

Acetophenone is an interesting synthon in the most organic reactions. Acetophenone has been utilized in the synthesis of many heterocyclic compounds. Acetophenone and most of its derivatives are commercially available or readily accessible and hence are ideal synthon for multicomponent reactions including the three- and four-component reactions. Also, the biological activities of some compounds were studied. Herein, we want to review the application of the acetophenone as starting material in the synthesis of various heterocyclic compounds including fused and five-, six-, seven-membered rings via multicomponent reactions.

Graphic abstract

Extended author information available on the last page of the article

Keywords Acetophenone · Heterocyclic compounds · Multicomponent reactions · Five-membered rings · Six-membered rings

Abbreviations

NBSac	*N*-Bromosaccharin
THF	Tetrahydrofuran
Yb(OTF)	Ytterbium(III) triflate
TBBDA-MNPs@ SiO_2-Pr-AP	*N*,*N*,*N*′,*N*′-tetrabromobenzene-1,3-disulfonamide [TBBDA], poly(*N*,*N*′-dibromo-*N*-ethylbenzene-1,3-disulfonamide) [PBBS]
BIL	Basic ionic liquid
$Y(OTf)_3$	Yttrium triflate
DMF-DMA	*N*,*N*-dimethylformamide-dimethylacetal
NBS	*N*-bromosuccinimide
BDMS	Bromodimethylsulfonium bromide
TCRT	Three-component ring transformation
MWCNTs	Metal oxide nanocomposites
SBA-Pr-SO_3H	Sulfonic acid functionalized silica
STO	Sulfated tin oxide
CAN	Ceric ammonium nitrate
FeAlP-550	Amorphous mesoporous iron aluminophosphate
TBBDA	*N*,*N*,*N*′,*N*′-tetrabromobenzene-1,3-disulfonamide
[Bmim]HSO_4	1-Butyl-3-methylimidazolium hydrogen sulfate
PFPAT	Penta fluorophenylammoniumtriflate
Wet-TCT	Wet 2,4,6-trichloro-1,3,5-triazine
[Hmim]NO_3–[Bmim]BF_3	1-Methylimidazolium nitrate in 1-butyl-3-methylimidazolium tetrafluoroborate
GO	Graphene oxide

Introduction

Acetophenone is a useful precursor in the organic reactions for the synthesis of heterocyclic compounds [1–4]. There are several methods for the preparation of acetophenone **2**; one of them comprises the reaction of aryl triflates **1** with a mixture of $SnMe_4$, Pd(0) and CO (balloon) in the presence of Et_3N in DMF at 60 °C (Scheme 1) [5–8].

Acetophenone and its derivatives use in the organic reactions, including in (pseudo)-two-, three- and four-component reactions [9–12]. Furthermore, acetophenone is the main constituent of many natural compounds. For example, the three new acetophenone derivatives were isolated from the leaves of Acronychiaoligophlebia [13, 14]. Recent studies have demonstrated the antifungal activities of some naturally occurring acetophenone derivatives. For example, xanthoxylin isolated from Melicope borbonica leaves exhibited the antifungal activity against Candida albicans and Penicillium expansum; 4-hydroxy-3-(isopentent-2-yl) acetophenone, from Helichrysum sp., showed antifungal activity against Cladosporium herbarum (Fig. 1) [7, 15]. We have already published the synthesis of heterocyclic compounds via multicomponent reactions [16–24]. Based on previously published articles, in the review, we will try to highlight the applications of acetophenone as starting materials in the synthesis of various heterocycles.

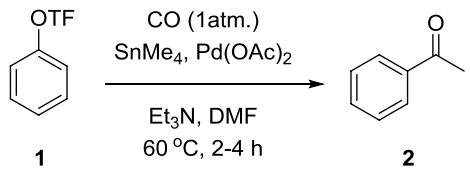
Acetophenone reactions

Acetophenones have been applied in the structure of different types of heterocyclic frameworks. In this review, a range of heterocyclic compounds from acetophenone involving: five-, six-, seven-membered through three-, four-component reactions, are presented.

Synthesis of five-membered rings

Five-membered rings containing O atom

Initially, intermediate compounds **4** were prepared in excellent yields via the Claisen condensation of acetophenones **2** with methyl 2-methoxytetrafluoropropionate **3**. 5-Aryl-2-hydroxy-2-(trifluoromethyl)furan-3(2*H*)-ones



Scheme 1 Synthesis of acetophenone 2

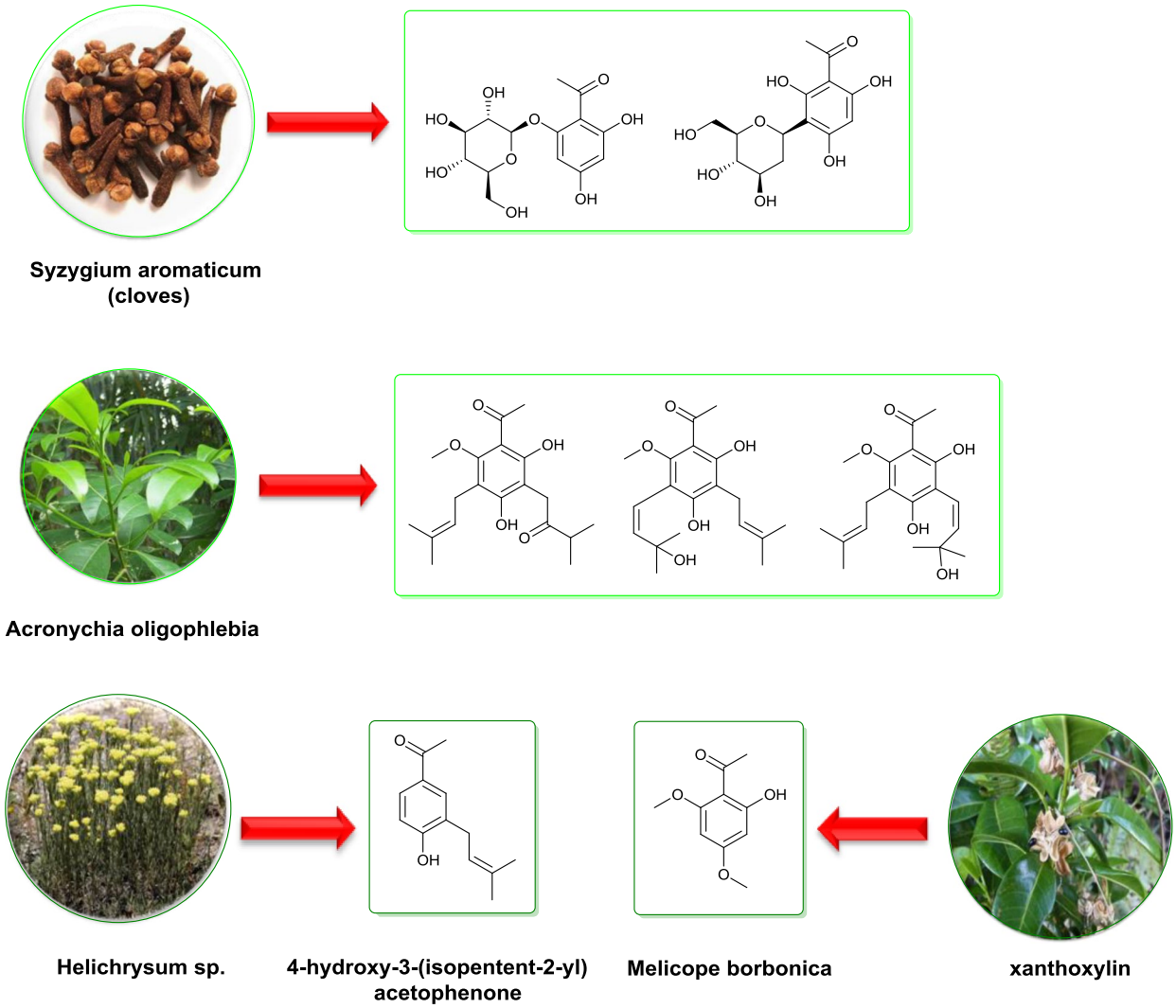
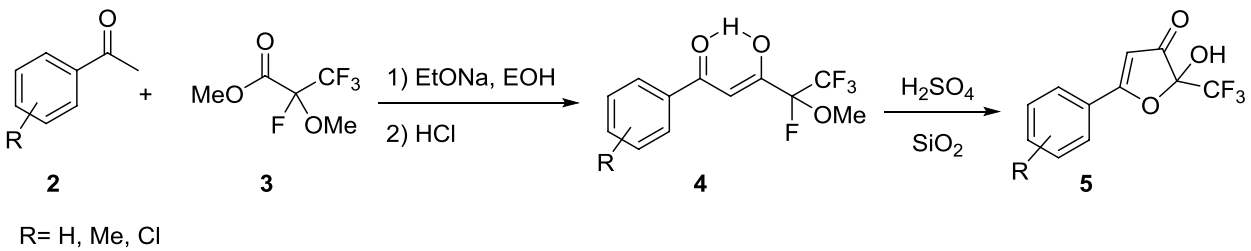


Fig. 1 Medicinally significant natural products containing acetophenone

5 were generated via intramolecular cyclization of intermediate compounds **4** in the presence of H_2SO_4 and SiO_2 as a catalyst (Scheme 2) [25].

Various 2,3-substituted-butyrolactones **8** have been prepared by three-component reaction of acetophenone **2**, aryl bromides **6** and dimethyl itaconate **7** in MeCN as a solvent at 60 °C in the excellent yield (Scheme 3) [26].

A novel copper-catalyzed domino reaction of acetophenone derivatives **2**, α,β-unsaturated dicarboxylate **9** and diethyl zinc **10** produced lactones **11** in the high yield,



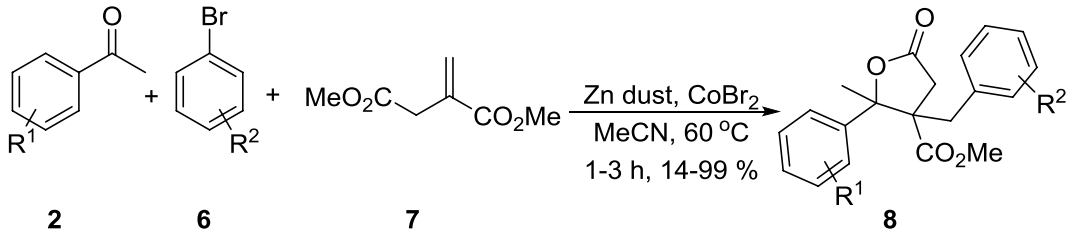
Scheme 2 Synthesis of 5-aryl-2-hydroxy-2-(trifluoromethyl)furan-3(2*H*)-one 5

the role of diethyl zinc was as an alkyl Michael donor (Scheme 4) [27].

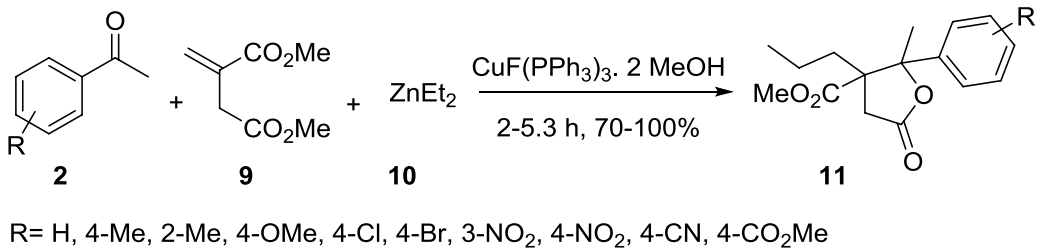
A large number of furo[3,2-*c*]coumarin derivatives **13** were obtained via three-component reaction of acetophenone derivatives **2** and two moles of various coumarins **12** in the presence of molecular iodine in DMSO at 80 °C (Scheme 5) [28].

Five-membered rings containing *N* atom

Rad-Moghadam et al. [29] developed a sequential tandem reaction for the synthesis of new series of oxindolyl-7-deazapurine derivatives **16** via the novel cyclocondensation reaction between acetophenones **2**, isatins **14** and 6-amino-uracils **15** in ethanol under reflux (Scheme 6). 5-(2-Oxoindolin-3-yl)-1*H*-pyrrolo[2,3-*d*]pyrimidine-2,4(3*H*,7*H*)-dione and its derivatives **16** were evaluated for their antimicrobial activities [29].

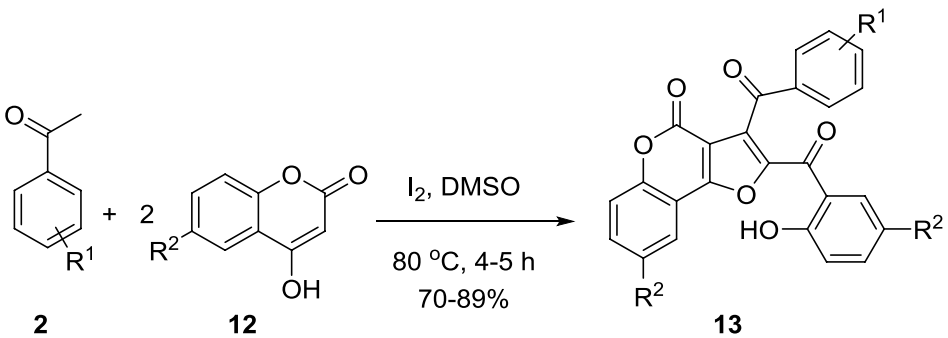


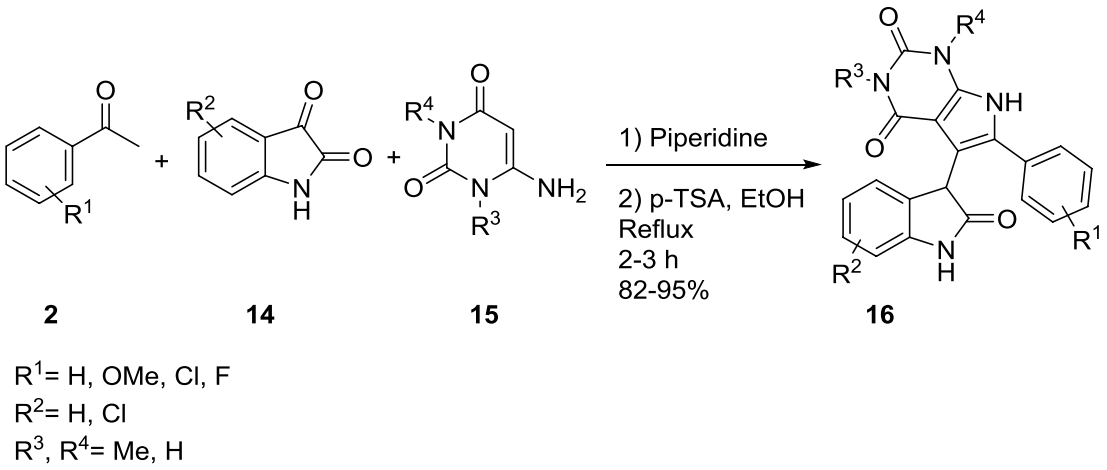
Scheme 3 Synthesis of various 2,3-substituted–butyrolactones 8



Scheme 4 Synthesis of lactones 11

Scheme 5 Synthesis of furo[3,2-*c*]coumarin derivatives 13





Scheme 6 Synthesis of 2-oxoindolin-3-yl-pyrrolo[2,3-*d*]pyrimidine-2,4(3*H*,7*H*)-diones 16

The one-pot reaction of substituted acetophenone **2**, pyridine **17**, acetic acid **18** and molecular iodine **19** in the presence of ceric ammonium nitrate (CAN) as a catalyst was carried out for the synthesis of 1-iodoindolizines **20** in 45–56% yields (Scheme 7) [30].

Yahyavi et al. [31] prepared the synthesis of 2,3-disubstituted-chromeno[4,3-*b*]pyrrole-4(1*H*)-ones **31** or **32** via multicomponent reactions of phenylglyoxals **28**, active methylene compounds **29** and 4-amino coumarin **30** (Scheme 8).

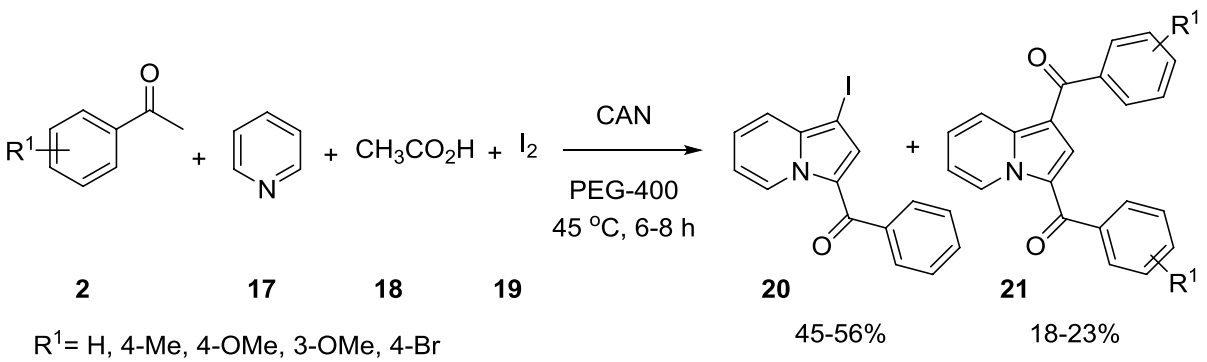
The synthesis of pyrrole derivatives **29** was accomplished with good yield using acetophenone **2** and trimethylacetaldehyde **27** and TosMIC in $LiOH{\cdot}H_2O$ at room temperature (Scheme 9) [32]

In 2018, Mishra et al. [33] developed the synthesis of pyrroles via multicomponent reaction of acetophenone, 4-hydroxycoumarin and amino chromones in the presence of I_2 as a catalyst in DMSO. The products confirmed with high yield in a short time (Scheme 10).

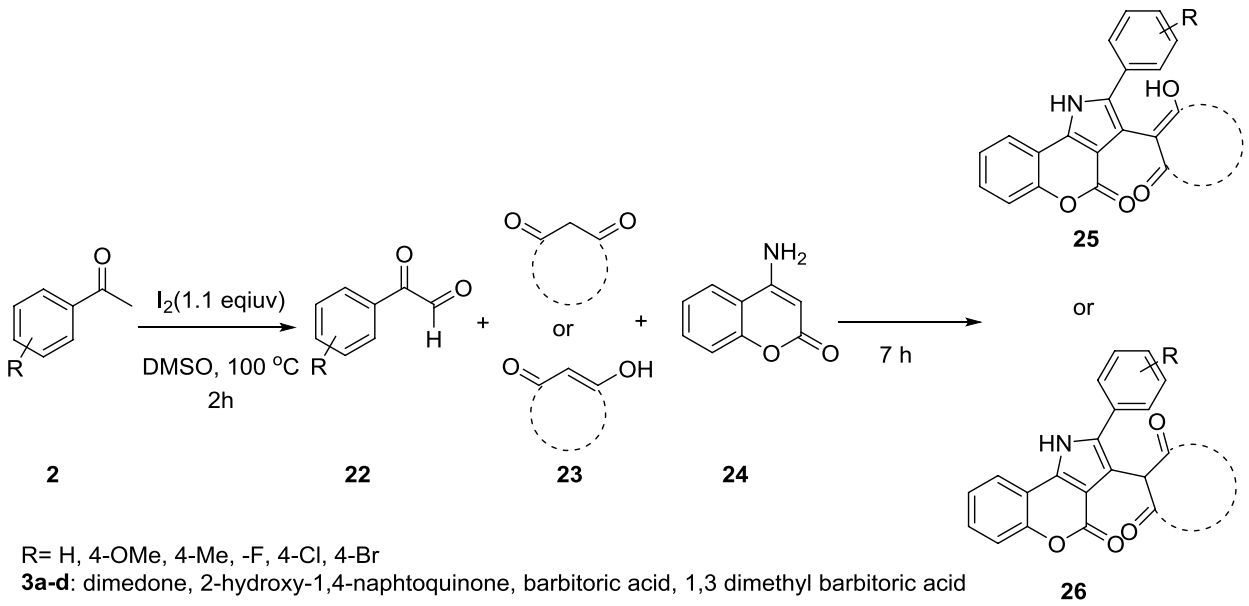
Five-membered rings containing two hetero atoms

The synthesis of 1,3-oxathiolane **34** has been developed via the carbonyl group protection of acetophenone **2** using mercaptoethanol **33** in the presence of Tin(IV) hydrogen phosphate $[Sn(HPO_4)_2\ H_2O]$ nanodisks as an efficient heterogeneous catalyst at room temperature (Scheme 11) [34]. Alinezhad et al. [35] performed this reaction with *N*-bromosaccharin (NBSac) as a catalyst and obtained the product. Two different methods for the synthesis of this product are compared in Table 1.

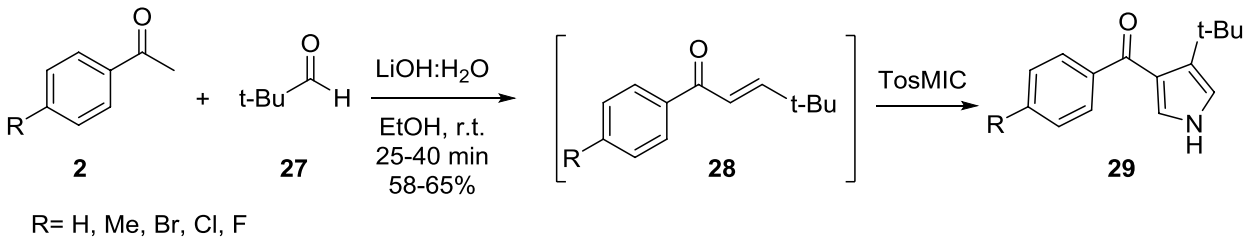
Initially, 1-(substituted methylbenzoyl)-3-arylthioureas **37** were prepared via condensation of benzoyl chlorides **34** potassium thiocyanate **36** in acetone that followed by reaction of suitably substituted anilines **38**. Next cyclization of 1-aroyl-3-arylthioureas **39** with acetophenone **2** in the presence of bromine and triethyl amine to afford 2-aroylimino-3-aryl-4-phenyl-1,3-thiazolines **40** (Scheme 12) [36].



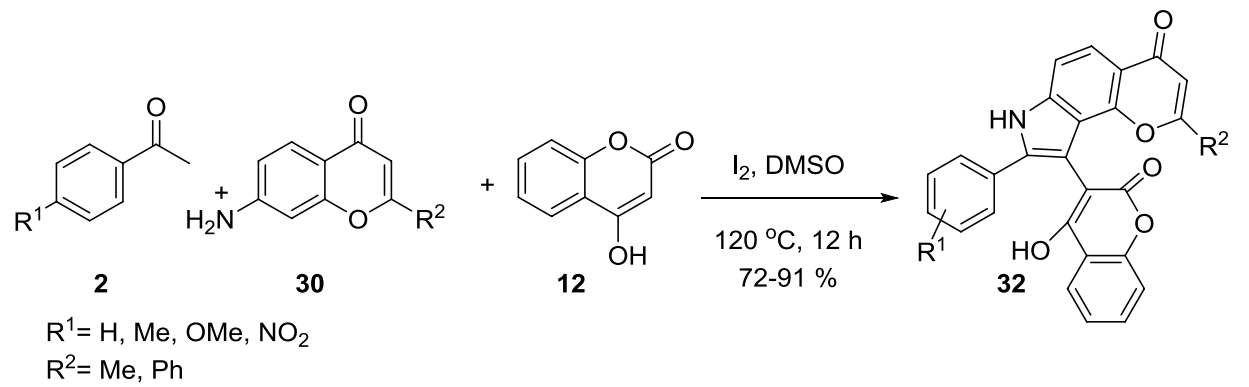
Scheme 7 Synthesis of 1-iodoindolizines 20



Scheme 8 Synthesis of 2,3-disubstituted-chromeno[4,3-*b*]pyrrole-4(1*H*)-ones 25 or 26



Scheme 9 Synthesis of pyrrole derivatives 29



Scheme 10 Synthesis of pyrroles 32

A probable mechanism for the preparation of compounds **47** was shown in Scheme 13. Initially, the reaction of acetophenones **2** with anhydrous chloral **41** gave trichloroethylidene acetophenones **42**. According to the peculiar mechanism of this reaction, the 2,2-dichlorovinylacetophenones **42** were generated in high yields. These, 2,2-dichlorovinylacetophenones **44** reacted with hydroxylamine **45** to create oxime intermediates **46** which was treated with

Scheme 11 Synthesis of 1,3-oxathiolane 28

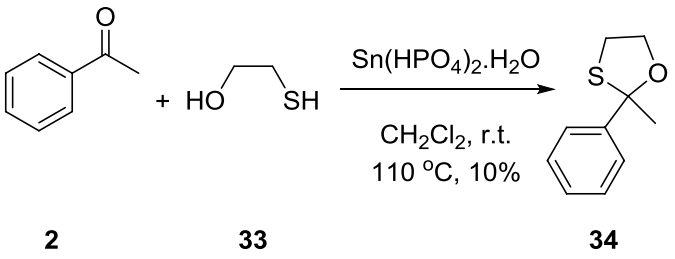
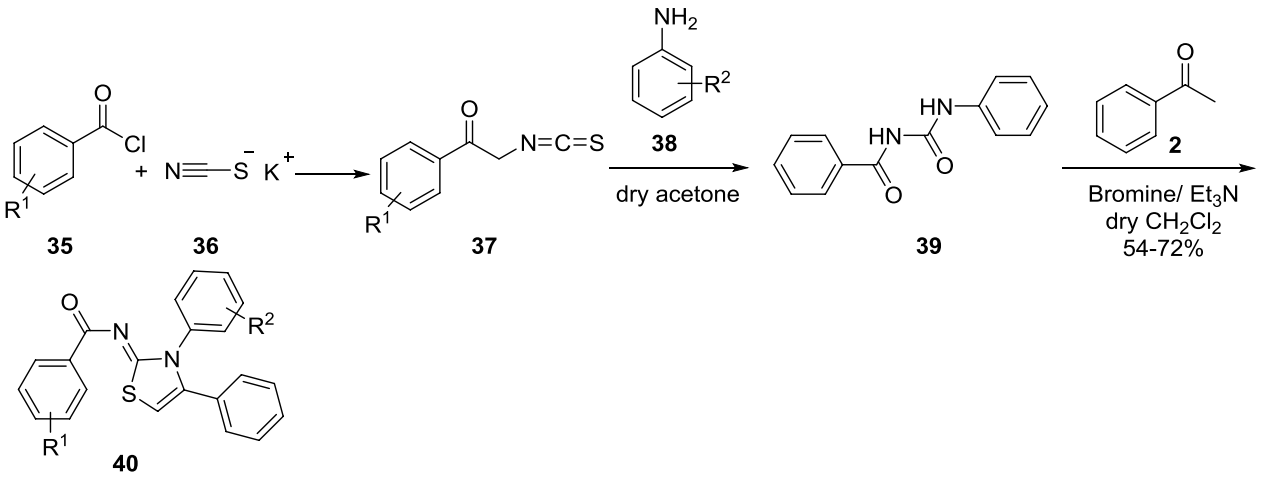


Table 1 Different reported strategies for the synthesis of 1,3-oxathiolane 34 in CH_2Cl_2 as solvent at room temperature

Entry	Catalyst	Time (min)	Yield (%)	References
1	$Sn(HPO_4)_2{\cdot}H_2O$[a]	70	10	Hazarika et al. [34]
2	NBSac[b]	85	50	Alinezhad and Fallahi [35]

[a]Tin(IV) hydrogen phosphate

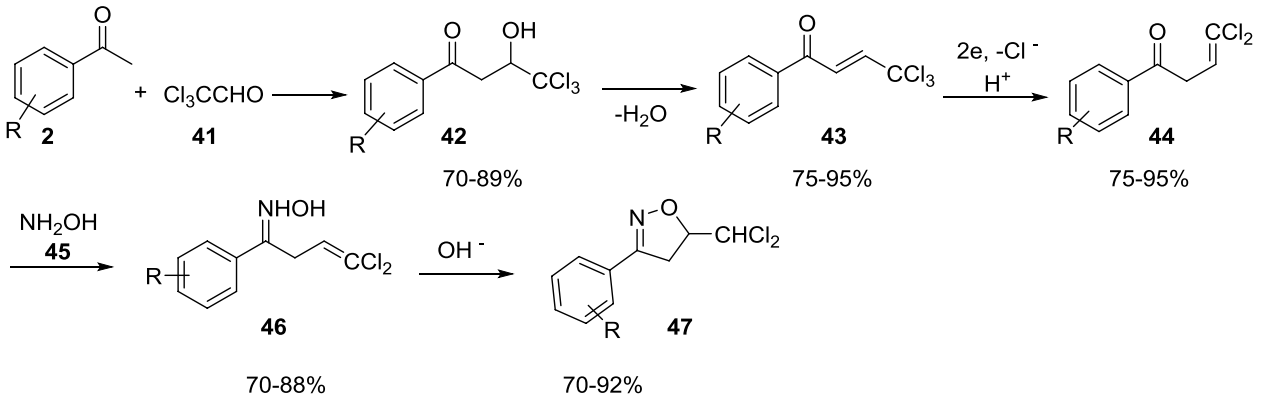
[b]*N*-Bromosaccharin



R^1 = H, 2-Me, 3-Me, 2-Br, 3-Cl

R^2 = H, 2-Me, 3-Me, 2-OMe, 2-Cl, 3-Cl, 2,4-$(Cl)_2$, 2-NO_2, 3-NO_2, 1-Naphthyl

Scheme 12 Synthetic route to 2-aroylimino-3-aryl-4-phenyl-1,3-thiazolines 40



R= H, 4-Me, 4-OMe, 4-F, 4-Cl, 4-Br, 4-NO_2 , 4-C_6H_5

Scheme 13 Synthesis of novel 3-aryl-5-dichloromethyl-2-isoxazolines 47

aqueous sodium hydroxide to afford novel 3-aryl-5-dichloromethyl-2-isoxazolines **47** (Scheme 13) [37].

Synthesis of trisubstituted isoxazoles **49** via the reaction of acetophenone **2** and ethyl nitroacetate **48** was carried out at in the presence of I_2/CuO as a catalyst in DMSO at 70 °C (Scheme 14) [38]. Many of these compounds were evaluated for the biological activities, such as antibacterial, antiviral, anticancer and antithrombotic activities [38].

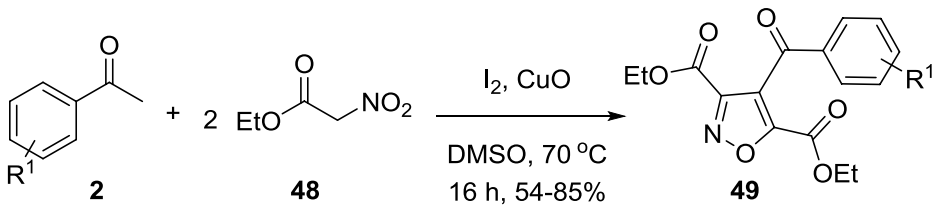
In 2014, Wang et al. [39] synthesized 4,5-dihydropyrazole derivatives **52** via one-pot three-component condensation of acetophenones **2**, arylacetylenes **50** and hydrazines **51** in the presence of KOtBu/DMSO (Scheme 15).

Yang et al. have established trisubstituted isoxazoles **54** via an efficient one-pot two-component reaction of acetophenone **2** and α-nitroketones **53** in DMSO at 70 °C in the presence of the I_2/CuO as a catalyst (Scheme 16) [38].

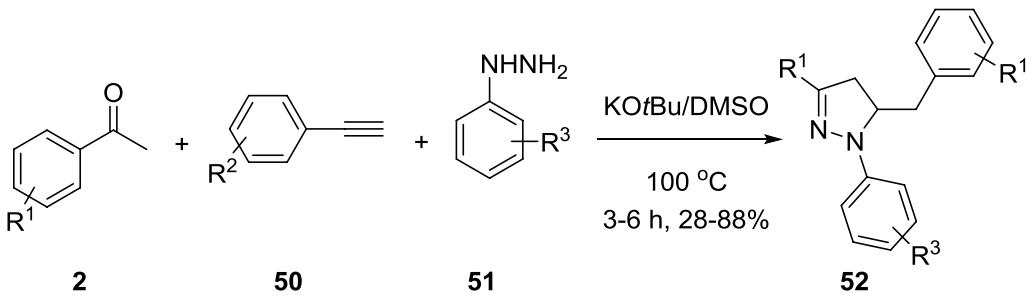
A novel series of coumarin-substituted thiazolyl-3-aryl-pyrazole-4-carbaldehydes **57** were synthesized through an efficient, one-pot multicomponent reaction of acetophenones **2**, 3-(2-bromoacetyl) coumarins **55** and thiosemicarbazide **56** utilizing Vilsmeier–Haack reaction condition with good yields (Scheme 17) [28].

Amer et al. [40] performed three-component reaction via condensation of acetophenones **2**, triazole **58** and

Scheme 14 Synthesis of trisubstituted isoxazoles 49



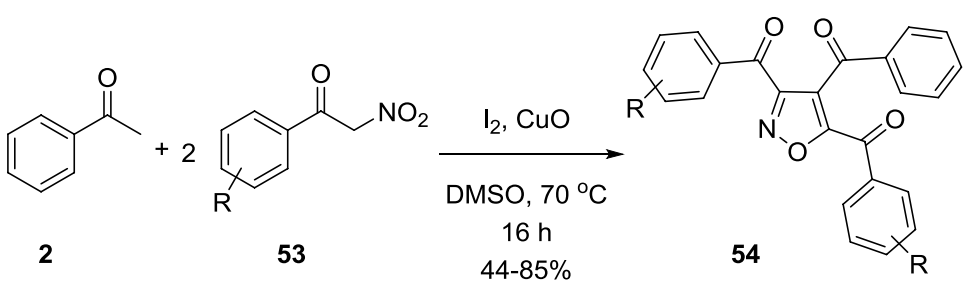
R^1= H, 4-Me, 4-MeO, 4-NO_2, 4-Cl, 4-Br, 4-F



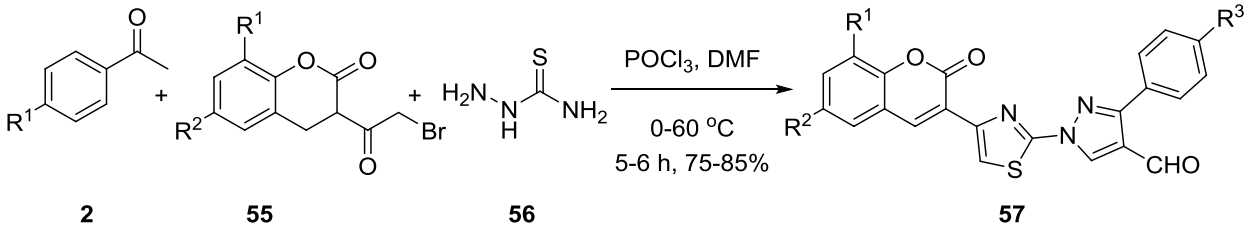
R^1= H, 4-Me, 2-Me, 4-OMe, 4-F, 4-Cl, 4-C_6H_5
R^2= Et, OMe, F
R^3= 4-OMe, 3-Cl

Scheme 15 One-pot synthesis of 5-benzyl-4,5-dihydropyrazoles 52

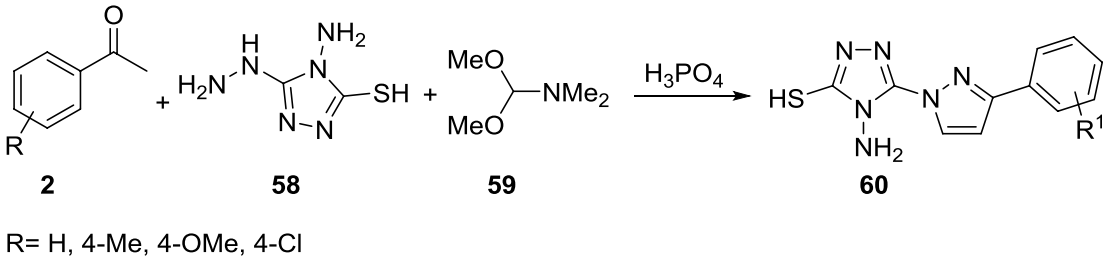
Scheme 16 Synthesis of isoxazoles 54



R= C_6H_5, 4-MeC_6H_4, 4-$MeOC_6H_4$, 4-$NO_2C_6H_4$, 4-ClC_6H_4, 4-BrC_6H_4



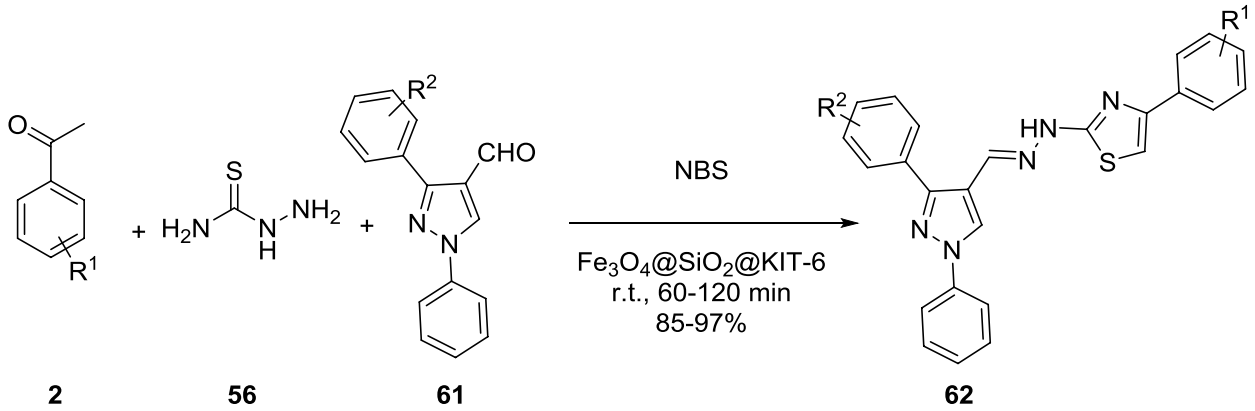
Scheme 17 Synthesis of coumarin-substituted thiazolyl-3-aryl-pyrazole-4-carbaldehydes 57



Scheme 18 Synthesis of coumarin-substituted thiazolyl-3-aryl-pyrazole-4-carbaldehydes 60

N,*N*-dimethylformamide-dimethylacetal (DMF-DMA) **59** in ortho-phosphoric acid as a solvent to obtain the target products **60** (Scheme 18). Triazolethiones possess various biological activities including anticancer, antiviral, anti-inflammatory, antiproliferative, antifungal, antidepressant and antioxidant.

Three-component condensation reaction of acetophenone derivatives **2**, thiosemicarbazide **56**, various aldehydes **61** and *N*-bromosuccinimide (NBS) **56** as a substrate instead of haloacetophenones in the presence of (KIT-6) mesoporous silica-coated magnetite nanoparticles as catalyst at room temperature was carried out by Nikpassand et al. [41] to achieve a series of benzothiazole derivatives **57** in the high yield (Scheme 19).



Scheme 19 Synthesis of benzothiazole 62

The synthesis of 1,3-dioxolanes **64** has been developed through the acetalization reaction of various acetophenone **2** and glycerol **63** in the presence of $FeCl_3{\cdot}6H_2O$ in tetrahydrofuran (THF) at 60 °C in the excellent yield (Scheme 20) [42].

Acetophenone derivatives **2** were reacted with thiourea **65** in the presence of HX/DMSO (X = Br or I) liquid system as the halogenating agent in EtOAc at 60 °C in which 2-aminothiazoles **66** and other analogous heterocyclic compounds were obtained in high yields (Scheme 21) (Table 2, entry 1) [43]. This reaction was also performed using *N*,*N*,*N*′,*N*′-tetrabromobenzene-1,3-disulfonamide [TBBDA], poly(*N*,*N*′-dibromo-*N*-ethylbenzene-1,3-disulfonamide) [PBBS] (TBBDA-MNPs@SiO_2-Pr-AP) [44] and I_2/CuO [45] as a catalyst. The efficiency of various conditions in the synthesis of 2-aminothiazoles **66** is compared in Table 2.

Liu et al. [48] described the synthesis of 2-aryl benzothiazole **69** via one-pot reaction of acetophenone **2**, aniline derivatives **67** and elemental sulfur **68** in the presence of iodine as catalyst (Scheme 22).

Alanthadka et al. [49] accomplished the one-pot reaction of acetophenone **2** and benzylamine **70** in the presence of *N*-heterocyclic carbene as a catalyst and under the solvent-free condition for the synthesis of imidazoles **71** (Scheme 23).

In 2019, Han et al. [50] applied poly(vinylbenzyltrimethylammonium hydroxide) resin (Amberlite 717) as a catalyst in the reaction of acetophenone **2** and ethylene glycol **72** for the synthesis of α-bromoacetal **73** in excellent yield (Scheme 24).

Khan et al. [51] synthesized various products including benzoxazole, benzothiazole and benzimidazole **74** via an interesting cyclization of acetophenone **2** and 2-amino aniline derivatives **73** in the presence of SeO_2 as a catalyst at 100 °C (Scheme 25).

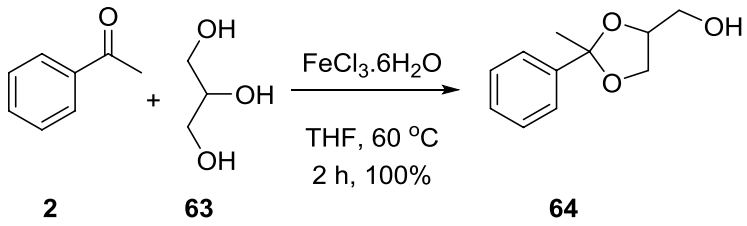
Farmani et al. [52] recorded the three-component reaction of acetophenone **2**, aldehydes **75** and thiosemicarbazide **56** in the presence of tetrabutylammonium hydroxide as catalyst under microwave irradiation for the synthesis of 4,5-dihydro-1H-pyrazole-1-carbothioamides **76** (Scheme 26).

Synthesis of six-membered rings

Six-membered rings containing O atom

The multicomponent reaction between acetophenone derivatives **2**, *α*-naphthol **77** and triethylorthobenzoate **78** catalyzed by bis[7-tert-butyl-2-anilinotropone] Ti complex in refluxing toluene afforded a new series of

Scheme 20 Acetalization of glycerol with ketones catalyzed by $FeCl_3$ $6H_2O$



Scheme 21 Synthesis of 2-aminothiazoles 66

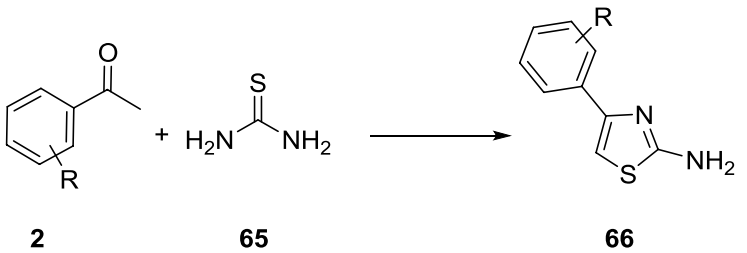
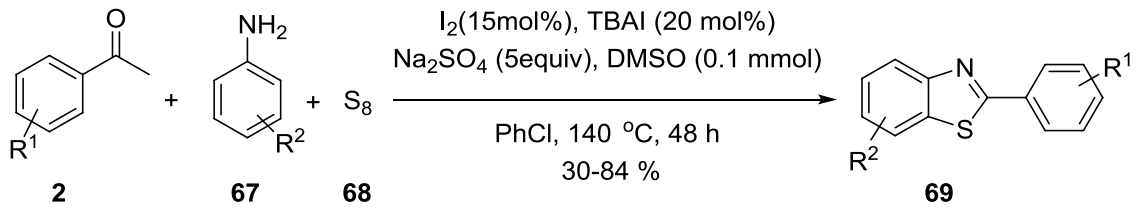
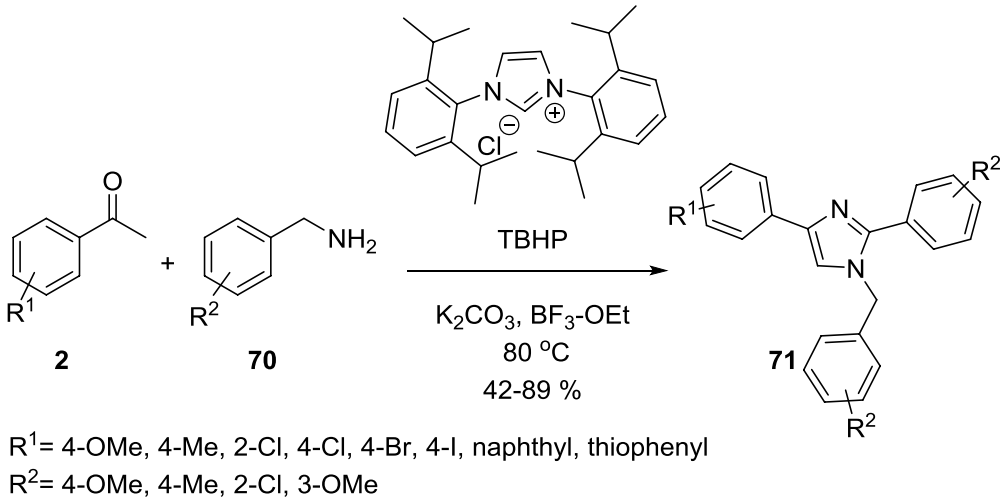


Table 2 Different reported methods for the synthesis of compounds 66

Entry	Catalyst	Solvent	Conditions	Time (h)	Yield (%)	References
1	HX/DMSO	EtOAc	60 °C	4	20–95	Zarnegar et al. [43]
2	TBBDA-MNPs@SiO_2-Pr-AP	–	80 °C	1–1.5	70–95	Ghorbani-Vaghei et al. [44]
3	I_2/CuO	EtOH	Reflux	1–13	48–90	Zhu et al. [45]

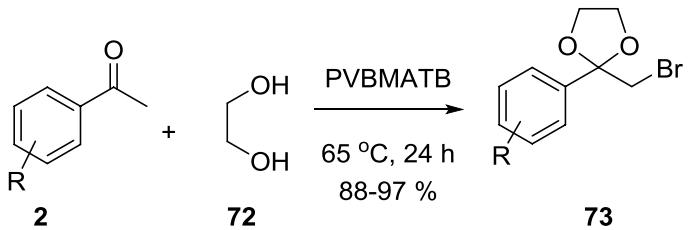


Scheme 22 Synthesis of 2-aryl benzothiazole 69



Scheme 23 Synthesis of imidazoles 71

Scheme 24 Synthesis of α-bromoacetal 73



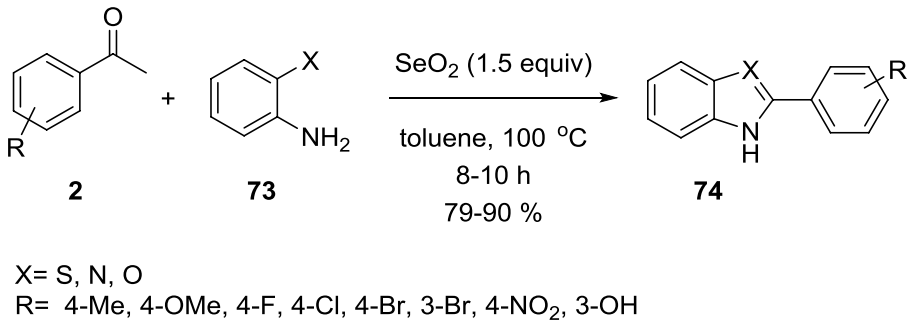
2-(4-aryl)-4-ethoxy-4-phenyl-4*H*-benzo[*h*]chromene derivatives **79** (Scheme 27) [53].

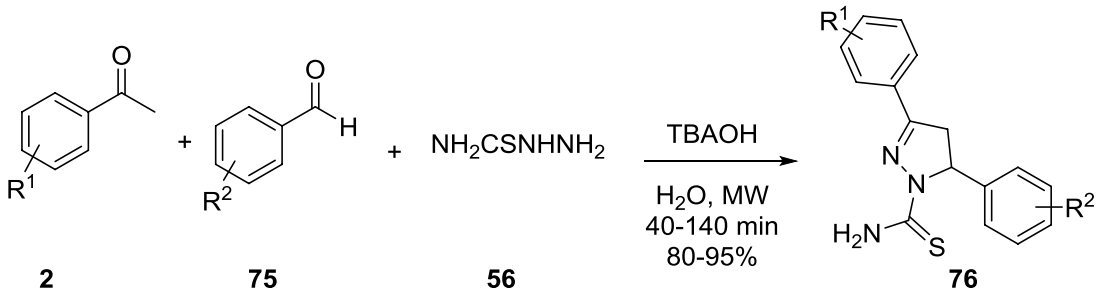
4-Phenacylidene flavenes **81** were synthesized by Bhattacharjee et al. [54] via the one-pot pseudo-three-component reaction between acetophenones **2** and salicylaldehydes **80** with a ratio of 2:1, respectively, in the presence of 20 mol% of bromodimethylsulfonium bromide (BDMS) as a catalyst in acetonitrile at room temperature (Scheme 28).

Reddy et al. [55] described an efficient method for the synthesis of pyrano[3,2-*c*]chromen-5(4*H*)-ones **82** via one-pot three-component reaction of acetophenone **2**, aldehydes **75** and 4-hydroxy-2*H*-chromen-2-one **12** without using catalyst and solvent under microwave irradiation (Scheme 29).

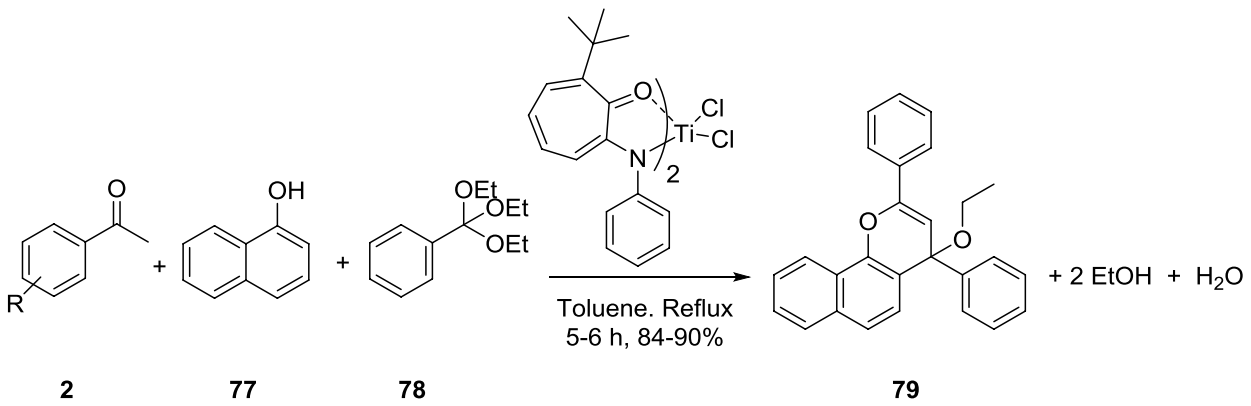
2-Amino-4-(3-methoxynaphthalen-2-yl)-6-phenyl-4*H*-pyran-3-carbonitrile **85** was synthesized via a three-component reaction of acetophenone **2**, malononitrile **83** and 2-methoxyquinoline-3-carbaldehyde **84** in the presence of NaOH as a catalyst in ethanol (Scheme 30) [56].

Scheme 25 Synthesis of various compounds including benzoxazole, benzothiazole and benzimidazole





Scheme 26 Synthesis of 4,5-dihydro-1H-pyrazole-1-carbothioamides 76

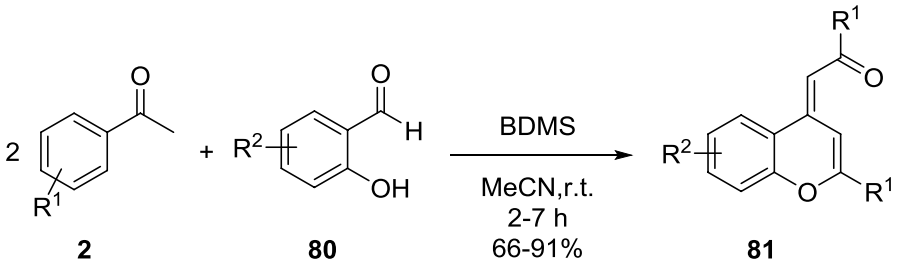


Scheme 27 Synthesis of 2-(4-aryl)-4-ethoxy-4-phenyl-4*H*-benzo[*h*]chromene derivatives 79

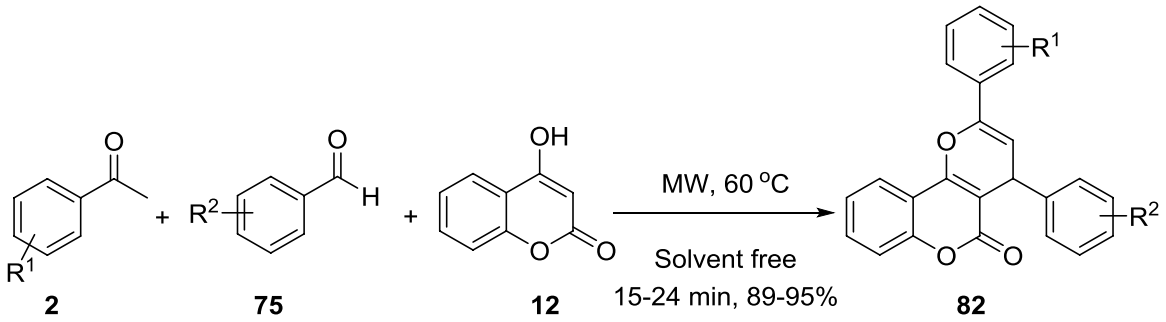
Sharifi et al. [57] applied a green method for the synthesis of chromene derivatives **88** via reaction of acetophenone **2**, 4-hydroxycoumarin **86** and aldehydes **87** in the presence of KF/clinoptilolite nanoparticles (KF/CP-NPs) under solvent-free conditions at 50 °C with high yield in low time (Scheme 31).

The bis(2-anilinotropone) Ti complex was applied as a catalyst for the synthesis of 1-ethoxy-3-(4-aryl)-1-phenyl-1H-benzo[f] chromenes **90** via multicomponent reaction of acetophenone derivatives **2**, *β*-naphthol **89**, and triethyl orthobenzoate **78** under refluxing in toluene as a solvent (Scheme 32) [58].

Scheme 28 Synthesis of 4-phenacylidene flavene derivatives 81

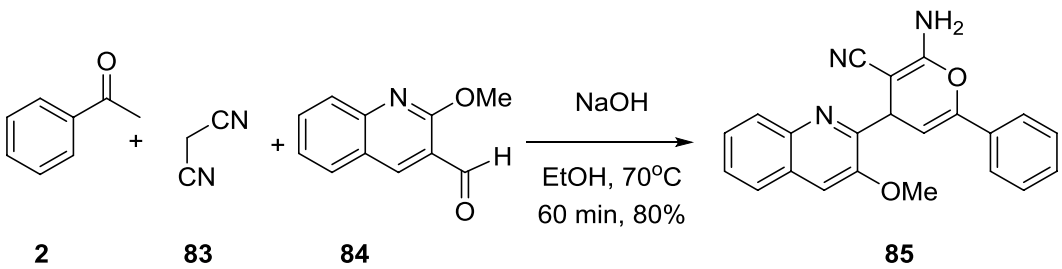


R^1= H, 4-Me, 4-OMe, 4-F, 4-Cl, 4-Br, 3-NO_2, 4-NO_2
R^2= H, 3-OEt, 3-OMe, 5-OMe, 5-Br



R^1= H, 4-Me, 4-OMe, 2-OMe, 2-Br, 4-Br, 4-Cl
R^2= H, 3-OMe, 4-OMe,3,4,5-$(OMe)_3$, 3-NO_2, 4-Br, 4-OEt

Scheme 29 Synthesis of pyrano[3,2-*c*]chromen-5(4*H*)-ones 82



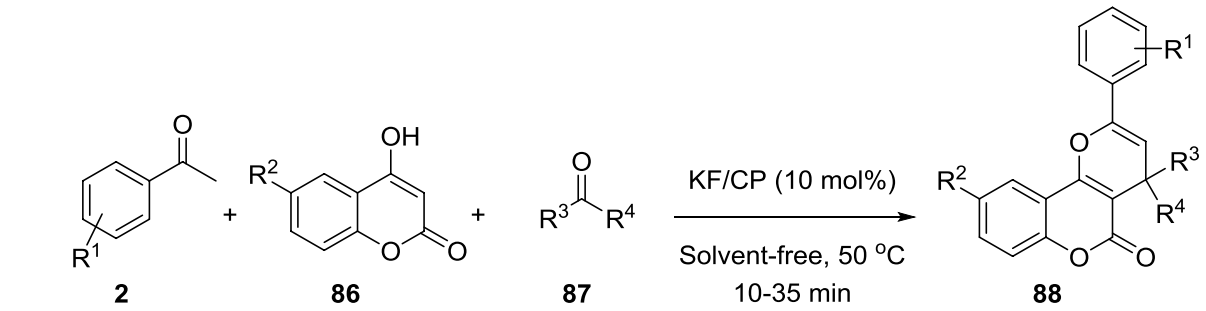
Scheme 30 Synthesis of pyrano[3,2-*c*]chromen-5(4*H*)-ones 85

Six-membered rings containing *N* atom

2-Chloronicotinonitriles **91** were synthesized by sequential cyclization and aromatization under Vilsmeier-Haack reaction of acetophenones **2** and malononitrile **83** (Scheme 33) [59].

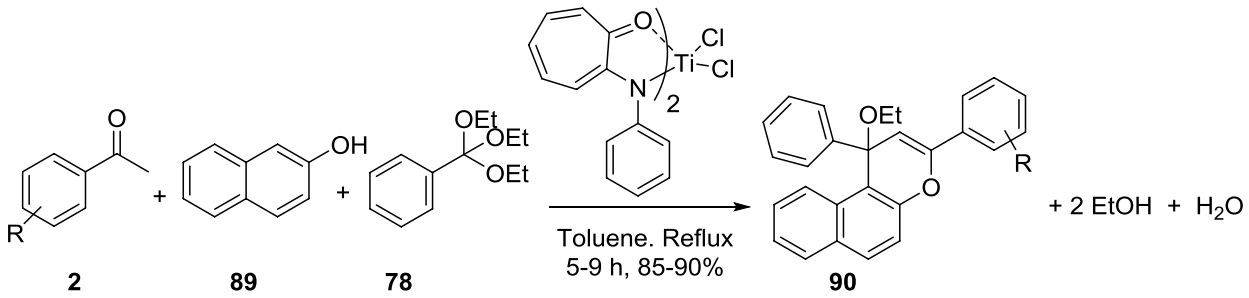
A plausible mechanism of this reaction is shown in Scheme 34. Initially, acetophenones **2** underwent Vilsmeier–Haack reaction in the presence of $POCl_3$ and DMF to afford chloromethyleneiminium salt intermediates **92**. Malononitrile **83** was added on the chloromethyleneiminium salt intermediates **93** to give intermediate **94**. As a result, by intramolecular cyclization, elimination of dimethylamine, 1,3-shift of the chlorine atom and by aromatization, respectively, afforded 2-chloropyridines **96** (Scheme 34) [59].

Synthesis of aminothieno[2,3-*b*]pyridine derivatives 99 was reported by the reaction of acetophenone derivatives 2 and 2-amino-3-thiophenecarbonitriles 98 in the presence of a catalytic amount of ytterbium (III) triflate (Yb(OTF)) under microwave irradiation (Scheme 35) [60]. Thieno[2,3-*b*]



R^1= H, 4-Me, 4-OMe, 4-Cl, 4-Br
R^2= H, Me
R^3= Ph, 4-Me-C_6H_4, 4-NO_2-C_6H_4, 4-Cl-C_6H_4, 3-OMe-C_6H_4, 4-OEt-C_6H_4
R^4= H, Me

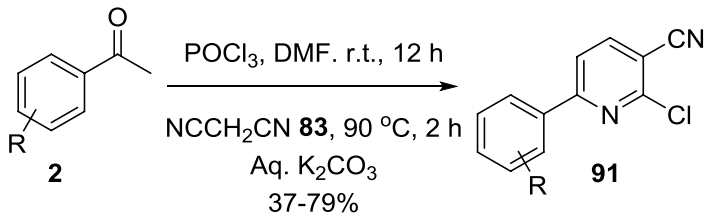
Scheme 31 Synthesis of chromene derivatives 88



R= H, 4-Me, 4-OMe, 2-OMe, 4-OH, 4-NO_2, 4-Br, 4-Cl, 4-F

Scheme 32 Synthesis of 1-ethoxy-3-(4-aryl)-1-phenyl-1H-benzo[f] chromenes 90

Scheme 33 Synthesis of 2-chloronicotinonitriles 91



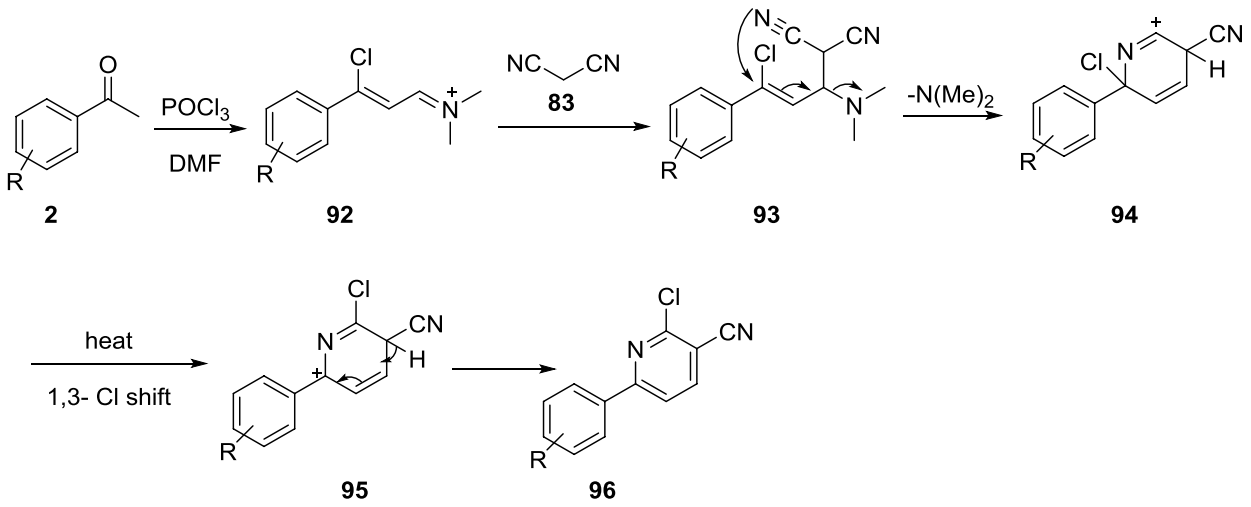
R= H, 4-Me, 4-OMe, 3-OMe, 4-Br, 4-Cl

pyridine derivatives have important structures in many alkaloids and biologically active natural products [60].

Two series of heterocyclic compounds including 4,6-diaryl-2-oxo-1,2-dihydropyridine-3-carbonitriles and their isosteric 2-oxopyridine derivatives 102 were synthesized by the multicomponent reaction of the 4-bromo acetophenone 2, aromatic aldehyde 74, malononitrile or ethyl cyanoacetate 100 and ammonium acetate 101 in ethanol under reflux condition (Scheme 36) [61].

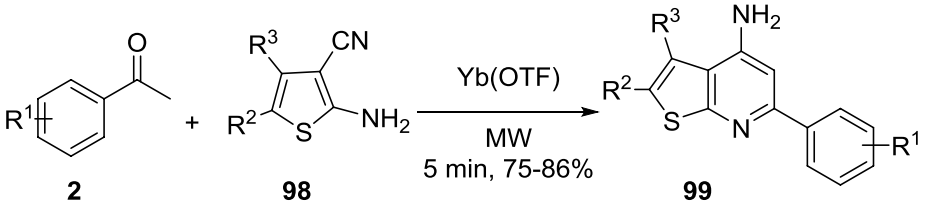
In the first step, ortho-nitro-chalcones 104 were obtained by the Claisene Schmidt condensation of acetophenone 2 and 2-nitrobenzaldehydes 75. Next 2-substituted-1,2,3,4-tetrahydroquinolines 103 has been achieved by the one-pot reductive intramolecular cyclization of ortho-nitro-chalcones with gaseous hydrogen in the presence of a Pd/C as a catalyst and CH_2Cl_2 as a solvent (Scheme 37) [62].

Safari et al. [46] reported in 2011 the Hantzsch condensation of acetophenones 2, aromatic aldehydes 75, ammonium

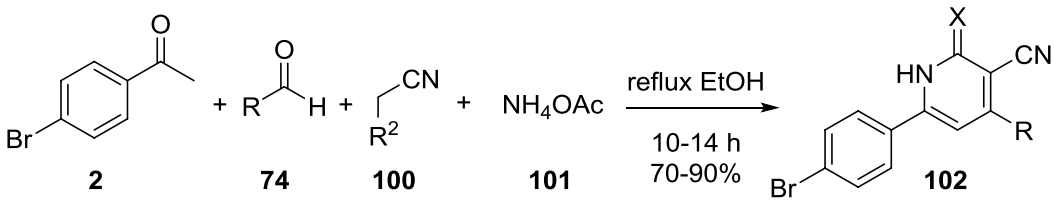


Scheme 34 A plausible mechanism for the synthesis of 2-chloropyridines **96**

Scheme 35 Synthesis of aminothieno[2,3-*b*]pyridine derivatives 99



R^1= H, 4-Me,4-OMe, 3-Cl, 4-Cl, 4-F, 4-NO_2
R^2,R^3= H, Me, -$(CH_2)_4$-, -$(CH_2)_3$-

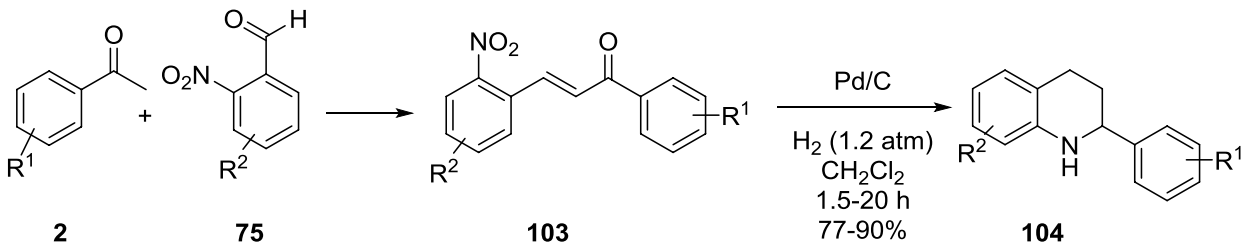


X= NH, O
R= 4-BrC_6H_4, 3,4-$Cl_2C_6H_3$, 2,4-$Cl_2C_6H_3$, 3,4,5-$(OMe)_3C_6H_2$, 2-Methyl-2-furanyl, 3-Pyridinyl, Indol-3-yl
R^2= CN, -CO_2Et

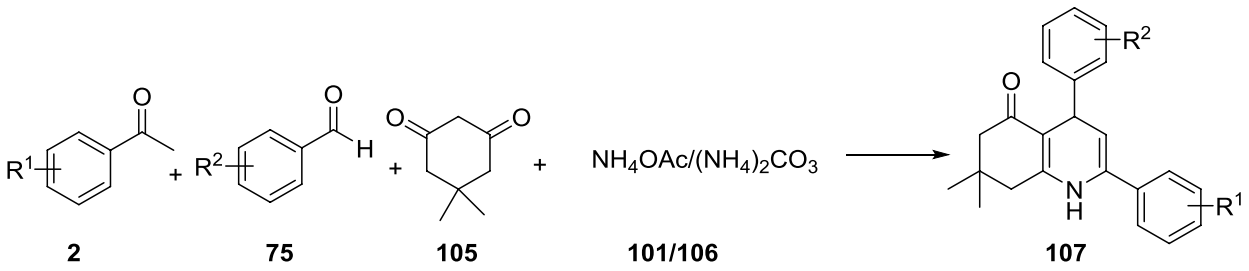
Scheme 36 Synthesis of 4,6-diaryl-2-oxo-1,2-dihydropyridine-3-carbonitriles 102

acetate 101 and dimedone 75 in the presence of a catalytic amount of Co nanoparticles at ambient temperature to produce C5-unsubstituted 1,4-dihydropyridines 107 in 30–97% yields (Scheme 38). In another study, Ray et al. developed a one-pot synthesis of C5-unsubstituted 1,4-dihydropyridines 107 using ammonium carbonate $(NH_4)_2CO_3$ 106 as nitrogen source at room temperature under solvent-free conditions (Table 3, entry 2) [47].

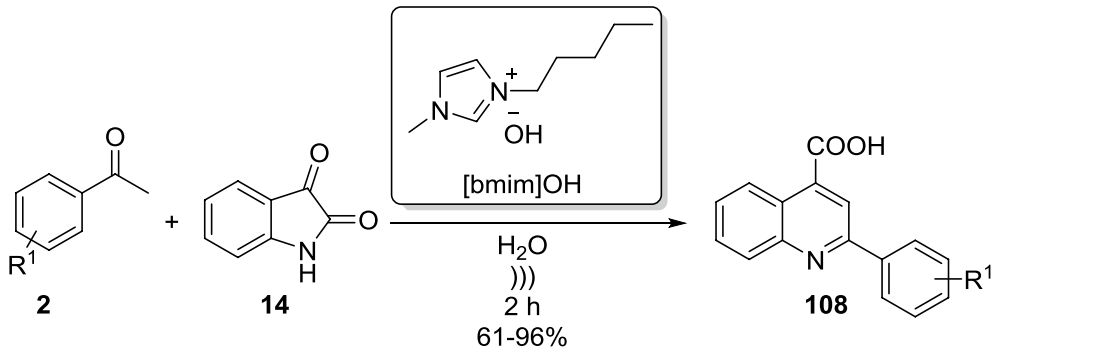
Kowsari et al. [63] investigated the synthesis of quinoline 108 via the condensation reaction of acetophenone 2 with isatin 14 in the presence of the basic ionic liquid (BIL) based on imidazolium cation under ultrasonic irradiation in



Scheme 37 Synthesis of tetrahydroquinolines 104



Scheme 38 Synthesis of 1,4-dihydropyridines 107



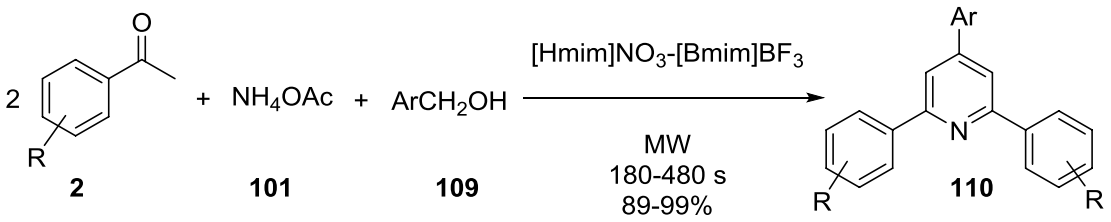
Scheme 39 Synthesis of quinoline compounds 104

Table 3 Comparison of different conditions in the synthesis of 1,4-dihydropyridines 107 at room temperature

Entry	Catalyst	Solvent	Time (h)	Yield (%)	References
1	Co-NPs	–	1–3	30–94	Safari et al. [46]
2	MSPA-10	H_2O	3	74–99	Ray et al. [47]

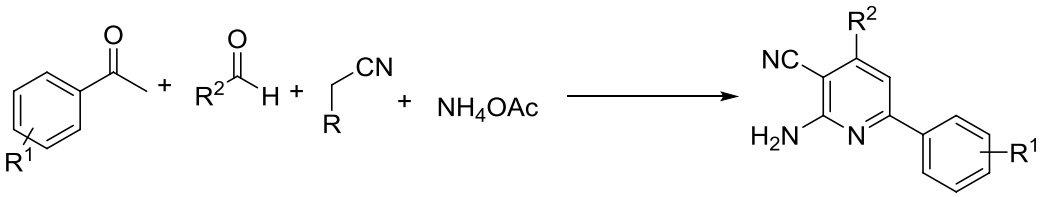
aqueous media with excellent yields (Scheme 39). Quinolines are the main constituents of many natural products, and also the quinoline nucleus plays an important role as an intermediate to design many pharmacologically active compounds [63].

A series of 2,4,6-triphenylpyrdine 110 was synthesized through a one-pot multicomponent reaction of acetophenone derivatives 2, ammonium acetate 101 and alcohols 109 using 1-methylimidazolium nitrate in 1-butyl-3-methylimidazolium tetrafluoroborate [Hmim]NO_3-[Bmim]BF_3 as a binary task-specific ionic liquid under microwave irradiation (Scheme 40) [64].



R= H, 4-Me, 4-Cl
Ar= C_6H_5, 4-BrC_6H_5, 2-FC_6H_5, 4-ClC_6H_5, 4-NO2C_6H_5, 4-MeC_6H_5, 2-MeC_6H_5, 4-OMeC_6H_5, 2-naphthyl, 2-thiophenyl, 5-Me-2-thiophenyl

Scheme 40 Synthesis of 2,4,6-triphenylpyrdine 110



R^1= C_6H_5, 4-OMeC_6H_4, 4-MeC_6H_4, 4-ClC_6H_4, 4-$NO_2C_6H_4$,3-$NO_2C_6H_4$, Et
R^2= H, 4-OMe, 4-OH
R= CN, CO_2Et

Scheme 41 The one-pot synthesis of 2-amino-3-cyanopyridines 113

A family of 2-amino-3-cyanopyridine derivatives 113 was synthesized by Jun Tang et al. [65] via a four-component reaction of acetophenones 2, aldehydes 75, malononitrile or ethyl cyanoacetate 111 and ammonium acetate 112 using [Yb(PFO)$_3$] as a catalyst under refluxing in EtOH (Scheme 41). This reaction was also developed using different catalysts including MgO [66], graphene oxide [67], [Bmim][BF_4] [68], PEG-400 [69], Fe_3O_4@SiO_2@$(CH_2)_3$-urea-benzimidazole sulfonic acid [70], $SrFe_{12}O_{19}$ [71], (Fe_3O_4@TiO_2@$O_2PO_2(CH_2)$NHSO_3H) [72], morpholine tags [73], catalyst free along with Ultrasound-promoted method and Cu@imineZCMNPs [74, 76]. A comparison of different catalysts is demonstrated in Table 4.

A series of novel 1-benzyl-2-butyl-4-chloroimidazole embodied 4-azafluorenone hybrids 116 was synthesized in excellent yields via one-pot condensation of acetophenone derivatives 2, ammonium acetate 101, 1,3-indanedione 114 and 1-benzyl-2-butyl-4-chloroimidazole-5-carboxaldehyde 115 under refluxing DMF in 77–86% yields (Scheme 42) [77].

2,4,6-Triarylpyridines 117 were synthesized via a multicomponent reaction of acetophenone 2, aldehyde 75 and ammonium acetate 101 under different conditions (Scheme 43). According to Table 5 various catalysts including wet 2,4,6-trichloro-1,3,5-triazine (Wet-TCT) [78], ZnO [79], pentafluorophenylammoniumtriflate (PFPAT) [80], $ZrOCl_2$ [81], nanotitania-supported sulfonic acid (*N*-TSA) [82], silica vanadic acid [SiO_2–$VO(OH)_2$] (SVA) [83], Fe_3O_4@TiO_2@$O_2PO_2(CH_2)_2NHSO_3H$ [72], chitosan-supported oxo-vanadium (CSVO) [84], $MgAl_2O_4$ [85], $HNTf_2$ [86] and PPA-SiO_2 [87] were reported to be effective in this reaction.

The multicomponent reaction of acetophenone 2, phenylacetic acids 118 and ammonium acetate 101 in the presence of VNU-22 {[Fe_3(BTC)-(BPDC)$_2$]·11.97H_2O} was accomplished by Doan et al. [88] (Scheme 44).

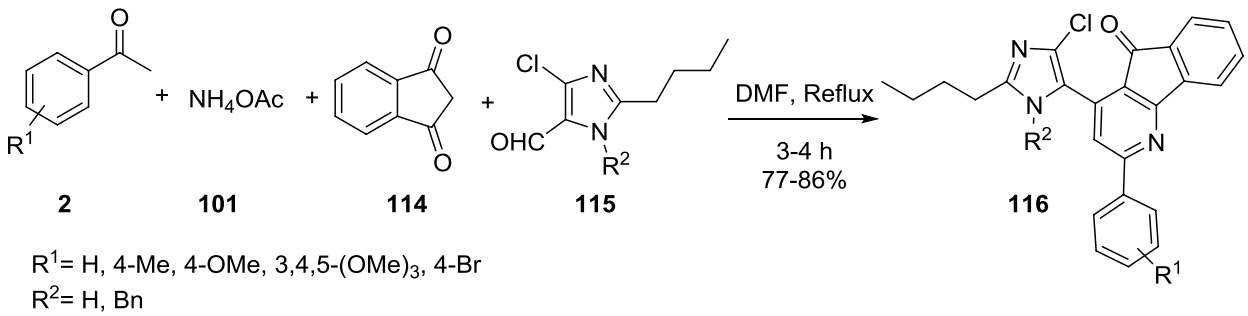
The tandem reaction of acetophenone derivatives 2 and simple nicotinamide salts 120 was carried out for the synthesis of substituted 2,7-naphthyridin-1(7*H*)-ones 121 in the high yield (Scheme 45) [89].

A series of quinoline derivatives 124 was synthesized by Friedländer reaction of acetophenone derivatives 2, 2-bromobenzaldehydes 122 and aqueous ammonia 123 as the nitrogen source in the presence of CuBr as a catalyst in high yields (Scheme 46) [90].

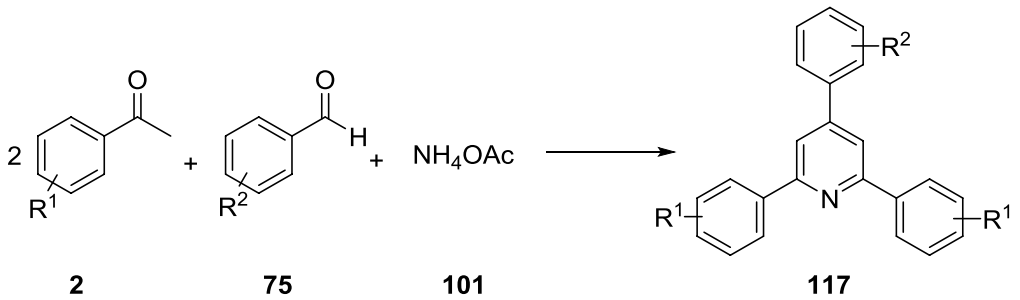
Table 4 Comparison of different conditions in the synthesis of 2-amino-3-cyanopyridines 113

Entry	Catalyst	Solvent	Conditions	Time (min)	Yield (%)	References
1	$[Yb(PFO)_3]$	EtOH	r.t.	1.5 h	85–90	Tang et al. [65]
2	MgO	DMF	Reflux	7–10	79–86	Sheibani et al. [66]
3	–	–	50 °C	10–35	75–99	Safari et al. [74]
4	GO[a]	H_2O	80 °C	5 h	75–97	Khalili et al. [67]
5	PEG-400	H_2O	80 °C	6 h	75–85	Mansoor et al. [68]
6	$[Bmim][BF_4]$	–	60 °C	4–5.5 h	85–94	Mansoor et al. [69]
7	$Fe_3O_4@SiO_2@(CH_2)_3$-urea-benzimidazole sulfonic acid	–	70 °C	10–40	70–92	Torabi et al. [70]
8	$SrFe_{12}O_{19}$	–	80 °C			Kheilkordi et al. [71]
9	$(Fe_3O_4@TiO_2@O_2PO_2(CH_2)NHSO_3H)$	–	90 °C	15–35	82–91	Zolfigol et al. [72]
10	Morpholine tags	–	80 °C	10–25	81–95	Kalhor et al. [73]
11	Cu@imineZCMNPs	–	80 °C	12–25	85–95	Yahyazadeh et al. [75]
12	–	–	MW	1–5	72–84	Amer et al. [76]

[a]Graphene oxide



Scheme 42 Synthesis of 4-azafluorenone hybrids 116



Scheme 43 Synthesis of 2,4,6-triarylpyridines 117

Sarmah et al. [91] developed an efficient method for the synthesis of pyrido[2,3-*d*]pyrimidine derivatives 126 by the multicomponent aza-Diels–Alder reaction of acetophenones 2, aromatic aldehydes 75 and uracil analogues 125 in the presence of Na_2CO_3 in DMF at 153 °C (Scheme 47).

Alinezhad et al. [92] in 2014 utilized Cu-doped ZnO nanocrystalline powder (10 mol%) in water/ethanol (50:50) as a solvent at room temperature to obtain indeno[1,2-*b*] pyridines 127 by multicomponent reaction of acetophenones 2, aldehydes 75, ammonium acetate 101 and 1.3-indandione 114 in 1.5-2 h (Scheme 48). Tapaswi et al. applied Ceric ammonium nitrate (CAN) as a catalyst in this reaction and obtained the products in good yields (Table 6, entry 2). This

Table 5 Comparison of different conditions in the synthesis of 2,4,6-triarylpyridines 117

Entry	Catalyst	Conditions (°C)	Time (min)	Yield (%)	References
1	ZnO	120	20–150	75–95	Maleki et al. [78]
2	*n*-TSA[a]	110	80–150	87–94	Shafiee et al. [79]
3	PFPAT[b]	120	2 h	84–94	Montazeri et al. [80]
4	$ZrOCl_2$	100	170–240	85–93	Moosavi-Zare et al. [81]
5	Wet-TCT[c]	130	4–7.5 h	58–86	Tabrizian et al. [82]
6	Silica vanadic acid [SiO_2–$VO(OH)_2$] (SVA)	130	45–60	81–88	Zolfigol et al. [83]
7	Fe_3O_4@TiO_2@$O_2PO_2(CH_2)_2NHSO_3H$	90	20–40	80–86	Zolfigol et al. [72]
8	CSVO[d]	130	55–75	53–88	Safaiee et al. [84]
9	$MgAl_2O_4$	120	3 h	70–90	Safari et al. [85]
10	$HNTf_2$[e]	80	30–60	84–96	Wang et al. [86]
11	PPA-SiO_2[f]	120	30–80	74–91	Davoodnia et al. [87]

[a]Nanotitania-supported sulfonic acid

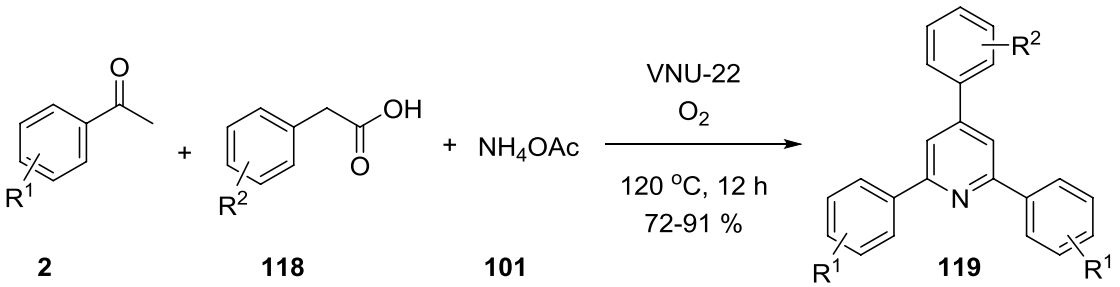
[b]Pentafluorophenylammoniumtriflate

[c]Wet 2,4,6-trichloro-1,3,5-triazine

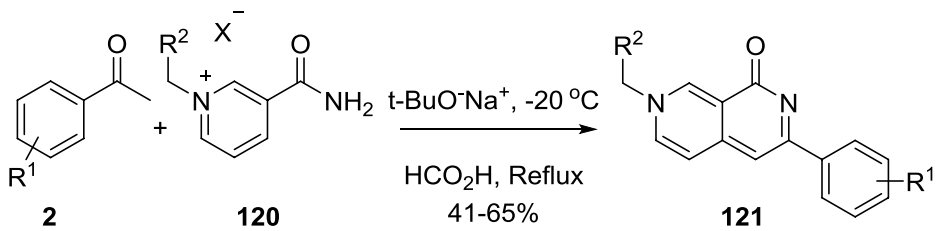
[d]Chitosan supported oxo-vanadium

[e]Triflimide

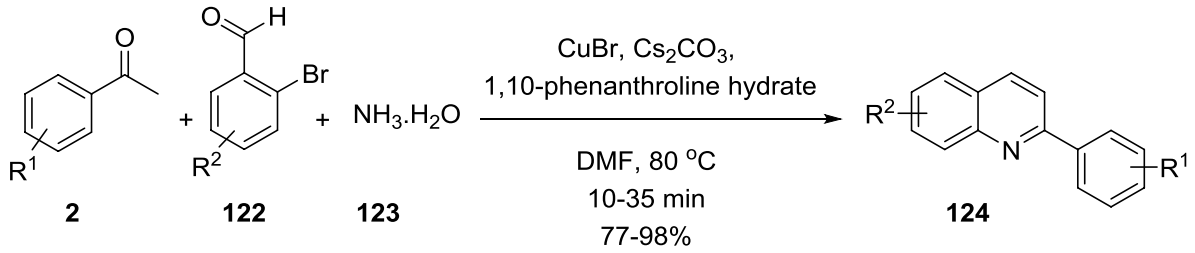
[f]Polyphosphoric acid-SiO_2



Scheme 44 Synthesis of 2,4,6-triarylpyridines 119

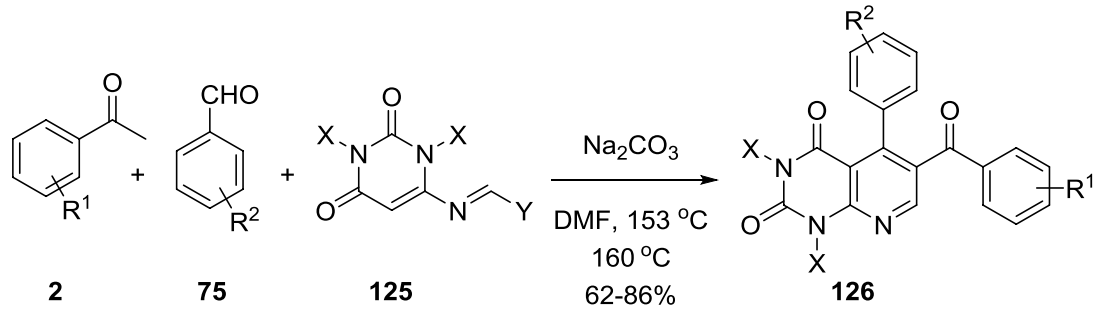


Scheme 45 Synthesis of substituted 2,7-naphthyridin-1(7*H*)-ones 121



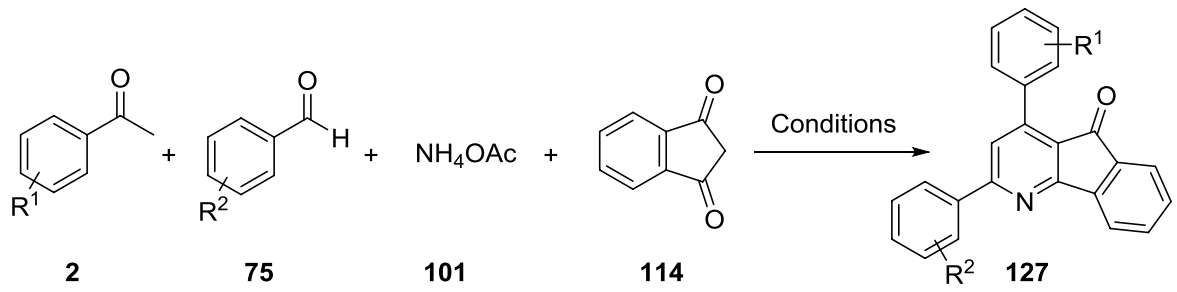
R^1= H, 4-OMe, 4-Me, 4-F, 4-CF_3, 4-NO_2, 3-Cl, 2-Cl
R^2= 2-Br-4-Me, 2-Br-4-Cl, 2-Br-4-F, 2-Br-4-CF_3, 2-Br-4-F

Scheme 46 Synthesis of quinoline derivatives 124



X= Me, Et
Y= $N(CH_2)_5$, $N(CH_2)_4$, $N(CH_2CH_2)_2O$
R^1= H, 4-F, 4-Cl, 4-Me, 4-OMe
R^2= H, 4-Me, 4-OMe, 4-Br, 3-Br, 2-Cl, 4-F

Scheme 47 Synthesis of pyrido[2,3-*d*]pyrimidines 126



Scheme 48 Synthesis of indeno[1,2-*b*]pyridines 127

Table 6 Comparison of the efficiency of various catalysts in the synthesis of indeno[1,2-*b*] pyridines 127

Entry	Catalyst	Solvent	Conditions	Time (h)	Yield (%)	References
3	Cu-doped ZnO	H_2O/EtOH	r.t.	1.5–2	85–95	Alinezhad et al. [92]
1	CAN[a]	EtOH	r.t.	3–5	84–94	Tapaswi et al. [93]
2	–	DMF	MW	6–15 min	57–89	Tu et al. [94]
4	–	TFE[b]	80 °C	2	85–95	Khaksar and Gholami [95]

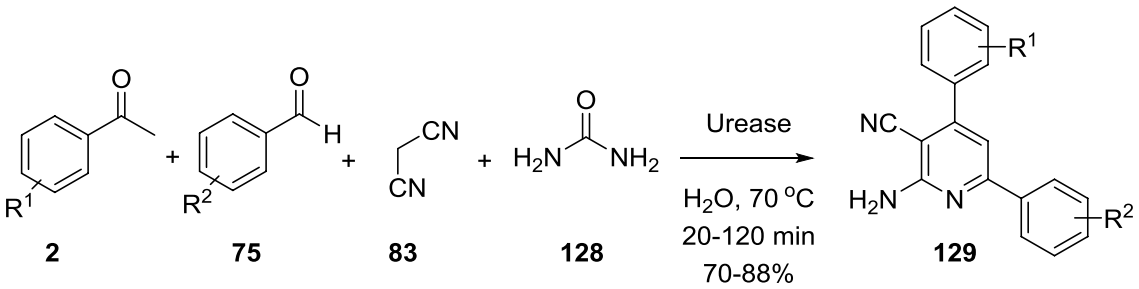
[a]CAN: ceric ammonium nitrate
[b]TFE: 2,2,2-trifluoroethanol

reaction was performed without any catalyst and obtained the products in good yields (Table 6, entry 3, 4).

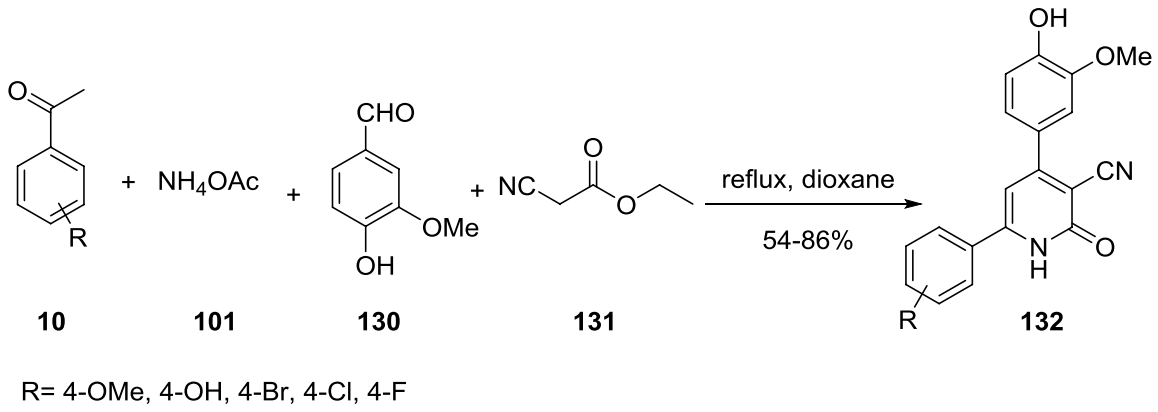
Tamaddon et al. [96] established the reaction of acetophenones 2, aldehydes 75, malononitrile 83 and urea 128 for the synthesis of 2-amino-3-cyanopyridines 129 using urease as the catalyst in the water at 70 °C in the high yield (Scheme 49).

Baluja et al. [97] described the synthesis of dihydropyridine derivatives 132 via condensation reaction of different substituted acetophenones 2, ammonium acetate 101, 4-hydroxy-3-methoxybenzaldehyde 130 and ethyl cyanoacetate 131 in refluxing dioxane (Scheme 50).

2-Phenyl pyridine 134 was synthesized via the cyclization of acetophenone 2 with 1,3-diamino propane 133 using palladium acetate in THF as a solvent (Scheme 51) [98]. Pyridine and its derivatives were evaluated for pharmaceuticals including etoricoxib (selective COX-II inhibitor), PMBI (antimalarial), topoisomerase type II inhibitor and zibotentan (endothelial antagonist) [98].

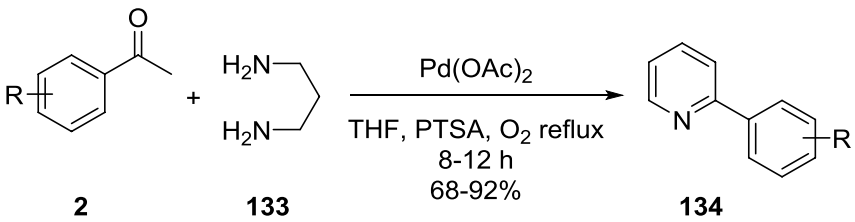


Scheme 49 Synthesis of 2-amino-3-cyanopyridines 129



Scheme 50 Synthesis of dihydropyridine derivatives 132

Scheme 51 Synthesis of 2-phenyl pyridine 134



Ladraa et al. [99] prepared a simple and convenient method for the synthesis of 3-cyanopyridine derivatives 136, 137 from the reaction of acetophenone derivatives 2, active methylene compounds 83, ammonium acetate 101 and 2-chloroquinolin-3-carbaldehydes 135 in the presence of PPh_3 as a catalyst at room temperature (Scheme 52).

The synthesis of various nitroarenes 139 has been developed through three-component ring transformation (TCRT) of acetophenone 2, ammonium acetate as nitrogen source 101 and dinitropyridone 139 in EtOH at 60 °C without using any catalyst (Scheme 53) [100].

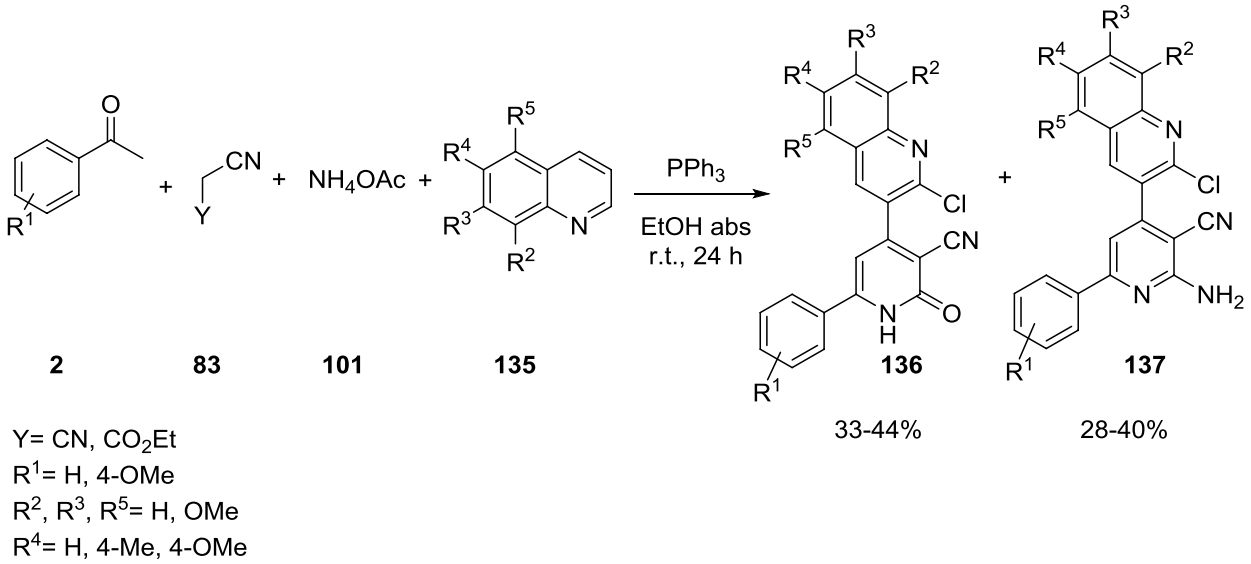
An efficient and environment-friendly procedure has been described for the preparation of substituted cyanopyridines 141 via four-component reaction of acetophenones 2, aromatic aldehydes 75, malononitrile 83 and sodium alkoxide 140 (molar ratio 1:1:1:1.3) in ethanol or methanol under MW (Scheme 54) [76].

A series of 2-substituted-1,8-naphthyridine derivatives 143 was synthesized by Friedlander condensation reaction of acetophenone derivatives 2 and 2-amino nicotinealdehyde 142 in refluxing methanol/water in the presence of potassium hydroxide as catalyst (Scheme 55) [101].

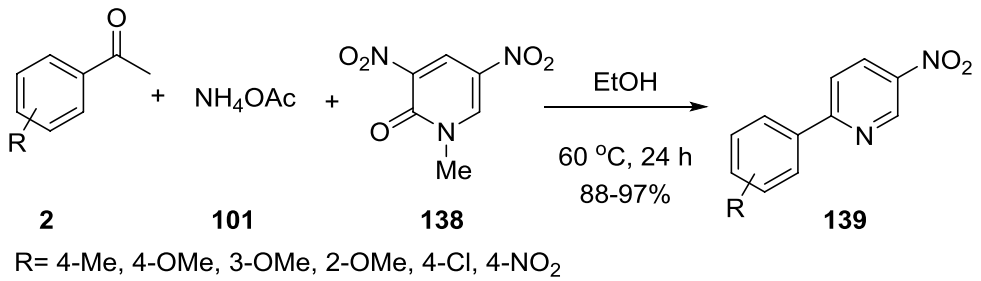
The reaction of acetophenone 2, alkyl amines 144 and malononitrile 145 was performed in the presence of KF/basic alumina as a catalyst for the synthesis of [1, 6] naphthyridines 146 [102] (Scheme 56).

The condensation reaction of acetophenone 2 and aniline derivatives 67 in the presence of CH_3SO_3H as a catalyst in DMSO solvent for the synthesis of quinolines 147 was reported by Jiang and co-works (Scheme 57) [103].

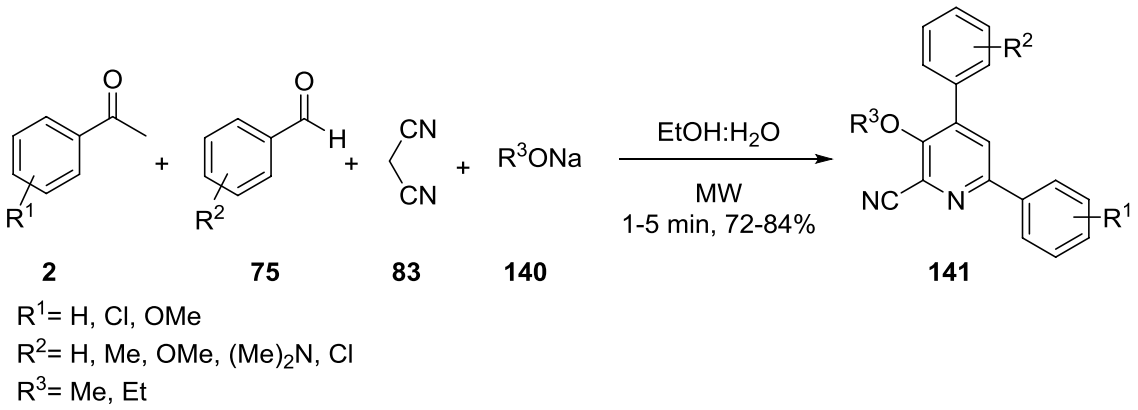
The quinoline derivatives 148 were obtained from the three-component reaction of acetophenone 2, aldehyde 75 and aromatic anilines 67 in the presence of CeO_2–TiO_2 under solvent-free conditions (Scheme 58) [104].



Scheme 52 Synthesis of 3-cyanopyridine derivatives 136, 137

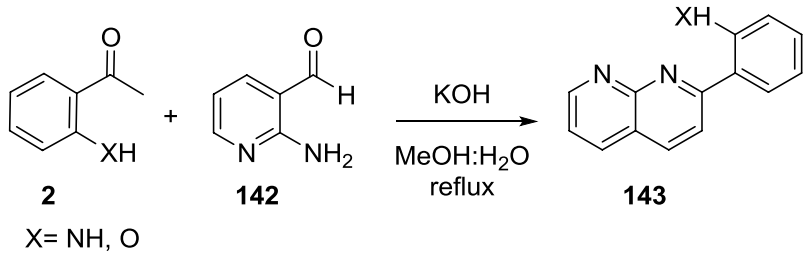


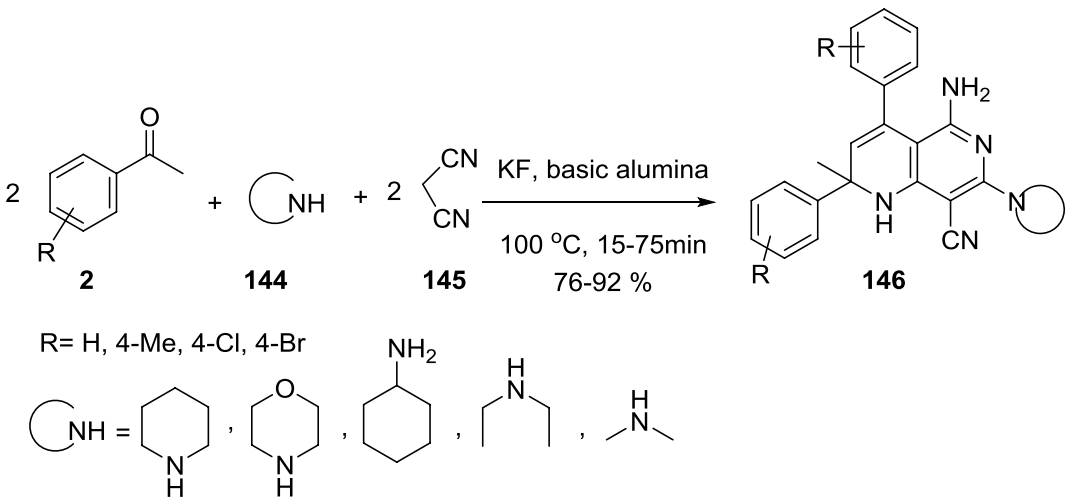
Scheme 53 Synthesis of nitroarenes 139



Scheme 54 Preparation of substituted cyanopyridines 141

Scheme 55 2-Substituted-1,8-naphthyridine derivatives 143





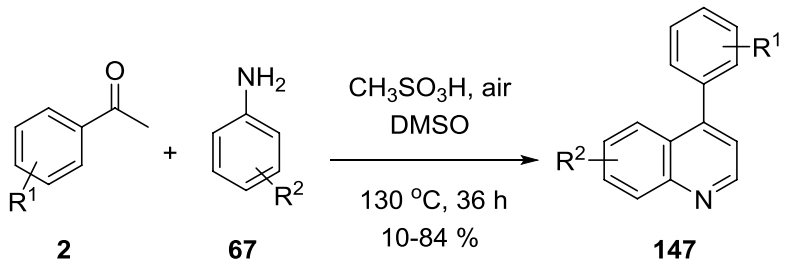
Scheme 56 Synthesis of [1, 6] naphthyridines 146

Six-membered rings containing two hetero atoms

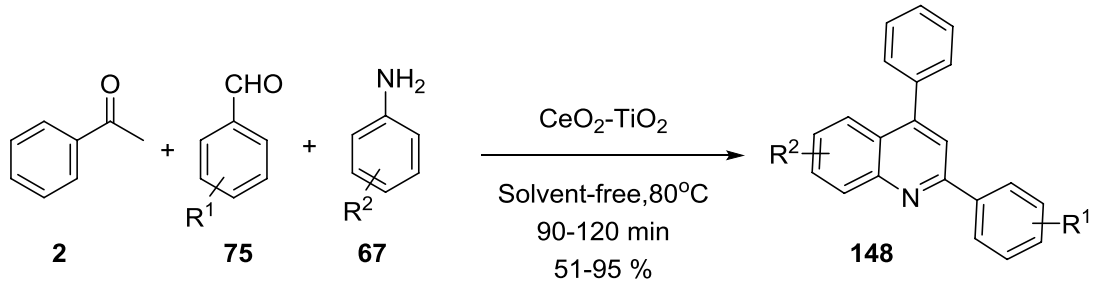
2-Methyl-2-phenyl-1,3-dithiane derivatives 150 were synthesized via the protection of acetophenone derivatives 2 with 1,3-propanedithiol 149 using the catalytic amount of yttrium triflate $Y(OTf)_3$ as a catalyst at room temperature (Scheme 59) [105]. The protection of carbonyl compounds played an important role during multistep syntheses in organic, medicinal, carbohydrate and drug design chemistry.

Wang et al. [106] studied a simple and efficient method for the synthesis of 5-unsubstituted 3,4-dihydropyrimidin-2-(1*H*)-ones 151 via the Biginelli-like three-component reactions of acetophenone 2, aldehyde 75 and urea 128 in the presence of $FeCl_3.6H_2O$ under refluxing in MeCN

Scheme 57 Synthesis of quinolines 147

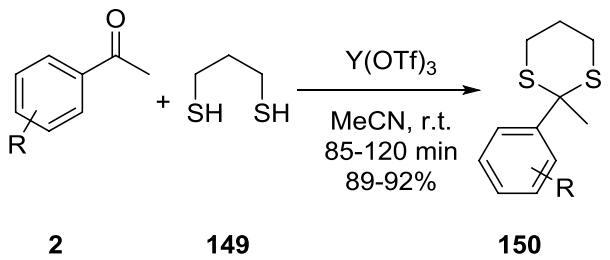


R^1= 4-OMe, 4-Me, 4-SMe, 4-Cl, 4-Br, 4-OH, 4-tBu, 4-iPr
R^2= H, 4-Me, 3-Me, 2,4-$(Me)_2$, 4-F, 4-Cl, 4-Br, 4-NO_2, 4-OH



R^1= H, 4-Me, 4-OMe, 4-Cl, 2-NO_2, 3-NO_2, 4-NO_2
R^2= H, 4-Me, 4-F, 4-OMe, 6-Cl

Scheme 58 Synthesis of quinoline derivatives 148



R= H, 4-OMe, 4-Cl

Scheme 59 Synthesis of 2-methyl-2-phenyl-1,3-dithiane derivatives 150

(Scheme 60). This reaction was also performed using a variety of catalysts such as MnO_2-CNTs [107], MnO_2 [107], TiO_2-MWCNTs [108], Na-atomazed [109], sulfonic acid functionalized silica (SBA-Pr-SO_3H) [110] and ionic liquid *N*,*N*,*N*′,*N*′-tetramethylethylenediaminium-*N*,*N*′-disulfonic acid hydrogen sulfate [TMEDSA][HSO_4]$_2$ [111] under different conditions. The efficiency of various conditions for this reaction is compared in Table 7.

The three-component condensation reaction of acetophenone 2, aromatic aldehydes 75 and thiourea 65 in the presence of inexpensive and efficient ceric ammonium nitrate (CAN) as a catalyst in PEG-400 was carried out by Singh et al. [112] to obtain 1,3-thiazine 152 with excellent yield (Scheme 61). 1,3-Thiazine and its derivatives were described as an inhibitor of Gram-negative bacteria and operated via inhibition of 4-diphosphocytidyl-2-C methyl-D-erythritol (IspE) kinase [112].

Magar et al. [113] synthesized 4,5,8*a*-triarylhexahydropyrimido[4,5-*d*]pyrimidine-2,7(1*H*,3*H*)-diones 153 with good-to-excellent yields via six-component reactions between acetophenone 2, aromatic aldehyde 75 and urea 128 in the presence of sulfated tin oxide (STO) as a reusable catalyst in ethanol at 60 °C (Scheme 62). Pyrimido pyrimidines have wide biological activities, such as antitumor, anti-inflammatory, antifungal and antibacterial activities [113].

A probable mechanism for the synthesis of substituted 2-aminopyrimidines 159 was shown in Scheme 58. Initially, condensation of acetophenone 2 with 3-hydroxybenzaldehyde 154 gave chalcone 155 and then reacted with carbamoyl chlorides 156 to generate carbamates intermediate 157

Scheme 60 Synthesis 5-unsubstituted 3,4-dihydropyrimidin-2-(1*H*)-ones 151

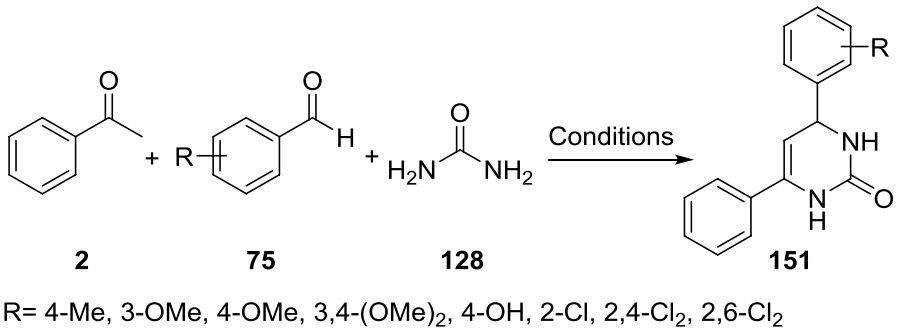


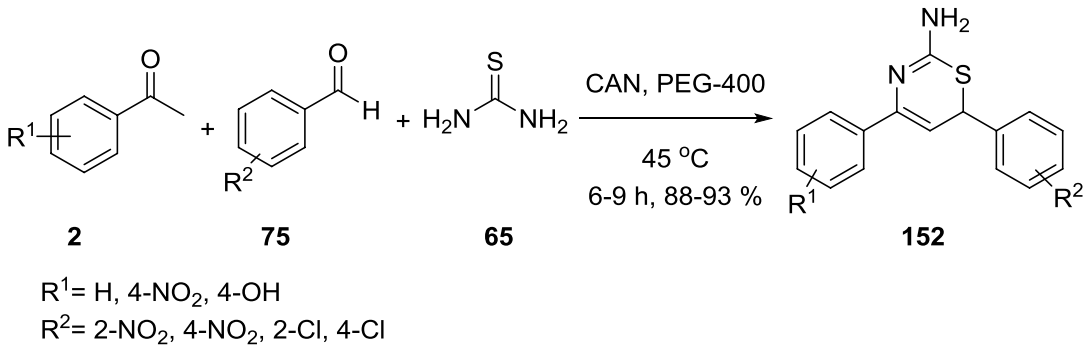
Table 7 The efficiency comparison of various catalysts in the synthesis of compound 151

Entry	Catalyst	Solvent	Conditions	Time (min)	Yield (%)	References
1	MnO_2-CNTs[a]	–	MW	5–25	87–97	Safari and Gandomi-Ravandi [107]
2	MnO_2	–	MW	30–80	58–73	Safari and Gandomi-Ravandi [108]
3	TiO_2-MWCNTs[b]	–	–	10–35	80–98	Safari and Gandomi-Ravandi [108]
4	$FeCl_3.6H_2O$	MeCN	Reflux	12 h	82–86	Pasha and Nagashree [109]
5	Na-atomazed	THF		10–14	86–90	Mohammadi Ziarani et al. [110]
6	SBA-Pr-SO_3H	–	110 °C	20–40	91–97	Khanivar and Zare [111]
7	[TMEDSA][HSO_4]$_2$c	–	80 °C	15–30	79–95	Singh et al. [112]

[a]Nanocomposites

[b]Metal oxide nanocomposites

[c]Ionic liquid *N*,*N*,*N*′,*N*′-tetramethylethylenediaminium-*N*,*N*′-disulfonic acid hydrogen sulfate



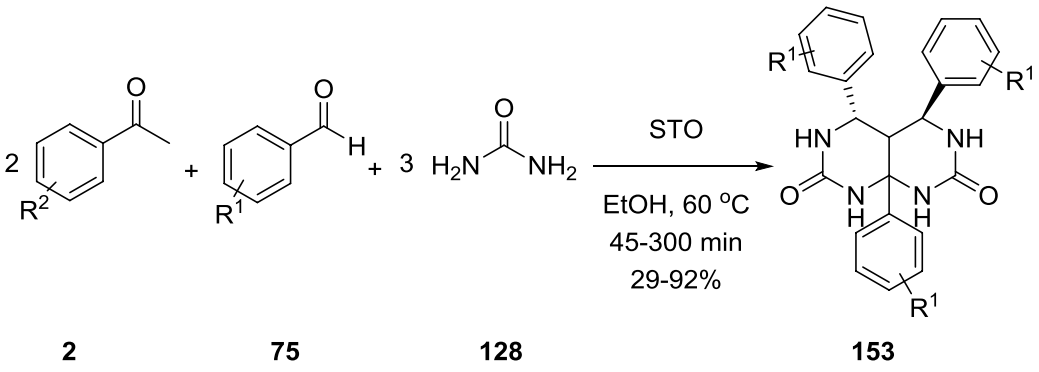
Scheme 61 Synthesis of 1,3-thiazines 152

which can be reacted with guanidine hydrochloride 158 in the presence of NaH in *N*, *N*-dimethylformamide (DMF) to produce 2-amino pyrimidines 159 (Scheme 63) [114].

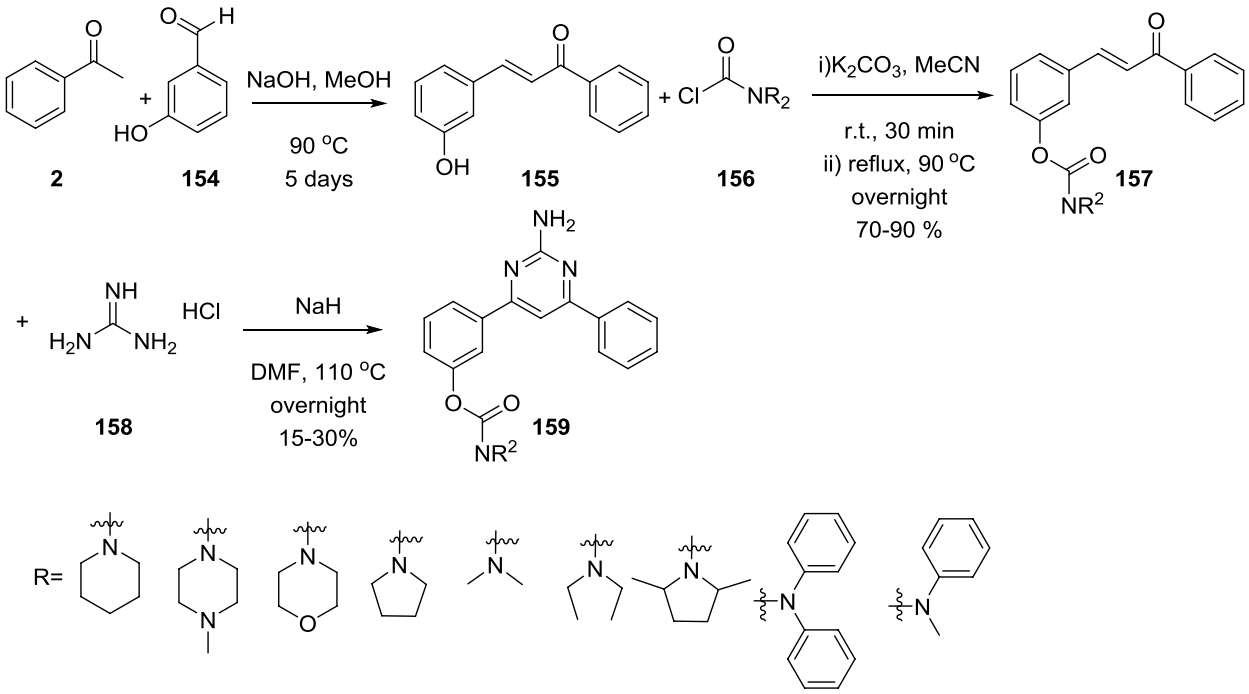
Thienothiophene-fused pyrimidine derivatives 161 were synthesized through the heterocondensation of acetophenone derivatives 2 with symmetric thieno[2,3-*b*]thiopheneoaminonitrile 160 under the reflux condition in ethanol for 2 h (Scheme 64) [115]. Thieno[2,3-*b*]thiophene ring skeleton and its derivatives possess a wide range of biological activities such as antiviral, antibacterial and anticancer activities [115].

Jadhav et al. synthesized the quinoxalines 164 which was generated by the reaction of acetophenone 2, succinamide 162 and aromatic amine 163 in the presence of I_2 in poly-ethylene glycol-400/water (2:1) as green solvent under microwave irradiation (Scheme 65) [116].

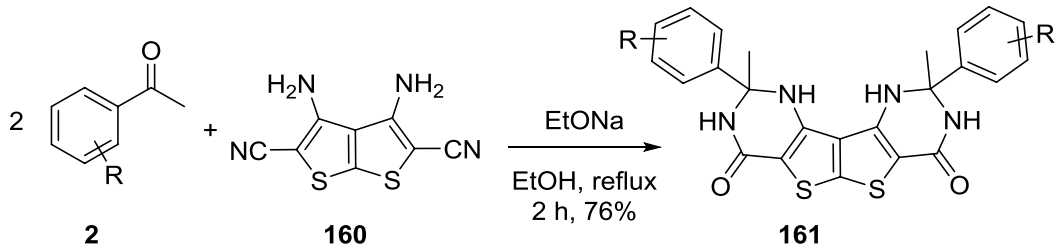
Aldol condensation of acetophenones 2 and aldehydes 75 gave intermediate chalcones 165 which were reacted with



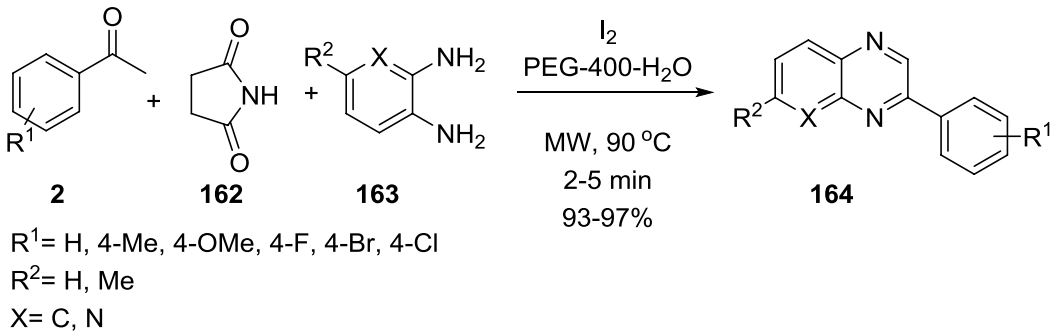
Scheme 62 Synthesis of 4,5,8 a-triarylhexahydropyrimido[4,5-*d*]pyrimidine-2,7(1*H*,3*H*)-diones 153



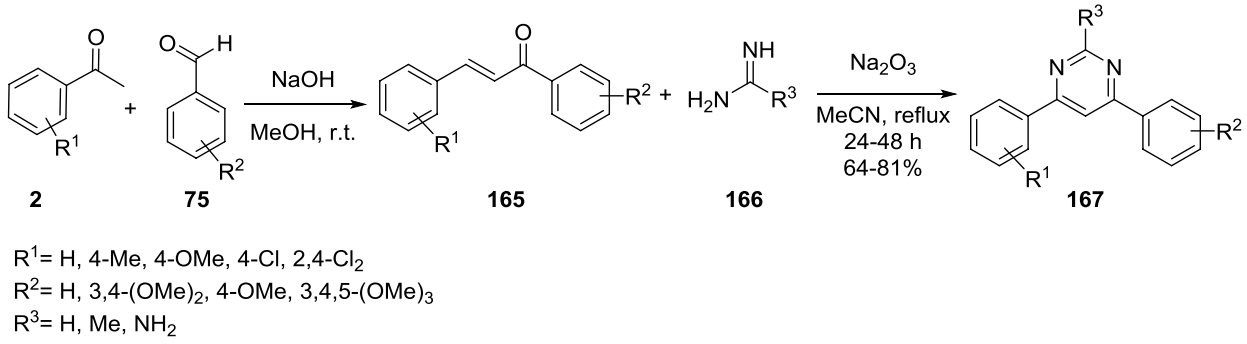
Scheme 63 Preparation of 2-aminopyrimidines 159



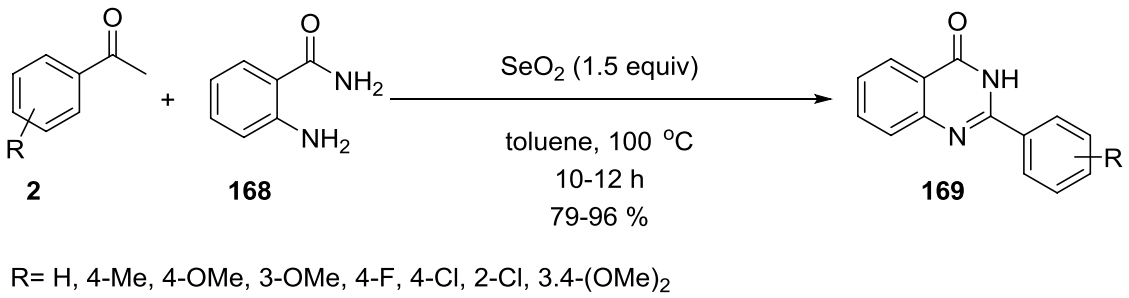
Scheme 64 Synthesis of thienothiophene-fused pyrimidine derivatives 161



Scheme 65 Synthesis of quinoxalines 164



Scheme 66 Synthesis of pyrimidine derivatives 167



Scheme 67 Synthesis of quinazolinone 169

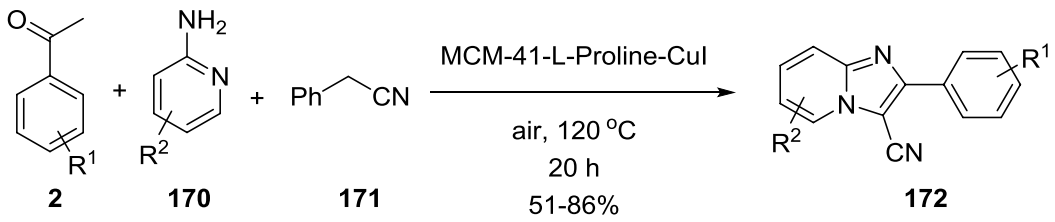
various compounds 166 to give pyrimidine derivatives 167 in 79–95% yields (Scheme 66) [117].

The reaction of acetophenone 2 and 2-aminobenzamidine 168 with the presence of SeO_2 as a catalyst for the synthesis of quinazolinone 169 was published by Khan et al. [51] (Scheme 67).

A class of 3-cyanoimidazo[1,2-a]pyridines 172 was achieved via three-component reaction of acetophenones 2, 2-aminopyridines 170, and benzyl cyanide 171 by using an MCM-41-anchored L-proline^{-} copper(I) complex [MCM-41-L-Proline-CuI] as a catalyst at 120 °C in high yields (Scheme 68) [118].

Synthesis of seven-membered rings

1-*H*-1,5-Benzodiazepine 174 was synthesized in good yields by the condensation of acetophenone 2 and phenylenediamine 173 in glycerol as a solvent in the catalyst-free condition (Scheme 69) [119]. As shown in Table 8, MIL-100 (v) [120], and amorphous mesoporous iron aluminophosphate (FeAlP-550) [121] were also used



Scheme 68 Synthesis of 3-cyanoimidazo[1,2-a]pyridines 172

Scheme 69 Synthesis of 1,5-benzodiazepine 174

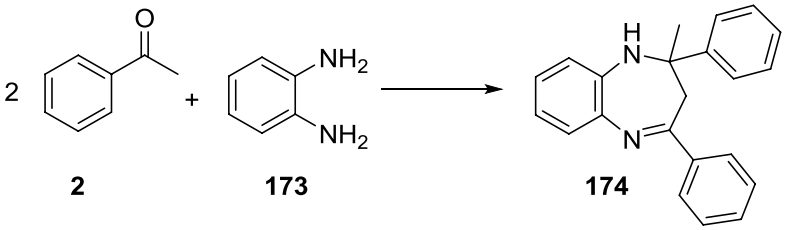


Table 8 Comparison of the efficiency of various catalysts in the synthesis of 1,5-benzodiazepine 174

Entry	Catalyst	Solvent	Conditions (°C)	Time (min)	Yield (%)	References
1	–	glycerol	90	4 h	96	Radatz et al. [119]
2	MIL-100(v)	MeOH	60	180	68	Timofeeva et al. [120]
3	FeAlP-550	–	80	75–120	83–92	Vijayasankar et al. [121]

Table 9 Comparison of different conditions in the synthesis of 5,7-diaryl-4,7-dihydrotetrazolo[1,5-*a*] pyrimidine derivatives 176

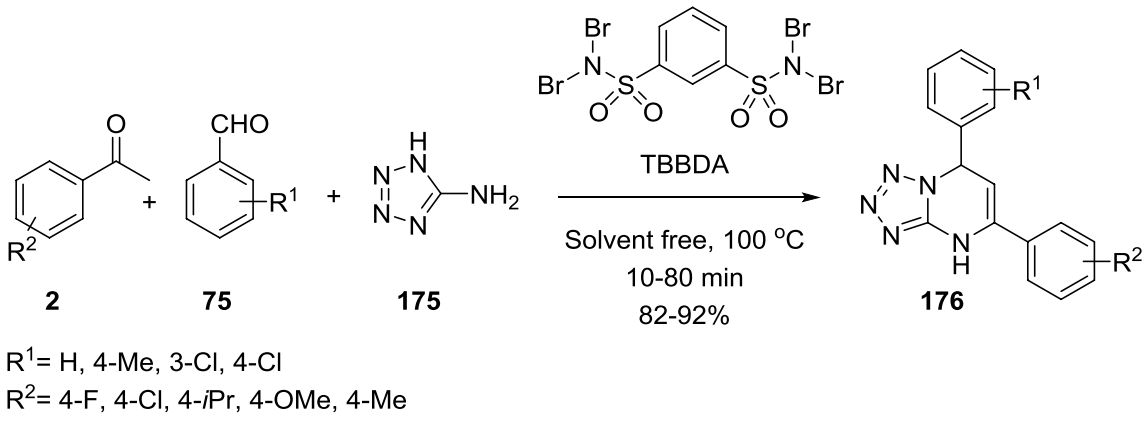
Entry	Catalyst	Solvent	Condition	Time (min)	Yield (%)	References
1	TBBDA[a]	–	100 °C	10–80	82–92	Ghorbani-Vaghei et al. [122]
2	AlCl3	MeCN	Reflux	3–5 h	88–92	Kour et al. [123]

[a]*N*,*N*,*N*′,*N*′-tetrabromobenzene-1,3-disulfonamide

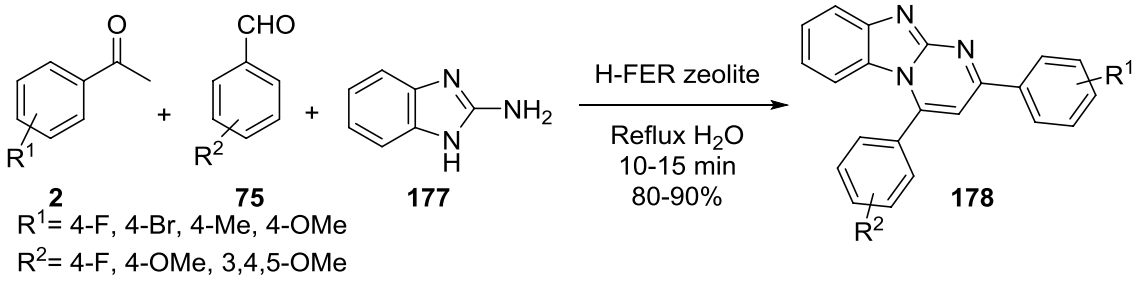
as catalysts in this reaction. Benzodiazepines and their derivatives have wide pharmacological properties such as anticonvulsant, analgesic, hypnotic, sedative and antidepressive agents [121].

Synthesis of fused heterocycle rings

A series of 5,7-diaryl-4,7-dihydrotetrazolo[1,5-*a*] pyrimidine derivatives 176 was obtained through the three-component reaction of acetophenones 2, aryl aldehydes 75 and 2-aminotetrazole 175 in the presence of *N*, *N*, *N*′, *N*′-tetrabromobenzene-1,3-di sulfonamide (TBBDA) as a catalyst under the solvent-free condition at 100 °C



Scheme 70 One-pot synthesis of 5,7-diaryl-4,7-dihydrotetrazolo[1,5-*a*] pyrimidine derivatives 176



Scheme 71 Synthesis of pyrimido[1,2-a]benzimidazole derivatives 178

[122]. $AlCl_3$ was used for the synthesis of 5,7-diaryl-4,7-dihydrotetrazolo[1,5-*a*] pyrimidine derivatives (Table 9, entry 2) [123]. According to Table 9, the best condition was in the presence of *N*, *N*, *N*′, *N*′-tetrabromobenzene-1,3-di sulfonamide (TBBDA) as a catalyst under solvent-free at 100 °C (Scheme 70) [122].

A class of pyrimido[1,2-a]benzimidazole derivatives 178 was synthesized from the multicomponent reaction of acetophenone derivatives 2, benzaldehyde derivatives 75 and heterocyclic amines 177 in the presence of H-ferrierite zeolite in short time and high yield (Scheme 71) [89].

Tris-dihydrotetrazolo[1,5-*a*]pyrimidine 180 was synthesized via a three-component reaction of acetophenone 2, 5-aminotetrazole 175 and trialdehyde (A15) 179 in the presence of *N*, *N*, *N*′, *N*′-tetrabromobenzene-1,3-disulfonamide (TBBDA) as a catalyst under solvent-free conditions in the excellent yield (Scheme 72) [122].

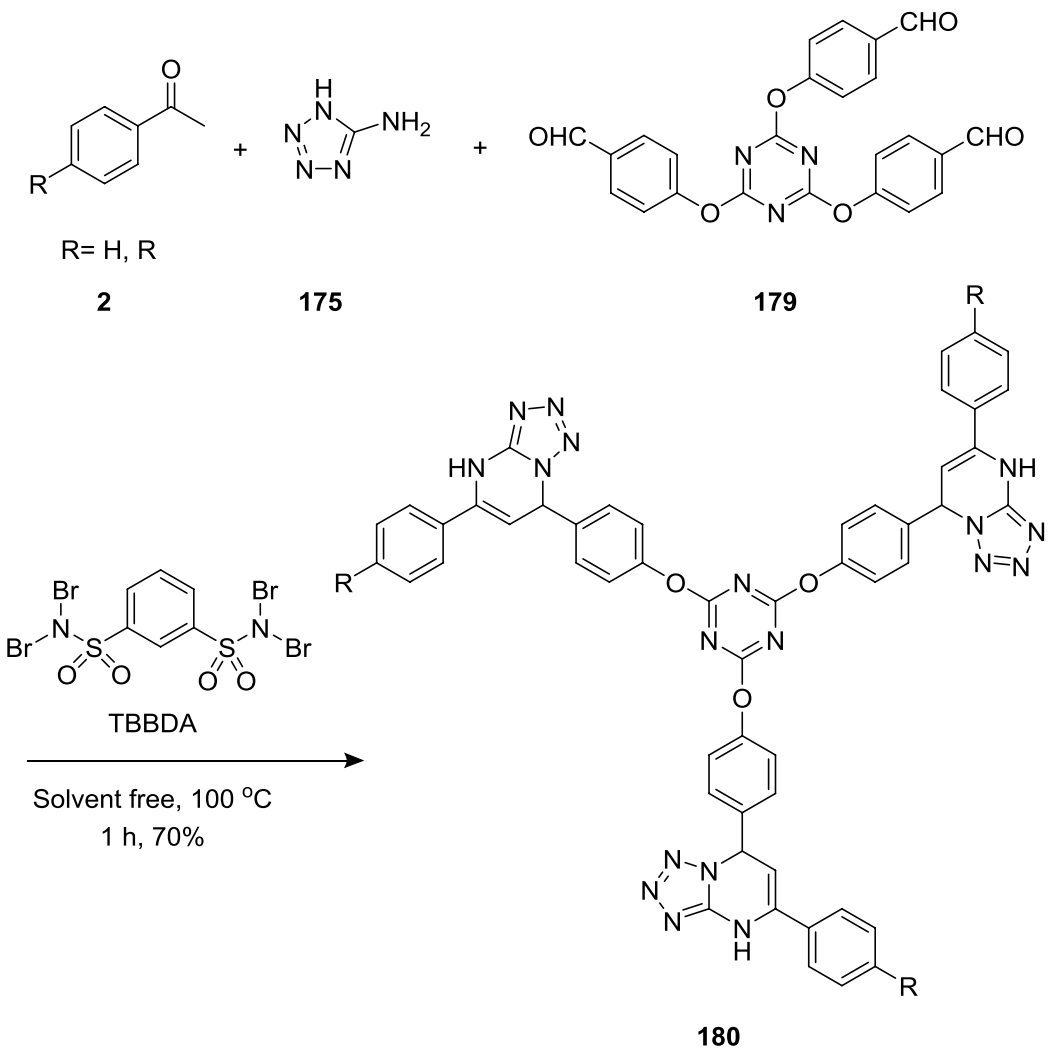
Qiao et al. [124] explained the synthesis of fused pyrazoles 183 through an efficient one-pot reaction of acetophenone 2, 2-phenylethynyl benzaldehyde 181 and hydrazine 182 in the presence of NaOMe under refluxing methanol (Scheme 73). The pyrazoles and their derivatives are an important class of bioactive heterocycles that display pharmaceutical properties, including anticancer agent, antipsychotic, auxin transport inhibitor and insecticidal activities.

The two-component condensation reaction of acetophenone 2 and 2-aminopyridine 173 in the presence of I_2-NH_4OAc in chloroform as a solvent was carried out by Kour et al. [125] to achieve 2-arylimidazo[1,2-*a*]pyridines 184 in high yield (Scheme 74).

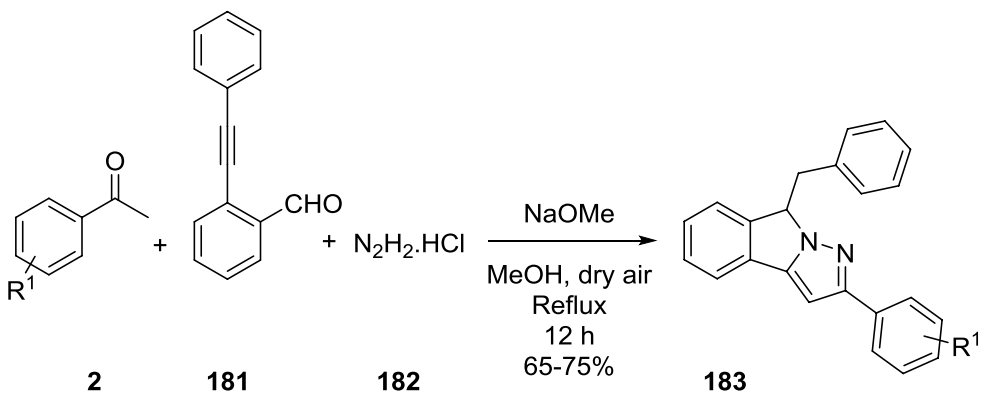
Pyrrolo[1,2-*a*]quinoxalines 186 were prepared via a three-component reaction of acetophenone derivatives 2, o-phenylenediamine 173 and 2-alkoxy-2,3-dihydrofuran 185 in the presence of boron trifluorideetherate as a catalyst (Scheme 75) [126].

Suresh et al. [127] worked on the multicomponent reaction of acetophenone 2, 5-aminotetrazole 175 and dimethylformamidedimethylacetal 187 in the presence of 1-butyl-3-methylimidazolium hydrogen sulfate [Bmim]HSO_4 ionic liquid to obtain fused tetrazolo[1,5-*a*]pyrimidine derivatives 188 in high yields (Scheme 76).

A new series of nitrogen bridgehead [1, 2, 4] triazolo[5,1-*c*] [1, 2, 4] triazepine derivatives 190 was synthesized by Moustafa's group via one-pot three-component reaction of acetophenone derivatives 2, aromatic aldehydes 75 and

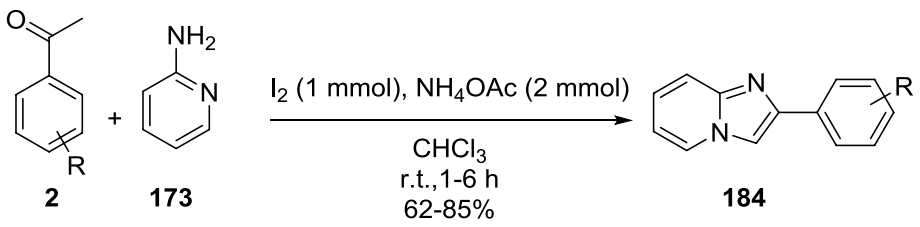


Scheme 72 Synthesis of tris-dihydrotetrazolo[1,5-*a*]pyrimidines 180

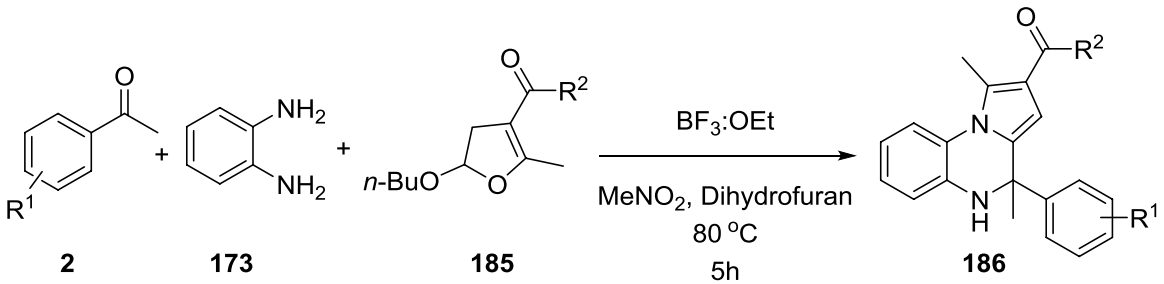


Scheme 73 Synthesis of fused pyrazoles 183

Scheme 74 Synthesis of 2-arylimidazo[1,2-*a*]pyridines 184



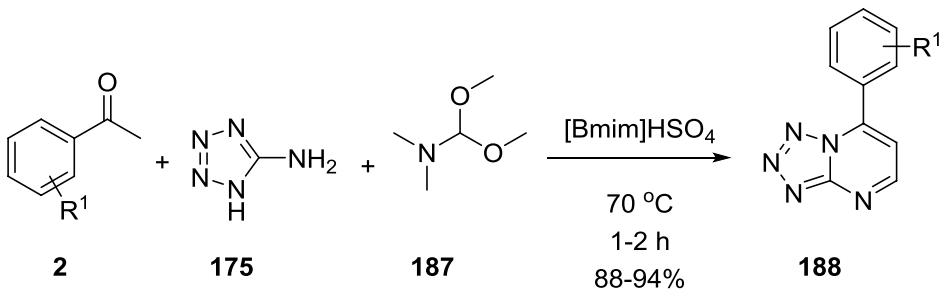
R= H, 4-Me, 3-OMe, 4-OMe, 3,4-$(OMe)_2$, 4-F, 4-Cl, 3-NO_2, 4-NO_2, (2-Br,4-Cl), (2-OH,5-Me)



R^1= 4-Me, 4-C_6H_5, 4-SMe, 4-*t*Bu, 4-Cl, 4-Br, 4-I, 4-CN, 3,4-$(OMe)_2$, 4-*n*C_5H_{11}
R^2= OMe, OEt, OCH_2CH_2OMe

Scheme 75 Synthesis of pyrrolo[1,2-*a*]quinoxalines 186

Scheme 76 Synthesis of fused tetrazolo[1,5-*a*]pyrimidine derivatives 188

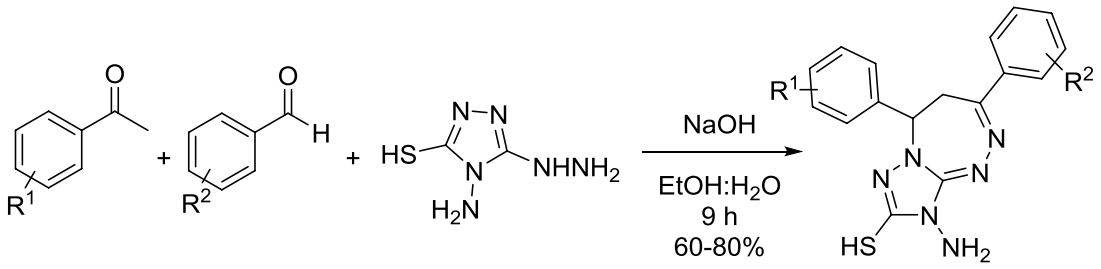


R= H, 4-Me, 4-OMe, 4-NO_2, 4-Cl, 4-F, 4-Br, 2-OH

polyfunctionaltriazole 189 using alcoholic sodium hydroxide solution (Scheme 77) [128].

Gálvez et al. [129] reported in 2018 two-component reaction between 4-chloro acetophenone 2 and 5-chloro-2-(1H-pyrazole-5-yl)aniline 191 in acetic acid at room temperature to produce 8-chloro-5-(4-chlorophenyl)-5-methyl-5,6-dihydropyrazolo[1,5-*c*]quinazoline 192 (Scheme 78).

The synthesis of thieno[2,3-d]pyrimidin-4-amines 195 was reported by Shi et al. [130] through a four-component reaction between acetophenone 2, formamide 193,

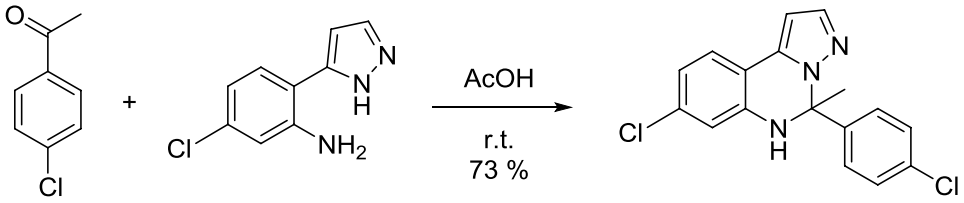


2 **75** **189** **190**

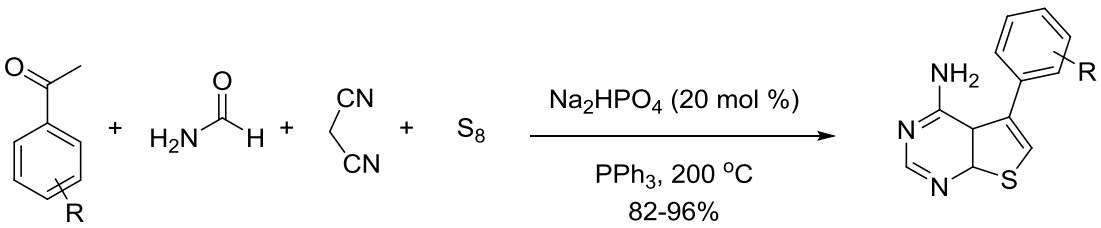
R^1= H, Cl, OMe
R^2= H, Me, OMe, Cl

Scheme 77 Synthesis of [1, 2, 4]triazolo[5,1-*c*][1, 2, 4]triazepine derivatives 190

Scheme 78 Synthesis of pyrazolo[1,5-*c*]quinazoline 192



2 **191** **192**

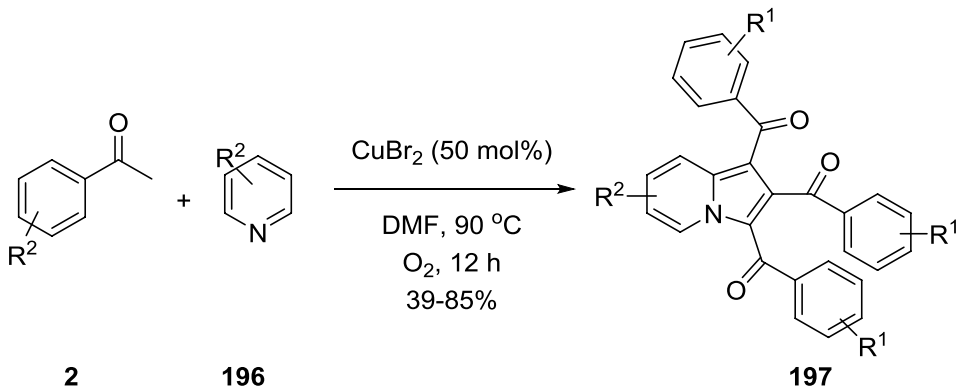


2 **193** **83** **194** **195**

R= H, 4-Me, 4-OMe, 4-F, 4-Cl, 4-Br, 4-I, 3-F

Scheme 79 Synthesis of thieno[2,3-d]pyrimidin-4-amines

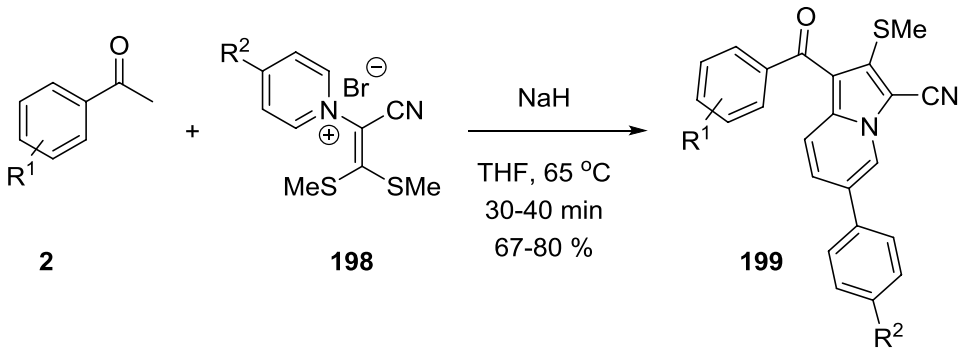
Scheme 80 Synthesis of 1,2,3-triaroylindolizines 197

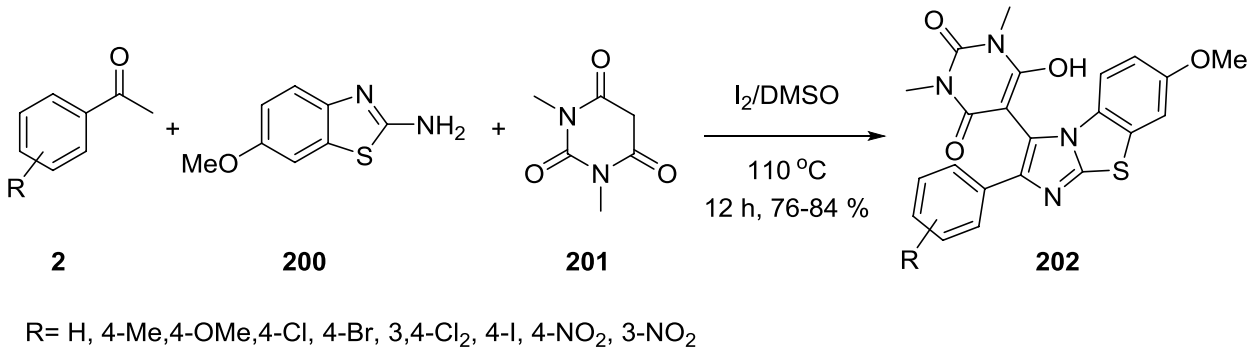


2 **196** **197**

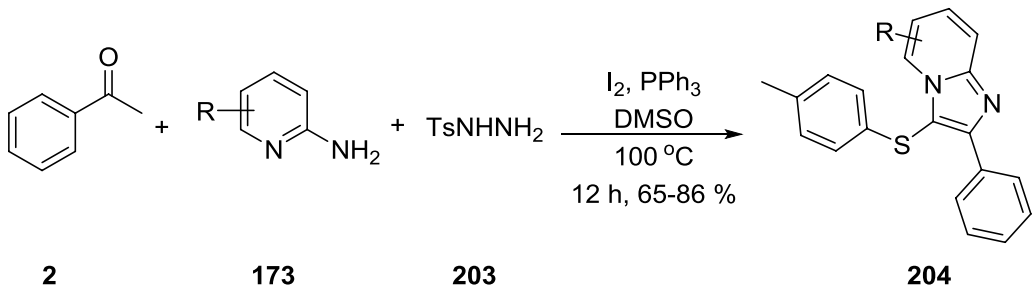
R^1= 4-F, 4-Cl, 3-Cl, 2-Cl, 4-Me, 4-Et, 4-OMe, 3-OMe, 2-OMe, 2.4-Me_2
R^2= 3-CO_2Me, 4-COPh, 4-H, 4-t-Bu, 4-CO_2Me

Scheme 81 Synthesis of the indolizine derivatives 199





Scheme 82 Synthesis of 2-arylbenzo[d]imidazo[2,1-b] thiazoles 202

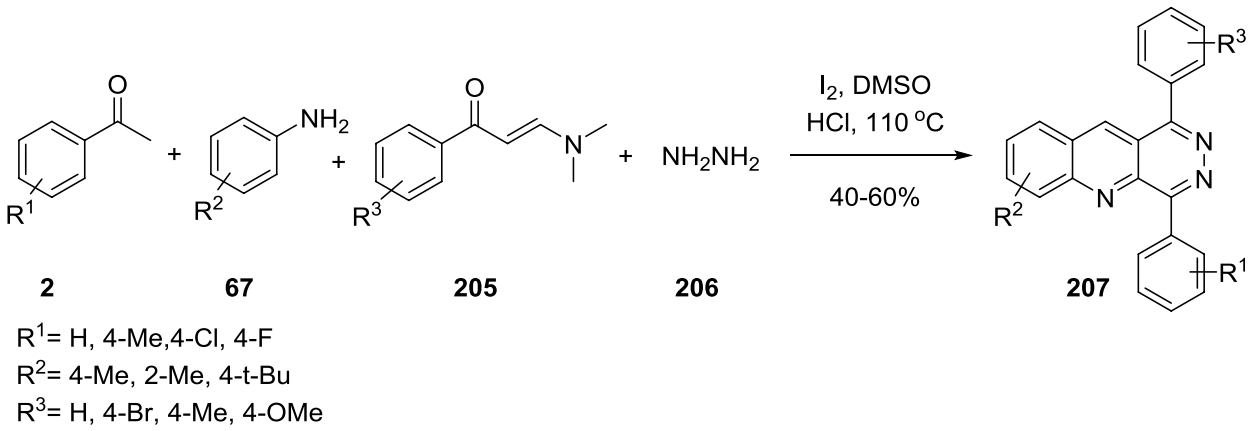


Scheme 83 Synthesis of 3-sulfenylimidazo[1,2-a]pyridines 204

malononitrile 83 and S_8 194 in the presence of Na_2HPO_4 as a catalyst at 200 °C (Scheme 79).

Sum et al. [131] synthesized a wide range of 1,2,3-triaroylindolizines 197 in excellent yield via the reaction of acetophenone 2 and pyridine derivatives 196 in the presence of $CuBr_2$ as a catalyst at 90 °C (Scheme 80).

Ramesh et al. [132] accomplished the synthesis of the indolizine derivatives 199 via the reaction of acetophenone



Scheme 84 Synthesis of pyridazino[4,5-b]quinolone skeletons 207

2 and 1-(1-cyano-2,2-bis(methylthio)vinyl)pyrdin-1-ium 198 in the presence of NaH at 65 °C in high yield (Scheme 81).

Synthesis of 2-arylbenzo[d]imidazo[2,1-b] thiazoles 202 was followed by a three-component reaction of acetophenone 2, 2-aminobenzothiazoles 200 and barbituric acids 201 in the presence of I_2 in DMSO (Scheme 82) [133].

The synthesis of 3-sulfenylimidazo[1,2-a]pyridines was studied by Hu et al. For the synthesis of 3-sulfenylimidazo[1,2-a]pyridines, the multicomponent reaction of acetophenone, 2-aminopyridine and 4-methylbenzenesulfonohydrazide was accomplished (Scheme 83) [134]

Various pyridazino[4,5-b]quinolone skeletons 207 were synthesized in 40–65% yields via three-component reaction of acetophenone 2, anilines 67, enaminones 205 and hydrazine 206 in the presence of I_2 as a catalyst at 100 °C (Scheme 84) [135].

Conclusions

In this review, different types of reactions which included acetophenone as a starting material have been studied. Also, we tried to highlight the application of acetophenone as a synthon in the synthesis of various heterocyclic systems.

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