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Arylidene Derivatives as Synthons in Heterocyclic Synthesis

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Abstract

This review describes the synthetic procedures for the preparation of arylideneacetophenones, arylidenecycloalkanones, 2-arylidene-1-indanones, 2-arylidene-1-tetralones, 2-arylidene-1-benzosuberones, aurones, 1-thioaurones, 3-arylidene-4-chromanones, 3-arylidene-1-thio-4-chromanones, 3-arylideneflavanones, 3-arylidene-1-thioflavanones, arylideneanilines, arylidenemalononitriles, diethyl arylidenemalonates, ethyl arylidenecyanoacetates, arylidenecyanoacetamides, 5-arylidene derivatives of barbituric and thiobarbituric acids, arylidene derivatives of Meldrum's acid and arylidene derivatives of dimedone. Also, it demonstrates the reactivity of these arylidene derivatives in heterocyclic synthesis with emphasis on the most recent findings. Some of these are the *α,β*-enones, *víz.* aurones and 3-arylidene-4-chromanones belong to the natural products. The others are synthetic substances which are convenient and important intermediates for the synthesis of a variety of useful and novel heterocyclic systems.

Keywords

Arylidenes, Synthesis, Reactions, Heterocycles

1. Introduction

The chemistry of different arylidene compounds has generated intensive scientific studies throughout the world. Especial interest has been focused on the synthesis and pharmacological activities of different arylidene compounds. They are versatile synthons so that a variety of novel heterocycles with good pharmaceutical profiles can be designed. Those arylidenes are usually prepared through Knoevenagel condensations of aldehydes with active methylene compounds, they are usually base [1], Lewis acid [2], or surfactant-catalyzed [3]; thus, create much wastes. Recently, there was an interest in so-called solvent-free [4] [5] Knoevenagel condensations on solid supports that were promoted by infrared [6] or microwave irradiation [7] [8]. Unfortunately, the latter techniques

require solvents for the extraction from the solid supports as well as for the preparation of the initial adsorbates, and do not yield pure products. Thus, further solvents are required for purifying the workup. Even catalyst-free Knoevenagel reactions in water could not reach quantitative yields [9]-[11]. This research deals with the various methods of preparation of arylideneacetophenones, arylidenecycloalkanones, 2-arylidene-1-indanones, 2-arylidene-1-tetralones, 2-arylidene-1-benzosuberones, aurones, 1-thioaurones, 3-arylidene-4-chromanones, 3-arylidene-1-thio-4-chromanones, 3-arylideneflavanones, 3-arylidene-1-thioflavanones, arylideneanilines, arylidenemalononitriles, diethyl arylidenemalonates, ethyl arylidenecyanoacetates, arylidenecyanoacetamides, 5-arylidene derivatives of barbituric and thiobarbituric acids, arylidene derivatives of Meldrum's acid and arylidene derivatives of dimedone as well as their utilization in heterocyclic synthesis.

2. Synthesis

2.1. Synthesis of Arylideneacetophenone Derivatives (Chalcones)

Different methods are available for the preparation of chalcones [12]-[14]. The most convenient method is the Claisen-Schimdt condensation of equimolar quantities of arylmethylketones with aryl aldehydes in the presence of aqueous alcoholic alkali [15]-[25]. Chalcones are used to synthesize several derivatives like cyanopyridines, pyrazolines, isoxazoles, and pyrimidines having different heterocyclic ring systems [26]-[29].

2.1.1. Various Condensing Agents Have Been Used in the Synthesis of Chalcones

1) Alkali

Alkalis are the most widely used condensing agents for the synthesis of chalcones. They are used as an aqueous alcoholic solution of suitable concentration *viz.* 30%, 40%, 50% and 70% [15]-[25].

2) Acids

Methanolic solution of dry hydrochloric acid gas at 0˚C was used for the synthesis of chalcones from aromatic ketones and aldehydes [26] [27]. In addition, concentrated sulfuric acid in acetic acid was used as a condensing agent in the synthesis of chalcones [28].

3) Other Condensing Agents

Raval and Shah [29] used phosphorous oxychloride as a condensing agent to synthesize chalcones. In addition, Szell and Sipos [30] condensed 2-hydroxy-5-nitroacetophenone with benzaldehyde using anhydrous $AlCl_3$.

Besides the above, other condensing agents have been used in the synthesis of chalcones; namely, amino acids [31], an aqueous solution of borax [32], perchloric acid [33], piperidine [34], boron trifloride [35], alkali metal alkoxides [36], magnesium *tert*-butoxide [37], and thionyl chloride [38].

In recent years, microwave assisted solid support solvent-free organic synthesis have attracted the attention as they offer several advantages such as simple procedure, fast reaction rate, mild reaction conditions, eco-friendly and improved yields as compared to conventional methods [39] [40].

2.1.2. Mechanism

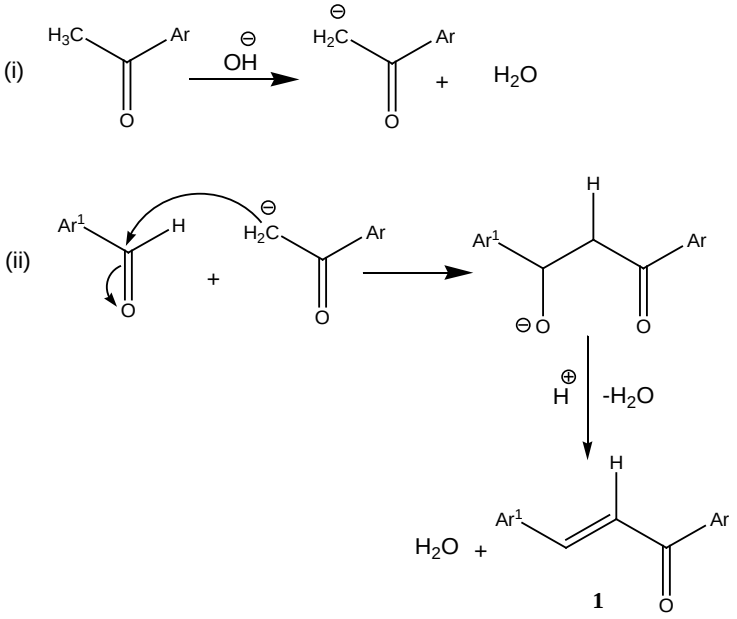
The following mechanisms have been suggested for the synthesis of chalcones:

1) Base catalyzed reaction (**Scheme 1**) [21] [41].

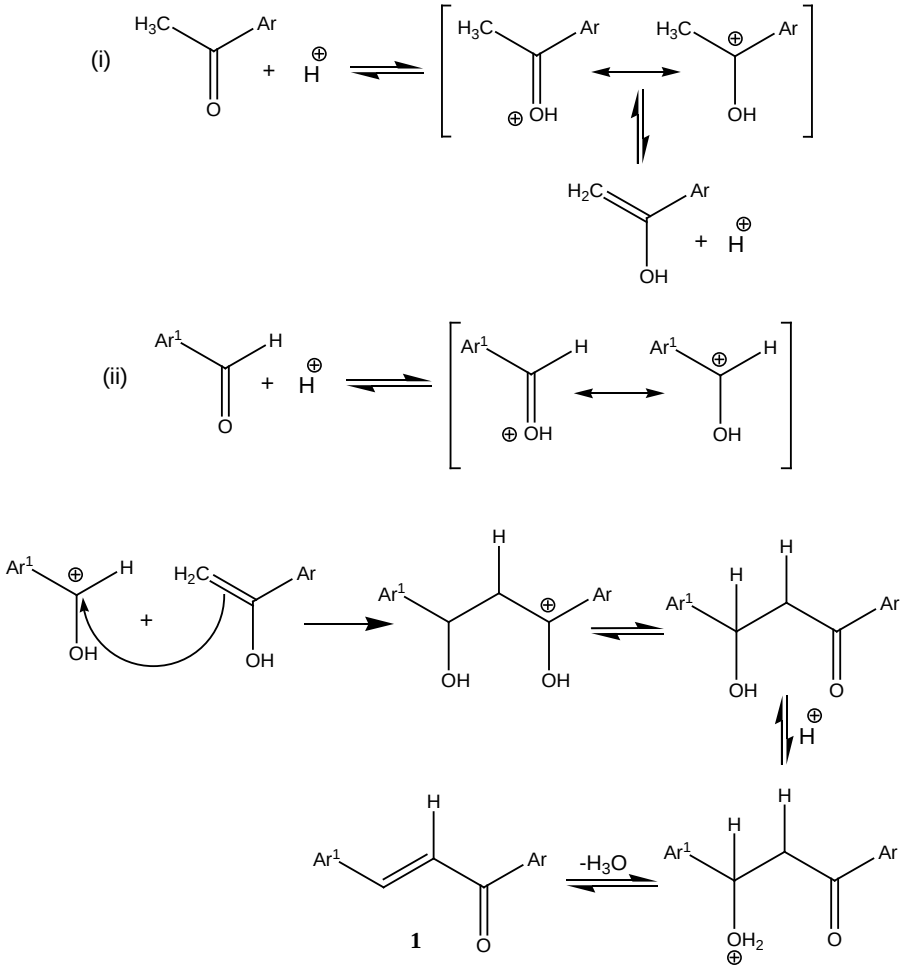
2) Acid catalyzed reaction (**Scheme 2**) [42].

2.1.3. Reactivity of Chalcone Derivatives

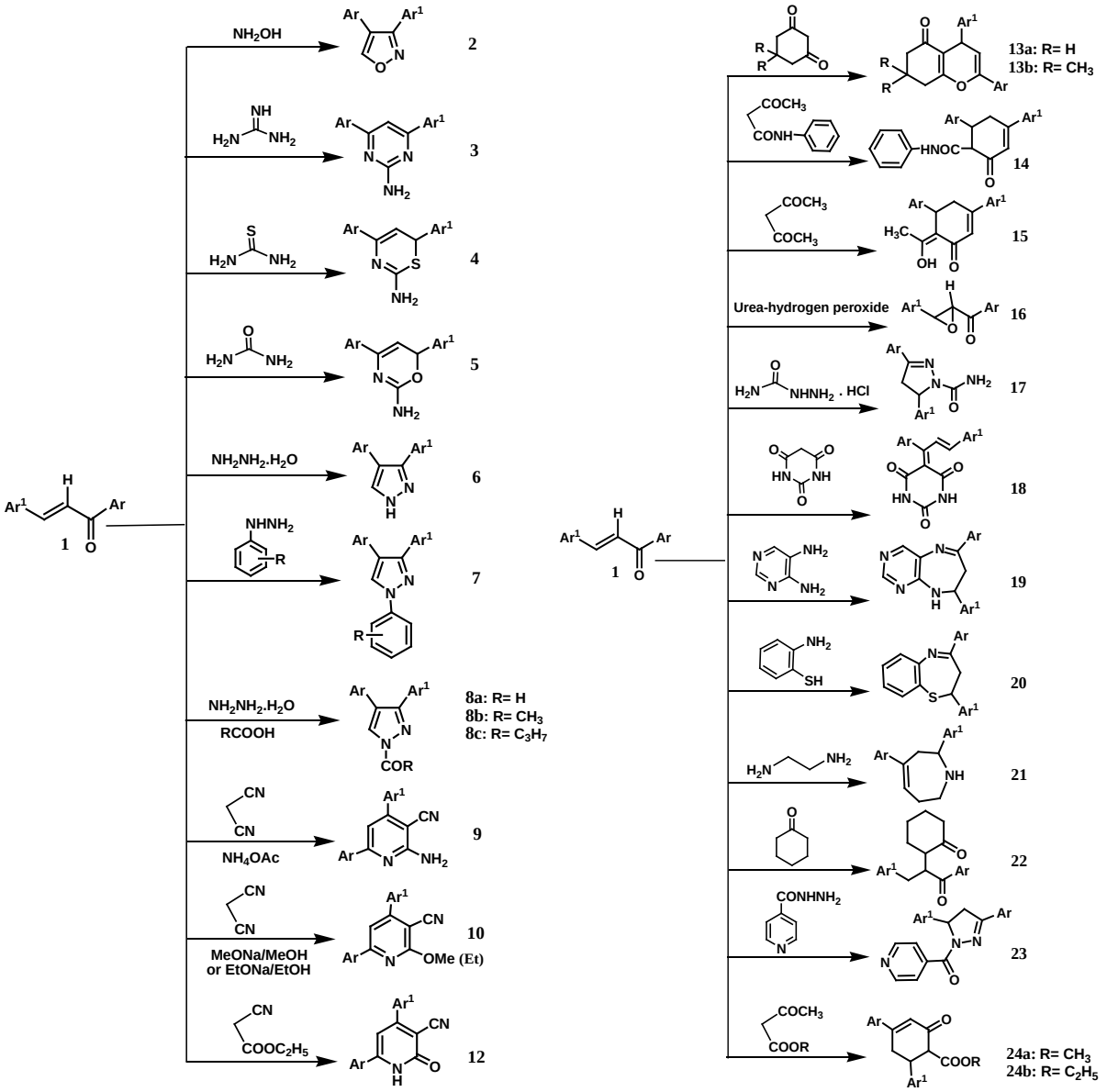
The chalcones **1** are useful intermediates for the synthesis of a variety of heterocyclic compounds. Isoxazoles **2** are prepared by the reaction of chalcones **1** with hydroxylamine hydrochloride and sodium acetate [43] (**Scheme 3**). Treatment of chalcones **1** with guanidine hydrochloride in the presence of alkali afforded 2-aminopyrimidines **3** [44] (**Scheme 3**). Thiazines **4** and oxazines **5** can be synthesized by reaction of chalcones **1** with thiourea and urea, respectively [45] (**Scheme 3**). Pyrazoles **6**, **7** are obtained through the reaction of chalcones **1** with hydrazine hydrate and phenyl hydrazine derivatives, respectively [46]-[49] (**Scheme 3**). Furthermore, reaction of hydrazine hydrate with **1** in the presence of different aliphatic acids resulted in the formation of pyrazole derivatives containing *N*-acyl moiety **8a-c** [49] [50] (**Scheme 3**). Condensation of chalcones **1** with malononitrile and ammonium acetate yields 2-amino-3-cyanopyridines **9** [49] [51] (**Scheme 3**). On the other hand, reaction of **1** with malononitrile in refluxing methanol or ethanol and in the presence of freshly prepared sodium alkoxide solution yielded 3-cyanopyridines **10** [52] (**Scheme 3**). Reaction of cyanopyridines **10** with hydrazine hydrate



Scheme 1. Mechanism of base catalyzed condensation of aromatic ketones with aldehydes.



Scheme 2. Mechanism of acid catalyzed condensation of aromatic ketones with aldehydes.



Scheme 3. Preparation of compounds **2-10** and **12-24**.

using Lewis acid (1.0 equivalent of $BF_3{\cdot}Et_2O$) in refluxing ethanol under anhydrous conditions afforded 1*H*-pyrazolo[3,4-*b*]pyridines **11** in very good yields and short reaction time [52] (**Scheme 4**). Similarly, treatment of **1** with ethyl cyanoacetate in absolute ethanol and in the presence of ammonium acetate afforded cyanopyridine derivatives **12** [49] (**Scheme 3**). Reaction of **1** with cyclohexane-1,3-diones produced 2,4-diaryl-5-oxo-5,6,7,8-tetrahydro-2-chromenes **13a**, **b**. [53] Similarly, acetoacetanilide and acetylacetone reacted with chalcones **1** to afford cyclohexenone derivatives **14** and **15**, respectively [49] (**Scheme 3**). Epoxidation of chalcones **1** with urea-hydrogen peroxide (UHP) under ultrasound irradiation gave oxirane derivatives **16** [54] (**Scheme 3**). Reaction of **1** with semicarbazide hydrochloride in glacial acetic acid/dioxane afforded pyrazoline-1-carboxamides **17** [55] (**Scheme 3**). The barbitones **18** are obtained upon condensation of chalcones **1** with barbituric acid [49] [56] [57] (**Scheme 3**). Treating **1** with 4,5-diaminopyrimidine gave [1,4]diazepine derivatives **19** [58] (**Scheme 3**). Condensation of chalcones **1** with 2-aminothiophenol afforded [1,4]benzothiazepines **20** [59] (**Scheme 3**). Tetrahydro-1*H*-azepines **21** were synthesized *via* reaction of **1** with ethylenediamine [60] (**Scheme 3**). Reaction of chalcones **1** with cyclohexanone in benzene in the presence of sodium hydroxide and a catalytic amount of

tributyl benzyl ammonium chloride (TBBAC) at room temperatue afforded 2-(1-oxo-1,3-diarylpropan-2-yl) cyclohexanones **22** [61] [62] (**Scheme 3**). Furthermore, reaction of **1** with isonicotinoyl hydrazide in refluxing ethanol gave *N*-isonicotinoyl-3,5-diarylpyrazolines **23** (**Scheme 3**) [49]. Reaction of **1** with methyl acetoacetate or ethyl acetoacetate in refluxing ethanol and in the presence of catalytic amounts of piperidine and basic alumina afforded 4,6-diaryl-2-oxocyclohex-3-enecarboxylates **24a**, **b** [49] [63] (**Scheme 3**). Condensation of cyclohexenecarboxylates **24a**, **b** with hydrazine hydrate produced 4,6-diaryl-3-oxo-2,3,4,5-tetrahydroindazoles **25** [49] (**Scheme 5**).

2.2. Synthesis of Arylidenecycloalkanone Derivatives

Arylidenecycloalkanones are frequently used *α*,*β*-unsaturated ketones. Their synthesis is based on the reaction of the appropriate cyclic ketone with aldehydes, through aldol condensation reaction. Several reports exist for their synthesis [64]-[69], involving the use of organic and inorganic bases, metal catalysts, different types of Friedel-Crafts catalysts and trichloro-1,3,5-triazine (TCT). A more convenient method used solid potassium hydroxide [70] [71] or sodium hydroxide [72] as a catalyst for the condensation of different aldehydes with cycloalkanones in ethanol and resulted in *α*,*α*'-*bis*(substituted benzylidene)cycloalkanones **26** and **27** in good yields. This method is economical and eco-friendly as neither any byproduct was formed nor any toxic material was used during the synthesis, and the reactions were carried out at ambient temperature. In addition, the same condensation was carried out in refluxing ethanol and in the presence of a catalytic amount of ammonium chloride to afford *α*,*α*'-*bis*(arylidene)cycloalkanones [73]. Moreover, a simple and efficient procedure for the synthesis of *α*,*α*'-bis (arylidene)cycloalkanones has been developed using *N*-bromosuccinimide (NBS) as a catalyst under mild reaction conditions [74]. Recently, a simple, improved and solvent-free synthesis of **26** and **27** was performed using activated barium hydroxide and grinding three to five minutes at room temperature [75].

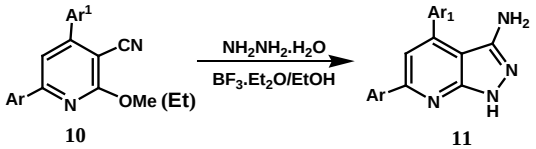
Reaction of cyclopentanone with substituted benzaldehydes (1:2 molar ratio) in alcoholic alkali solution produced *α*,*α*'-*bis*(substituted benzylidene)cyclopentanones **26** (**Scheme 6**) [76] [77].

Cyclohexanone was reacted with substituted benzaldehydes under alkaline reaction conditions (1:2 molar ratio) to afford *α*,*α*'-*bis*(substituted benzylidene)cyclohexanones **27** which could be easily separated (**Scheme 7**) [76]-[82]. The same condensation was carried out using amino-functionalized ionic liquid, 1-aminoethyl-3-methyl tetrafluoroborate ([2-aemim][BF_4]) as solvent and catalyst [83]. In addition, Brønsted acid-surfactant catalyst was utilized for synthesis of *α*,*ά*-bis(substituted benzylidene)cyclohexanones in aqueous media [84].

In addition, *α*,*α*'-bis(substituted benzylidene)cycloalkanones **26** and **27** could be obtained by refluxing 1,1-diacetates and cycloalkanones in tetrahydrofuran (THF) and in the presence of Samarium (III) Triiodide (SmI_3) as a catalyst (**Scheme 8**) [85].

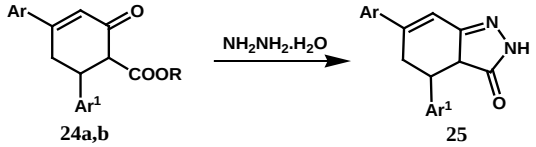
Cyclohexylphenyl methanols **28** were prepared by *D*-glucosamine catalyzed aldol reaction of cyclohexanone with substituted benzaldehydes (**Scheme 9**) [86].

3,5-Dibenzylidene-4*H*-pyran-4-ones (**30**, X = O) and 3,5-dibenzylidene-4*H*-1-thiopyran-4-ones (**30**, X = S) were synthesized *via* reaction of tetrahydro-4*H*-pyran-4-one (**29**, X = O) or tetrahydro-4*H*-1-thiopyran-4-one (**29**, X = S) with substituted benzaldehydes either under alkaline [87]-[90] or acidic [91] [92] reaction conditions (**Scheme 10**).



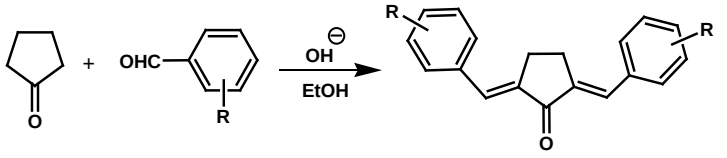
10 **11**

Scheme 4. Preparation of compounds **11**.



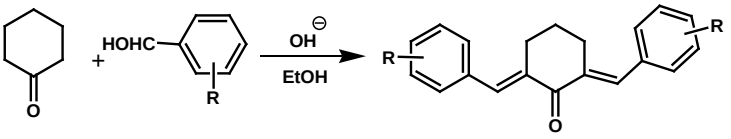
24a,b **25**

Scheme 5. Preparation of compounds **25**.



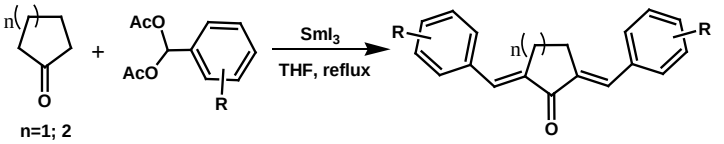
26

Scheme 6. Preparation of compounds **26** through reaction of cyclopentanone with substituted benzaldehydes under alkaline conditions.



27

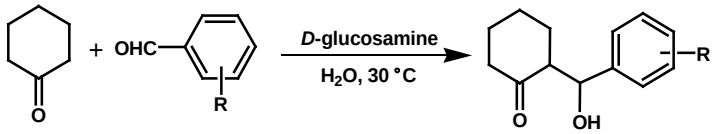
Scheme 7. Preparation of compounds **27** through reaction of cyclohexanone with substituted benzaldehydes under alkaline conditions.



n=1; 2

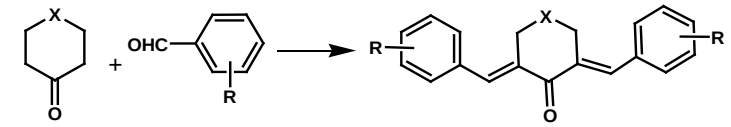
26: n=1
27: n=2

Scheme 8. Preparation of compounds **26** and **27** through reaction of 1,1-diacetates and cycloalkanones.



28

Scheme 9. Preparation of compounds **28**.



29: X = O; S

30: X = O; S

Scheme 10. Preparation of compounds **30**.

2.3. Synthesis of 2-Arylidene-1-Indanone Derivatives

2-Arylidene-1-indanones **32** are important intermediates for the synthesis of a wide variety of heterocyclic ring systems. For this reason, it is useful to have simple and convenient procedures for their preparation. Most of the utilized syntheses are based on the condensation of 1-indanones **31** with aldehydes in the presence of a catalyst to afford 2-arylidene-1-indanones **32** (**Scheme 11**).

In most cases sodium or potassium hydroxide is used as a catalyst [93]-[99] and 2-arylidene-1-indanones **32** are obtained in good yields. In addition, various inorganic acids, *viz.* sulfuric, phosphoric or hydrochloric acids were used as catalysts to prepare **32** [100]-[105]. It is worth mentioning that acetic anhydride was used to facilitate the condensation of indanones with substituted benzaldehydes [106]. Basavaiah and Reddy [107] have introduced a simple one-pot procedure for the preparation of 2-arylidene-1-indanones **32** starting from *tert*-butyl 3-aryl-3-hydroxy-2-methylenepropanoate **33**, which was allowed to react with a catalytic amount of concen-

trated sulfuric acid in benzene followed by reaction of the intermediates formed with trifluoroacetic anhydride (TFAA) in methylene chloride to afford **32** (**Scheme 12**).

Reactivity of 2-Arylidene-1-Indanone Derivatives

The synthesis of indeno[1,2-*c*]pyrazoles **34** was accomplished *via* reaction of 2-arylidene-1-indanones **32** with phenylsulfonylhydrazide in an inert solvent such as aromatic hydrocarbon and in the presence of a catalytic amount of acid [108] (**Scheme 13**). The [3 + 2] cycloaddition reactions of 2-arylideneindanones **32** with the arylnitrile oxides generated *in situ* from arylhydroxyaminoyl chlorides **35** and triethylamine led to the formation of the spiro derivatives **36** [109] (**Scheme 14**).

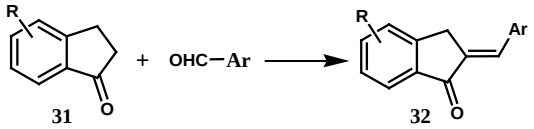
2.4. Synthesis of 2-Arylidene-1-Tetralone Derivatives

2-Arylidene-1-tetralones **38** are useful intermediates for the synthesis of polycyclic ring systems. Several synthetic methods have been developed for their preparation. The majority of compounds **38** have been synthesized by the condensation of 1-tetralones **37** with aromatic aldehydes in aqueous alcoholic solution of sodium or potassium hydroxide [94] [110]-[122]. Piperidine is another alkaline catalyst which has also been used to obtain 2-arylidene-1-tetralones **38** [108] [123]-[126]. In addition, acidic catalysts such as sulfuric, phosphoric and hydrochloric acids were utilized for this condensation [127]-[129] (**Scheme 15**).

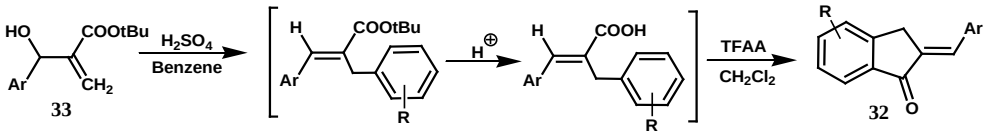
Reactivity of 2-Arylidene-1-Tetralone Derivatives

The naphtho[1,2-*c*]pyrazole derivatives **39** were prepared *via* reaction of 2-arylidene-1-tetralones **38** with phenylsulfonylhydrazide in an inert solvent such as aromatic hydrocarbon and in the presence of a catalytic amount of acid [108] (**Scheme 16**).

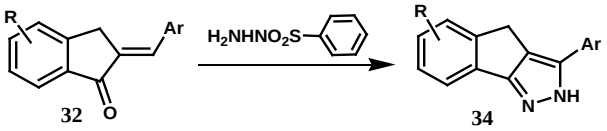
The reaction of **38** with the bicyclic carbonyl ylide **41** generated from the α-diazo ketone **40** in the presence of $Rh_2(OAc)_4$, afforded the spirodioxa ring systems **42** [130] (**Scheme 17**).



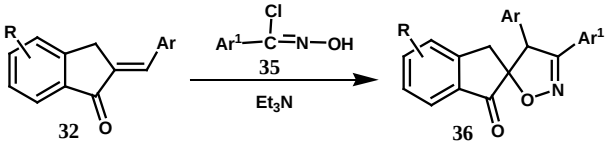
Scheme 11. Preparation of compounds **32** through condensation of 1-indanones **31** with aldehydes.



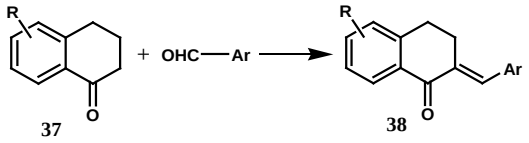
Scheme 12. Preparation of compounds **32** starting from *tert*-butyl 3-aryl-3-hydroxy-2-methylenepropanoate **33**.



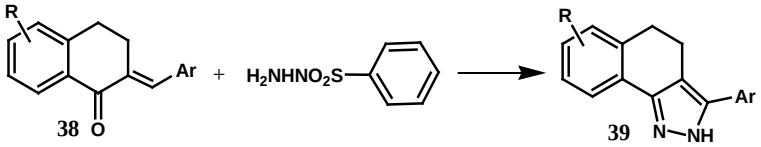
Scheme 13. Preparation of compounds **34**.



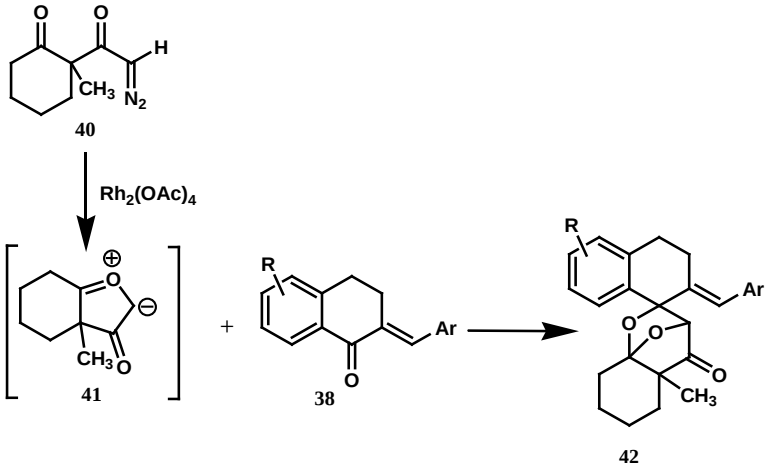
Scheme 14. Preparation of compounds **36**.



Scheme 15. Preparation of compounds **38**.



Scheme 16. Preparation of compounds **39**.



Scheme 17. Preparation of compounds **42**.

Moreover, the preparation of benzo[*g*]pyrazolo[3,4-*b*]quinolines **43** was accomplished *via* cyclocondensation reaction of **38** with aminopyrazoles under solvent-free conditions [131] (**Scheme 18**).

Treatment of **38** with potassium isothiocyanate gave 2-[aryl(isothiocyanato)methyl]-3,4-dihydronaphthalen-1(2*H*)-ones **44**. Reaction of **44** with primary aromatic amines gave 4-aryl-1-(substituted phenyl)-1,4,5,6-terahydrobenzo[*h*]quinazoline-2-thiols **45** [129] (**Scheme 19**).

Dispiropyrrolidinyl derivatives, 1',2',3',4'-tetrahydronaphthalen-1'-one-spiro[3'.3]-4-aryl-*N*-methylpyrrolidine-2-spiro-2"-acenaphthen-1"-ones **48** were obtained through reaction of 2-arylidene-1-tetralones **38**, acenaphthylenequinone **46** and sarcosine **47** in aqueous methanol [132] (**Scheme 20**).

Furthermore, 1',2',3',4'-tetrahydronaphthalen-1'-one-spiro[2'.3]-(4-aryl)pyrrolidine-spiro-[2.2"]oxindoles **50** were synthesized *via* reaction of 2-arylidene-1-tetralones **38**, isatin **(49)** and benzylamine in dry acetonitrile [132] (**Scheme 21**).

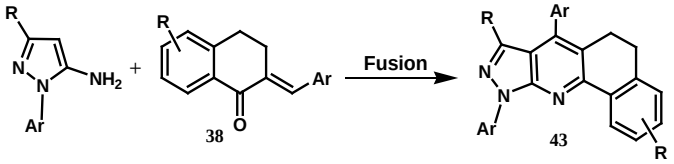
A new method to prepare benzo[*c*]xanthones **51** was reported by the ultraviolet radiation-mediated tandem reaction through irradiating a solution of 2-benzylidene-1-tetralones **38** in acetonitrile with ultraviolet light (500 W middle-pressure Hg) [133] (**Scheme 22**).

2.5. Synthesis of 2-Arylidene-1-Benzosuberone Derivatives

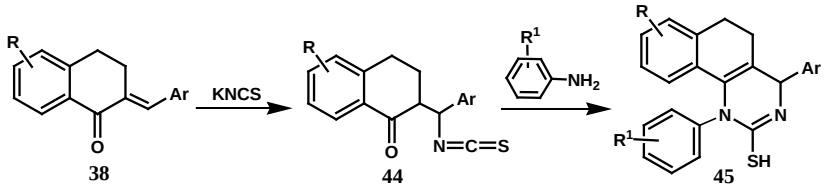
2-Arylidene-1-benzosuberones **53** were synthesized by the condensation of 1-benzosuberone **52** with aromatic aldehydes using alkaline [96] [121] [134] [135] or acidic [136] catalysts (**Scheme 23**).

2.6. Synthesis of Aurone Derivatives

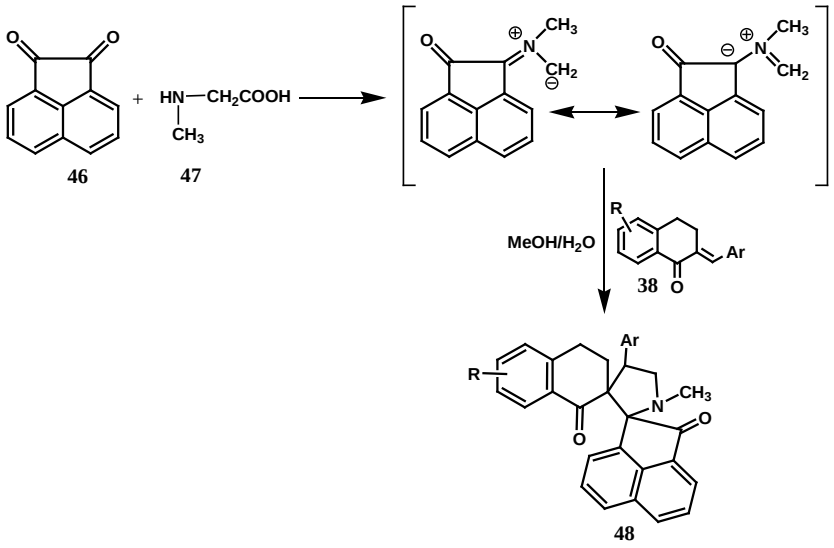
Aurones **55** are the oxa analogues of the 2-arylidene-1-indanones **32**, different procedures were adopted for their



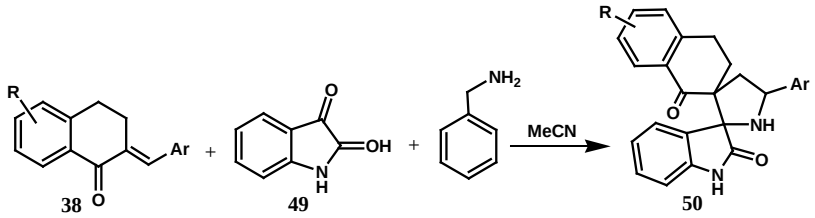
Scheme 18. Preparation of compounds **43**.



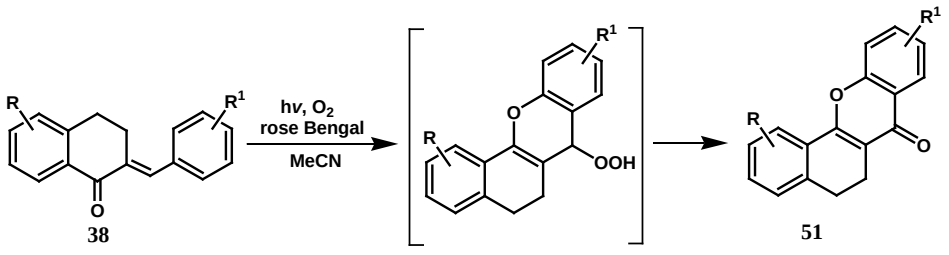
Scheme 19. Preparation of compounds **45**.



Scheme 20. Preparation of compounds **48**.



Scheme 21. Preparation of compounds **50**.



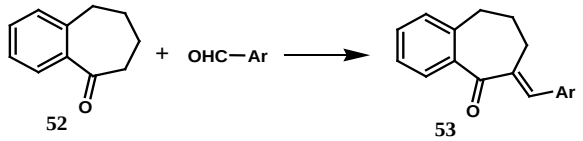
Scheme 22. Preparation of compounds **51**.

preparation. First, the Algar-Flynn-Oyamada reaction based on the oxidative cyclization of 2'-hydroxychalcones, where aurone is one of the products formed during preparation of 2'-hydroxychalcone [137]-[139]. Another procedure described by Donnelly and co-workers [140] [141] is based on bromomethylation of chalcones and 2'-acetoxychalcones followed by ring closure of the bromodihydro analogues providing aurones **55**. However, none of these procedures can be considered as a rational method for the synthesis of aurones.

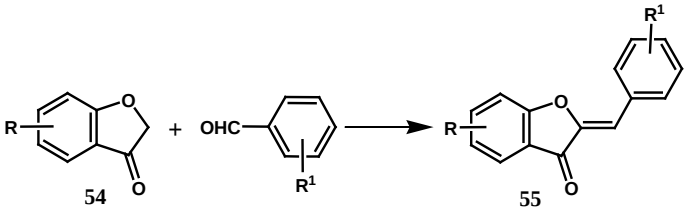
The most common synthetic procedures for aurones **55** are based on the condensation of coumaran-3-ones **54** with substituted benzaldehydes in the presence of a catalyst. As catalyst, sodium hydroxide [142] [143], potassium hydroxide [144], anhydrous sodium acetate [145], sulfuric [145], hydrochloric [146], and phosphoric acids [147] were used for this condensation (**Scheme 24**). Farkas *et al.* [148]-[150] performed the condensation of the appropriate coumaran-3-one **54** with substituted benzaldehydes in refluxing acetic anhydride to obtain aurones **55**.

Another synthetic procedure was developed for the preparation of aurones **55** through cyclization of 1-(2-hydroxyphenyl)-3-(substituted phenyl)prop-2-en-1-ones **56** in methanol and in the presence of a catalytic amount of silver nitrate [151] (**Scheme 25**).

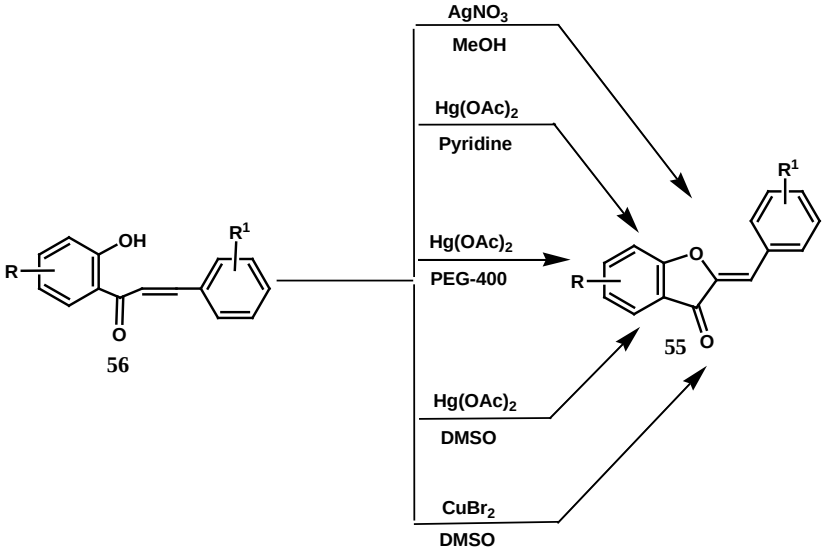
In addition, other synthetic procedures were adopted for the preparation of aurones **55** through cyclization of **56** using mercuric acetate in pyridine [152] [153], mercuric acetate in polyethylene glycol (PEG-400) [154], mercuric acetate in dimethyl sulfoxide (DMSO) [155], and cupric bromide in DMSO [152] (**Scheme 25**).



Scheme 23. Preparation of compounds **53**.



Scheme 24. Preparation of compounds **55** through condensation of coumaran-3-ones **54** with substituted benzaldehydes.



Scheme 25. Preparation of compounds **55** through cyclization of 1-(2-hydroxyphenyl)-3-(substituted phenyl)prop-2-en-1-ones **56**.

Mechanism of Cyclization Using Mercuric Acetate in Pyridine [152]

The mechanism of cyclization of **56** into aurone derivatives **55** is illustrated in **Scheme 26**.

Furthermore, gold-catalyzed cyclization of alkynol derivatives **57** has become an efficient tool in the synthesis of aurones **55** and provided the best results under mild reaction conditions and excellent selectivities, avoiding the formation of flavones as byproducts [156] [157] (**Scheme 27**).

2.7. Synthesis of 1-Thioaurone Derivatives

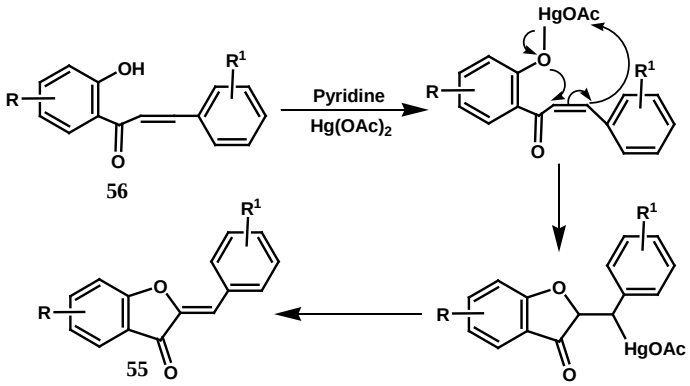
1-Thioaurones **59** are synthetic thio analogues of the naturally-occurring aurones, their synthesis has already been published [158]-[162]. Condensation of 1-thiocoumaran-3-ones **58** with aromatic aldehydes in the presence of phosphoric acid [147] or piperidine [159] afforded **59**. In addition, the same reaction was carried out in THF and in the presence of 1.5 equivalents of lithium diisopropylamide (LDA) at −10˚C [163] (**Scheme 28**).

Moreover, a convenient one-step synthesis has been published [161], whereas, equimolar amounts of (2-methylthio)benzoic acid derivatives **60** and aromatic aldehydes were allowed to react with 2.0 equivalents of LDA in THF at 0˚C to yield 1-thioaurones **59** (**Scheme 29**).

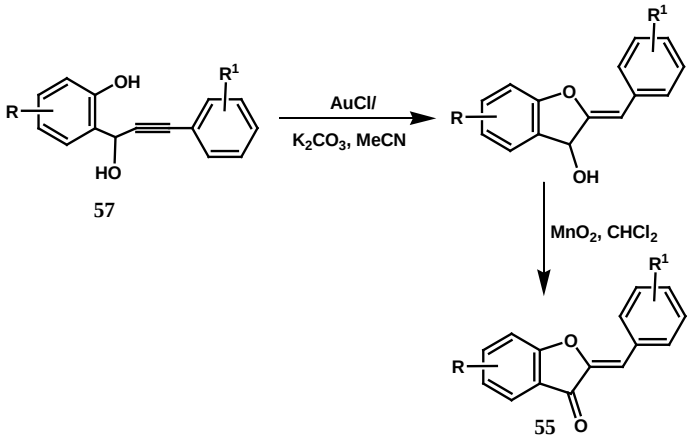
In 2010, Boughaleb *et al.* [162] described new synthetic pathway for the preparation of 1-thioaurones **59** (**Scheme 30**).

Reactivity of Aurone and 1-Thioaurone Derivatives

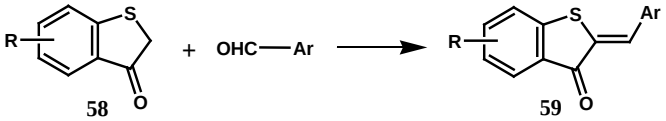
Reaction of **55** or **59** with 2-aminothiophenol in ethanol and in the presence of sodium ethoxide gave the spiro compounds **61a**, **b**. By the way of contrast, the 6,12-dihydrobenzofuro[2,3-*c*][1,5]benzothiazepines **62a** and the



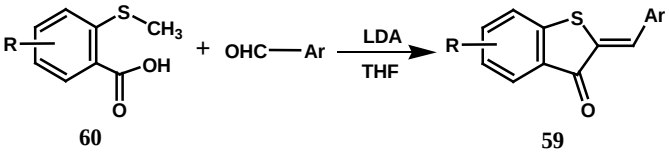
Scheme 26. Mechanism of cyclization of compounds **56** into aurone derivatives **55** using mercuric acetate in pyridine.



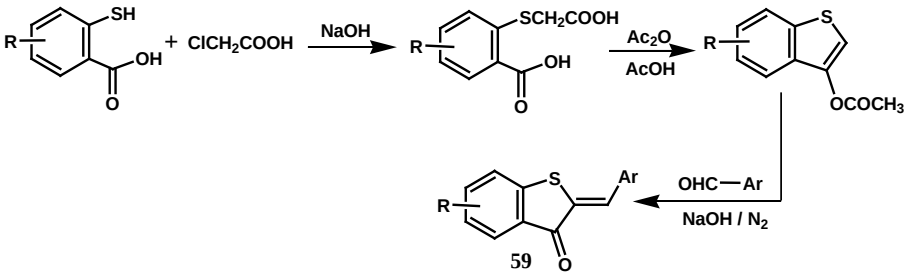
Scheme 27. Preparation of compounds **55** through cyclization of alkynol derivatives **57**.



Scheme 28. Preparation of compounds **59** through condensation of 1-thiocoumaran-3-ones **58** with aromatic aldehydes.



Scheme 29. Preparation of compounds **59** through reaction of (2-methylthio)benzoic acid derivatives **60** with aromatic aldehydes.



Scheme 30. Preparation of compounds **59** starting from 2-mercaptobenzoic acid derivatives.

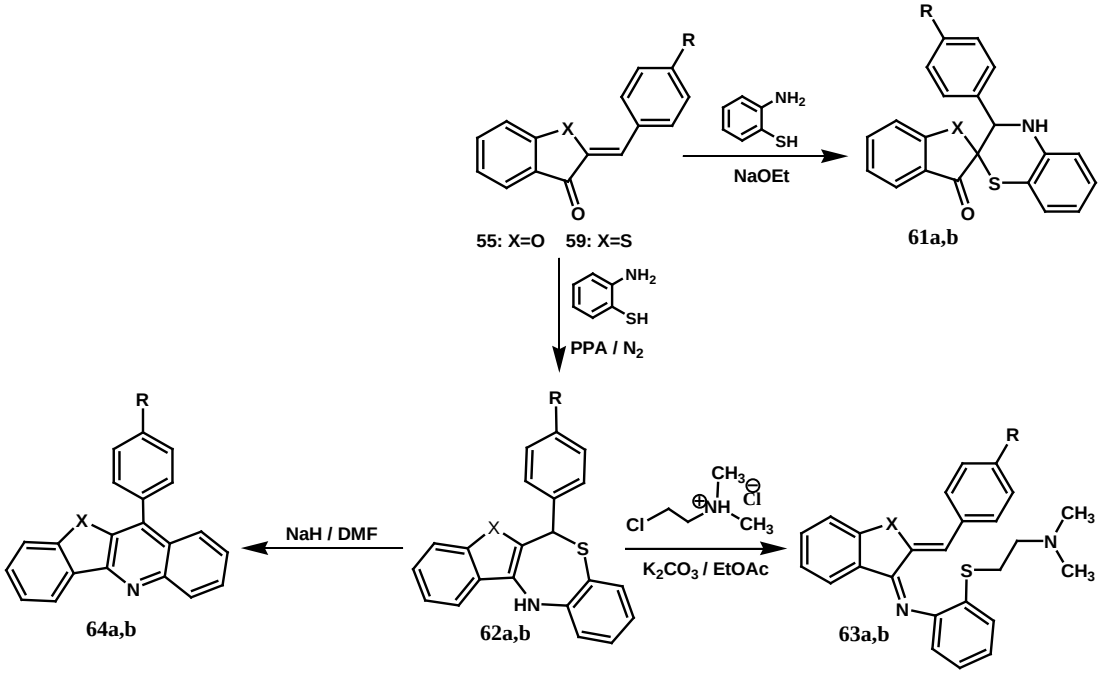
6,12-dihydrobenzothieno[2,3-*c*][1,5]benzothiazepines **62b** were obtained in good yields *via* heating **55** or **59**, respectively with 2-aminothiophenol in polyphosphoric acid (PPA) under nitrogen. Treatment of **62a**, **b** with 2-chloroethyl-*N*,*N*-dimethylammonium chloride and potassium carbonate in ethyl acetate produced the annulated benzofuran and benzothiophene derivatives **63a**, **b**. The tetracyclic derivatives **62a**, **b** were deprotonated with sodium hydride in DMF to afford compounds **64a**, **b** [164] (**Scheme 31**).

Reaction of aurones **55** with hydrazine hydrate in ethanol gave the benzofuro[3,2-*c*]pyrazole derivatives **65**. Refluxing aurones **55** with phenyl hydrazine in glacial acetic acid gave the benzofuro[3,2-*c*]pyrazole derivatives **66**. In addition, benzofuro[3,2-*c*]isoxazole derivatives **67** were synthesized by the reaction of aurones **55** with hydroxylamine hydrochloride in alcoholic solution of potassium hydroxide. Furthermore, benzofuro[2,3-*c*]pyridine derivatives **68** were obtained through reaction of aurones **55** with acetamide in alcoholic solution of potassium hydroxide. Finally, benzofuro[3,2-*d*]pyrimidine derivatives **69** were synthesized *via* reaction of aurones **55** with urea or thiourea in alcoholic solution of potassium hydroxide [165] (**Scheme 32**).

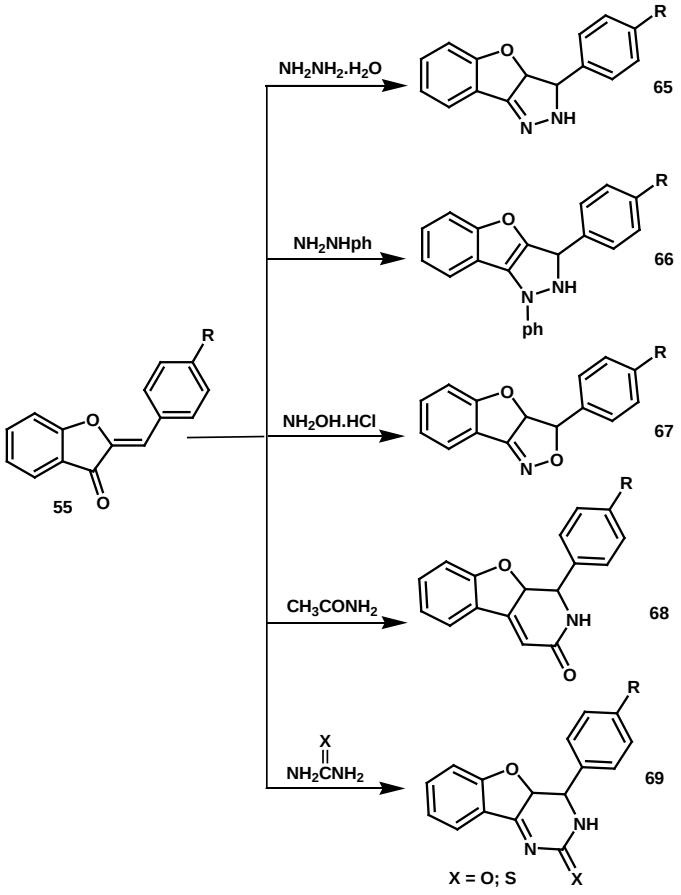
2.8. Synthesis of 3-Arylidene-4-Chromanone Derivatives

The synthesis and chemical transformation of 3-arylidene-4-chromanones and related compounds received much attention due to the abundance of this moiety in many natural products and biologically active substances [166]-[169]. Thiochromones are synthetic compounds and some of their derivatives are reported to have medicinal uses [170] [171]. Current literature showed that there has been an increasing trend towards the synthesis of heterocycles containing these two ring systems [172].

The synthesis of 3-arylidene-4-chromanones **71** is based on the condensation of 4-chromanones **70** with aromatic aldehydes in the presence of a catalyst (**Scheme 33**). Acid-catalyzed condensation (H_2SO_4, H_3PO_4 or HCl) of the two components was accomplished [173]-[180]. In addition, Farkas *et al.* [181]-[183] performed the same reaction in hot acetic anhydride, which is a very simple and convenient method, but sometimes it requires a prolonged time. Another procedure used for the synthesis of **71** is the base catalyzed condensation of 4-chromanones **70** with aromatic aldehydes using sodium hydroxide [184], sodium methoxide [185], anhydrous potassium acetate [186], piperidine [187]-[189], or pyrrolidine [190]. A new synthetic method for **71** was through



Scheme 31. Preparation of compounds **61-64**.



Scheme 32. Preparation of compounds **65-69**.

condensation of different aromatic aldehydes with 4-chromanones **70** using amberlyst-15 as a catalyst under microwave irradiation in solvent-free conditions [191]. However, it should be mentioned that in case of using piperidine as a catalyst, an exo-endo double bond migration takes place if the aldehyde has strong electron-withdrawing substituents [188] [192]. In such a case, 3-arylmethyl-4-chromenone (homoisoflavone) **72** is the product instead of the expected 3-arylidene-4-chromanone **71** (**Scheme 34**). Basavaiah *et al.* [107] [193] synthesized 3-arylidene-4-chromanones **71** by ring closure of the acrylic acid derivatives **73** with TFAA in methylene chloride (**Scheme 35**).

Reactivity of 3-Arylidene-4-Chromanone Derivatives

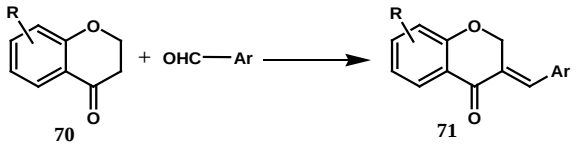
Refluxing a solution of 3-arylidene-4-chromanone **71**, isatin (**49**) and sarcosine (**74**) afforded 4-aryl-*N*-methyl-spiro[2.3'](2-oxoindoline)-spiro[3.3''](substituted 4-chromanone)pyrrolidines **75** [194] (**Scheme 36**). Whereas, refluxing a solution of **71**, isatin (**49**) and L-proline (**76**) in aqueous methanol gave 4-aryl-spiro[2.3'](2-oxoindoline)-spiro[3.3'']-(substituted 4-chromanone)hexahydropyrrolizines **77**. The reaction proceeded *via* formation of an azomethine ylide which readily undergoes 1,3-dipolar cycloaddition reaction with 3-arylidene-4-chromanones to give a single cycloadduct [194] (**Scheme 36**).

2.9. Synthesis of 3-Arylidene-1-Thio-4-Chromanone Derivatives

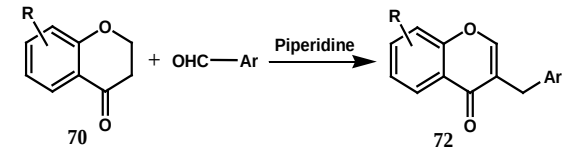
The synthesis of 3-arylidene-1-thio-4-chromanones **79** is based on the condensation of 1-thio-4-chromanones **78** with aromatic aldehydes under acidic conditions [195]-[199]. The same condensation was accomplished using piperidine as a catalyst [187]-[189], or amberlyst-15 under microwave irradiation [191] (**Scheme 37**). As described for the condensation of 4-chromanone **70** with aromatic aldehydes [187] [188], in case of aromatic aldehydes bearing strongly electron-withdrawing substituents, an exo-endo double bond transposition also takes place, resulting in the formation of 3-arylmethyl-1-thio-4-chromenones **80** instead of 3-arylidene-1-thio-4-chromanones **79** [188] (**Scheme 38**).

2.10. Synthesis of 3-Arylideneflavanone Derivatives

3-Arylideneflavanones (flavindognides) **82** are well known flavanone derivatives. They were first synthesized by Katschalowsky and von Kostanecki in 1904 [200]. They were also synthesized by the acid-catalyzed condensation

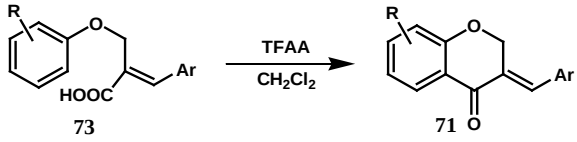


Scheme 33. Preparation of compounds **71** through condensation of 4-chromanones **70** with aromatic aldehydes.

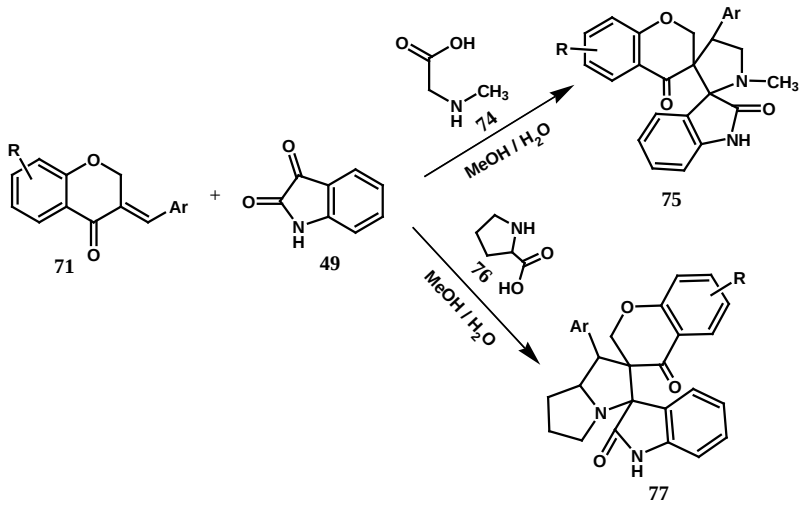


ArCHO = Aromatic aldehydes substituted with electron-withdrawing groups

Scheme 34. Preparation of compounds **72**.



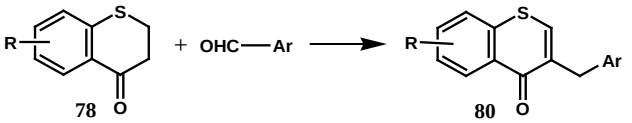
Scheme 35. Preparation of compounds **71** through cyclization of acrylic acid derivatives **73** with TFAA.



Scheme 36. Preparation of compounds **75** and **77**.

ArCHO = Aromatic aldehydes substituted with electron-donating groups

Scheme 37. Preparation of compounds **79**.



ArCHO = Aromatic aldehydes substituted with electron-withdrawing groups

Scheme 38. Preparation of compounds **80**.

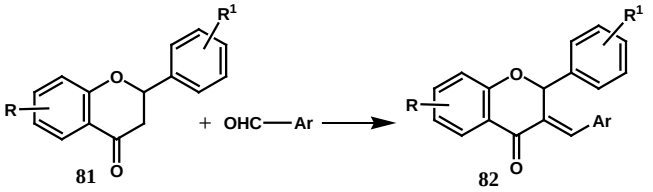
of flavanones **81** with aromatic aldehydes [200]-[204] (**Scheme 39**). In addition, glycine was described as a catalyst for this condensation [205]. It was reported that in some cases the base catalyzed condensation of hydroxyacetophenone with benzaldehyde gave 3-benzylideneflavanone as a coproduct of the corresponding hydroxychalcone [206]-[208]. Furthermore, the synthesis of **82** *via* base-catalyzed condensation of flavanone **81** with aromatic aldehydes was reported [189] [209]. It is worth mentioning that if aldehydes with strong electron-withdrawing substituents are used, 3-arylmethylflavones **83** are obtained instead of 3-arylideneflavanones **82** [210] (**Scheme 40**). 3-Arylmethylflavones **83** were also obtained *via* treatment of **82** with pyridinium chlorochromate (PCC) (5.0 equivalent) in DMF [211] (**Scheme 41**).

2.11. Synthesis of 3-Arylidene-1-Thioflavanone Derivatives

3-Arylidene-1-thioflavanones **85** were synthesized by the acid-catalyzed condensation of 1-thioflavanones **84** with aromatic aldehydes [212] [213] (**Scheme 42**). Also, base catalyzed condensation of thioflavanones **84** with aromatic aldehydes using piperidine was reported [214]. However, this procedure can be used only for the synthesis of 3-arylidene-1-thioflavanones substituted with electron-donating or slightly electron-withdrawing substituents in the arylidene moiety. When aromatic aldehydes substituted with strongly electron-withdrawing substituents were used, 3-arylmethyl-1-thioflavones **86** were obtained [214] [215] (**Scheme 43**).

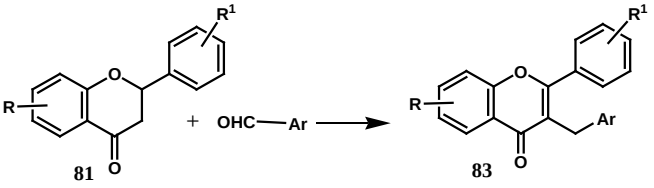
Reactivity of 3-Arylidene-1-Thioflavanone Derivatives

Reaction of 3-arylidene-1-thioflavanones **85** with sodium oxychloride and hydrogen peroxide gave 3-arylidene-



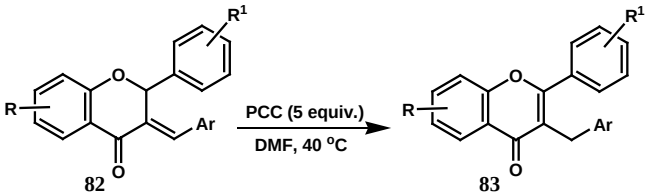
ArCHO = Aromatic aldehydes substituted with electron-donating groups

Scheme 39. Preparation of compounds **82** through condensation of flavanones **81** with aromatic aldehydes substituted with electron-donating groups.

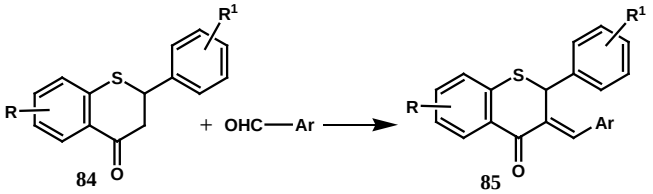


ArCHO = Aromatic aldehydes substituted with electron-withdrawing groups

Scheme 40. Preparation of compounds **83** through condensation of flavanones **81** with aromatic aldehydes substituted with strong electron-withdrawing groups.

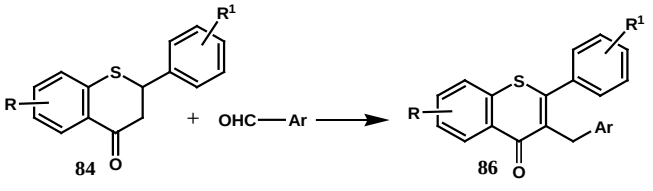


Scheme 41. Preparation of compounds **83** through treatment of 3-arylideneflavanones **82** with pyridinium chlorochromate (PCC).



ArCHO = Aromatic aldehydes substituted with electron-donating groups

Scheme 42. Preparation of compounds **85** through condensation of 1-thioflavanones **84** with aromatic aldehydes substituted with electron-donating groups.



ArCHO = Aromatic aldehydes substituted with electron-withdrawing groups

Scheme 43. Preparation of compounds **86** through condensation of 1-thioflavanones **84** with aromatic aldehydes substituted with strong electron-withdrawing groups.

1-thioflavanone epoxides **87** [216]. Reaction of epoxide derivatives **87** with dimethyldioxirane (DMD) yielded the sulfoxide and sulfone derivatives **88** and **89** [216] (**Scheme 44**).

2.12. Synthesis of Arylideneaniline Derivatives (Schiff Bases)

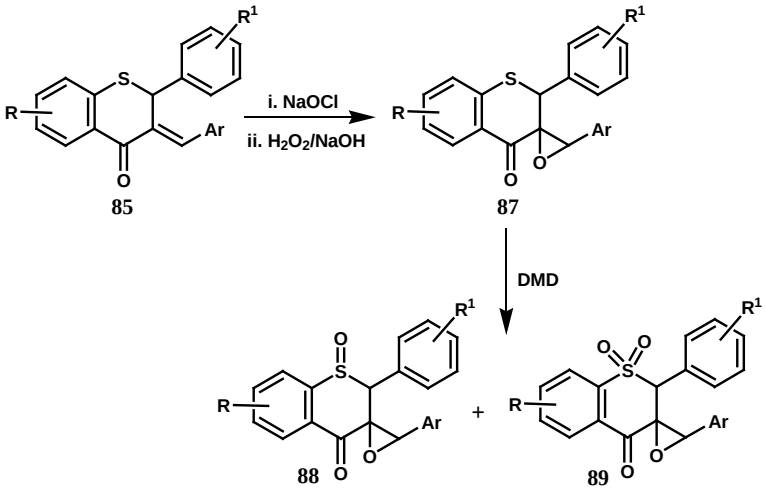
Schiff bases are typically formed by the condensation of primary amines with aldehydes. Schiff bases are important intermediates for the synthesis of various bioactive compounds. Literature survey revealed that these compounds have been associated with diverse chemotherapeutic activities, including antimalarial [217], anticancer [218], antibacterial [219], antifungal [220], antitubercular [221], anti-inflammatory [222], antimicrobial [222] and antiviral [223] activities. On the other hand, they are fundamental materials for the synthesis of various Schiff base ligands which are used as chiral auxiliaries in asymmetric synthesis [224]. Metal complex Schiff bases have also been used in oxidation reactions [225].

2.12.1. Various Reaction Conditions Have Been Used in the Synthesis of Schiff Bases

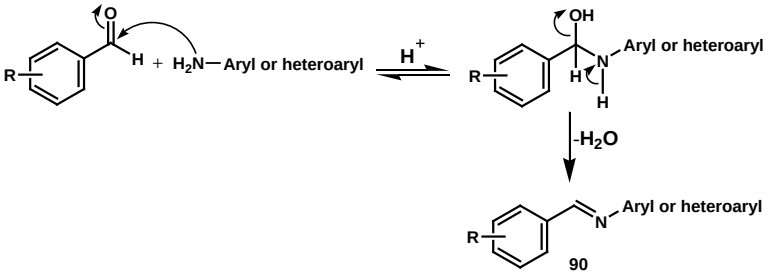
Schiff bases are compounds containing an azomethine group (-C=N-). They are usually formed by condensation of primary amines with carbonyl compounds according to the following equation [226]; R-NH_2 + R^1-CHO → R-N = CH-R^1 + H_2O, where R, R^1 may be an aliphatic or aromatic group. Schiff bases of aromatic aldehydes have an effective conjugated system and are more stable [227]. They are prepared under various reaction conditions, the use of organic solvents such as THF and 1,2-dichloroethane (DCE) was reported [228]. The reaction was also carried out in ethanol at room temperature [229] [230], in refluxing ethanol [231], in refluxing ethanol and in the presence of a catalytic amount of glacial acetic acid [222] [232], in refluxing methanol and in the presence of a catalytic amount of glacial acetic acid [233], in methanol at room temperature and in the presence of a catalytic amount of concentrated hydrochloric acid [234], in refluxing ethanol and in the presence of a catalytic amount of concentrated sulfuric acid [235], in refluxing mixture of ethanol/dioxane and in the presence of a catalytic amount of glacial acetic acid [236], in refluxing ethanol and in the presence of a catalytic amount of anhydrous zinc chloride [237], in refluxing benzene [238], in dichloromethane (DCM) at room temperature and in the presence of anhydrous magnesium sulfate [238], using DCM and a catalytic amount of neutral alumina under microwave irradiation [238], under solvent-free conditions in the presence of lemon juice as natural acid catalyst [239], in refluxing methanol and in the presence of a catalytic amount of nickel nitrate [240], and using phosphorus pentoxide/silica gel (P_2O_5/SiO_2) [241]. In addition, a green and efficient method for the synthesis of Schiff bases in aqueous media was described [242].

2.12.2. Mechanism

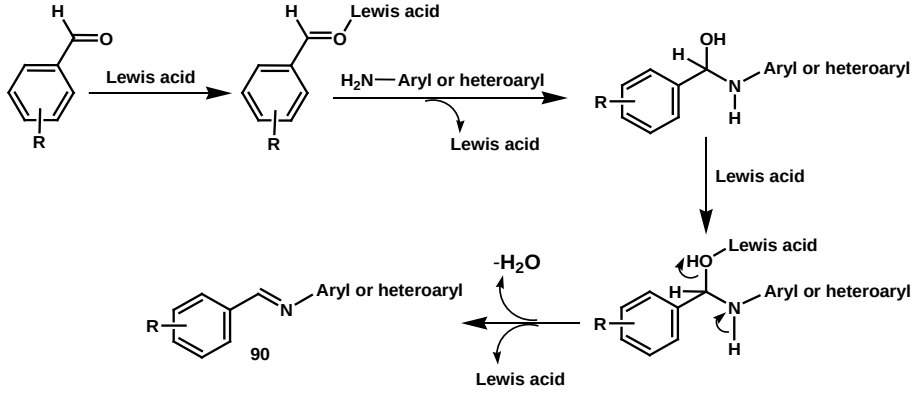
Concerning the mechanism of the transformation of aldehydes and amines into Schiff bases **90**, two possible pathways are illustrated (**Scheme 45** and **Scheme 46**) [239]. In **Scheme 45**, there is nucleophilic attack of a primary amine on carbonyl carbon that affords hydroxyl compound which on dehydration gives Schiff bases. The



Scheme 44. Preparation of compounds **87-89**.



Scheme 45. Mechanism of condensation of benzaldehyde derivatives with primary amines without using Lewis acid as a catalyst.



Scheme 46. Mechanism of condensation of benzaldehyde derivatives with primary amines using Lewis acid as a catalyst.

formation of Schiff bases **90** in this method largely depends on the rate of removal of water from the reaction mixture. Originally, the classical synthetic route for preparation of Schiff bases was reported by Schiff [243] which involves the condensation of primary amines with carbonyl compounds under azeotropic distillation [244] with the simultaneous removal of water. The removal of water during this condensation was conventionally facilitated by using molecular sieves [245] or a Dean-Stark apparatus [246]. In literature, the removal of water *in situ* has been accomplished by using dehydrating solvents such as tetramethyl orthosilicate [247] and trimethyl orthoformate [248] [249].

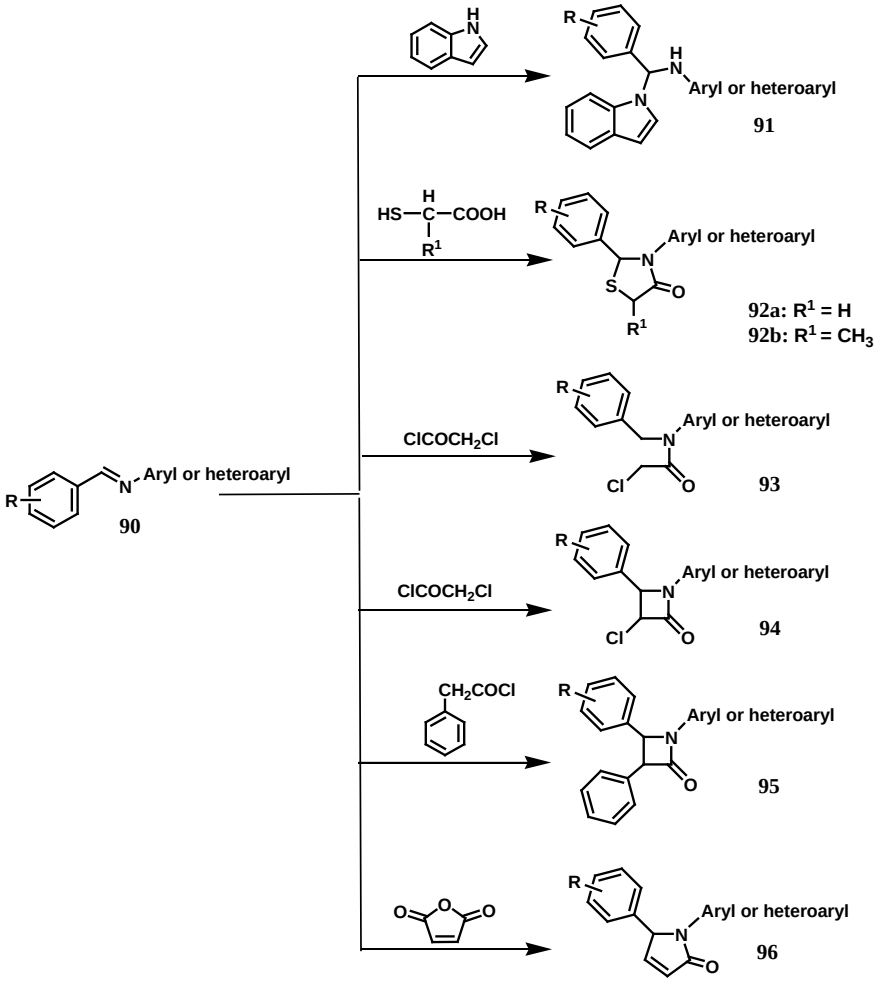
To overcome the difficulties in the removal of water, an alternative method has been employed in which Lewis acid is used as a catalyst that accelerates nucleophilic attack of amines on carbonyl carbon as well as serving as a dehydrating agent for removal of water in the second step (**Scheme 46**). Several modified methods for synthesis of Schiff bases have been reported in literature in which Lewis acids were used as catalysts such as $ZnCl_2$ [250], $TiCl_4$ [251], alumina [252], P_2O_5 [253] and also by using hydrotalcite [254].

2.12.3. Reactivity of Schiff Bases

Schiff bases are important intermediates for the synthesis of many heterocyclic compounds. Condensation of **90** with indole in basic medium afforded *N*-substituted indoles **91** [255]-[258]. The thiazolidin-4-one derivatives **92a,b** were obtained *via* reaction of **90** with thioglycolic [259]-[268] and thiolactic acids [269], respectively. Reaction of Schiff bases **90** with chloroacetyl chloride in dioxane and in the presence of triethylamine gave chloroacetamido derivatives **93** [266] or the azetidin-2-ones **94** [265] [270]. Similarly, reaction of **91** with phenylacetyl chloride in dioxane and in the presence of triethylamine produced the azetidinones **95** [271]. The pyrrol-2-one derivatives **96** can be synthesized by reaction of **90** with maleic anhydride [272]-[275] (**Scheme 47**).

2.13. Synthesis of Arylidenemalononitrile Derivatives

The Knoevenagel reaction is the most simple and straightforward method used to produce the substituted alkenes [276]. Classically, the process consists of condensation of aldehydes or ketones with active methylene



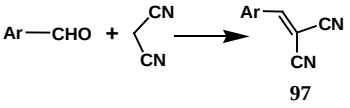
Scheme 47. Preparation of compounds **91-96**.

compounds in the presence of a variety of reagents. Several bases [276]-[279], Lewis acids [280] [281] or heterogeneous media [282]-[289] were used as catalysts in Knoevenagel condensation reaction for the synthesis of arylidenemalononitriles **97**. In addition, a simple and more efficient procedure was developed for the condensation of malononitrile with aromatic, heteroaromatic and aliphatic aldehydes in water [290] (**Scheme 48**).

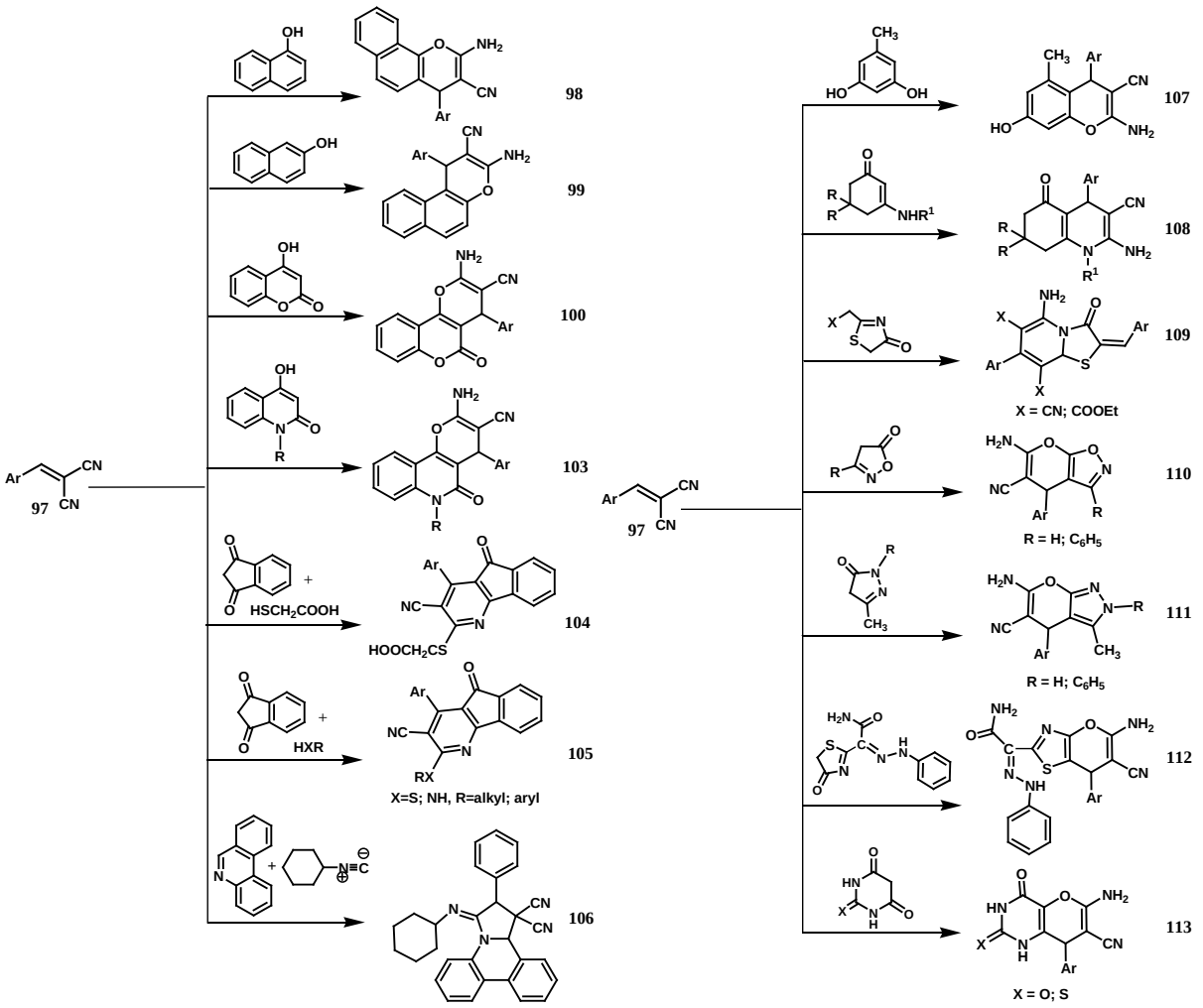
Reactivity of Arylidenemalononitrile Derivatives

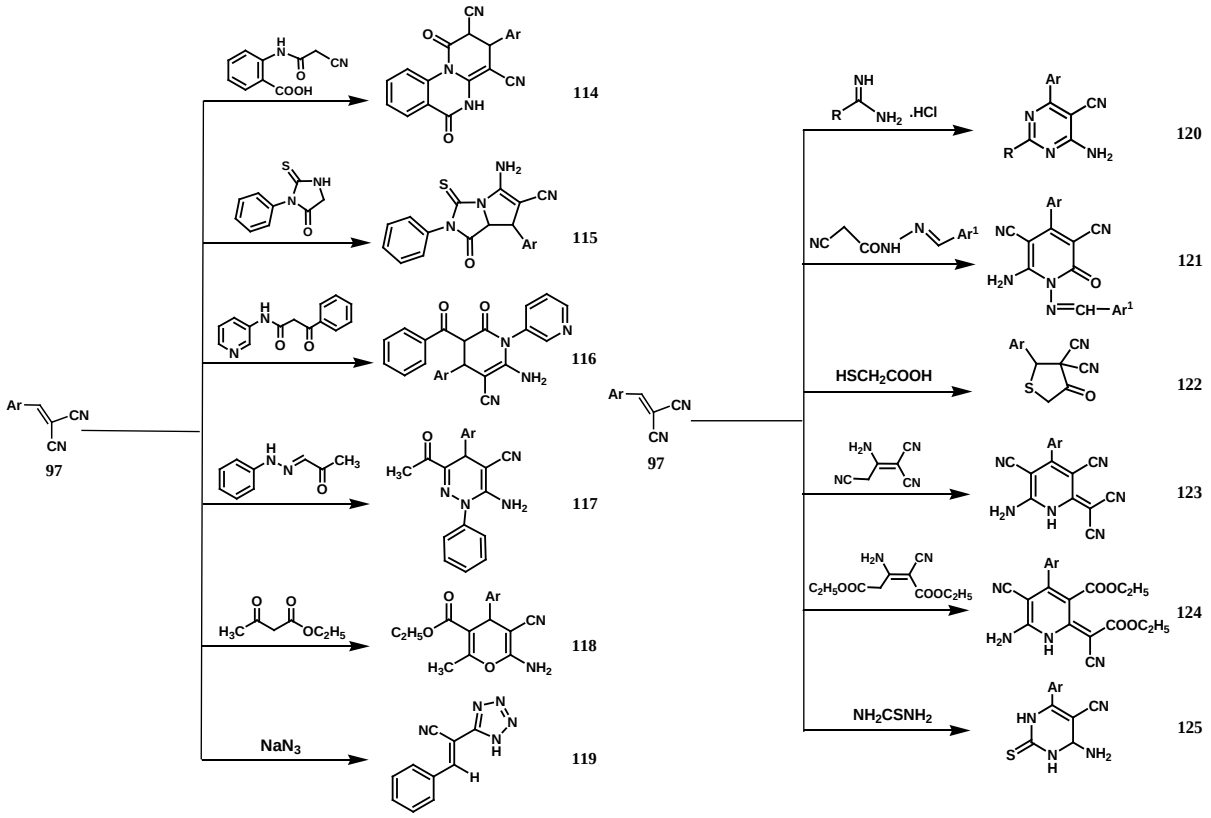
The arylidenemalononitriles **97** are useful intermediates for the synthesis of a variety of heterocyclic compounds, 2-aminobenzo[*h*]chromene-3-carbonitrile derivatives **98** were synthesized *via* condensation of **97** with *α*-naphthol in the presence of Mg/Al hydrotalcite under single-mode microwave irradiation [291] (**Scheme 49**). On the other hand, 3-aminobenzo[*f*]chromene-2-carbonitrile derivatives **99** were obtained through condensation of **97** with *β*-naphthol [292] (**Scheme 49**). Pyrano[3,2-*c*]chromenes **100** were prepared by heating 4-hydroxycoumarin with **97** in pyridine [293], in ethanol and in the presence of triethylamine as a catalyst [294] or in water [295] (**Scheme 49**). Acid hydrolysis of pyrano[3,2-*c*]chromenes **100** produced compound **101**, which is subsequently transformed into warfarin **102** (Ar = phenyl) [293] [296]-[298] (**Scheme 50**). Several pyrano[3,2-*c*]quinolines **103** were prepared *via* reaction of 4-hydroxyquinolin-2(1*H*)-ones with arylidenemalononitriles **97** [299] [300] (**Scheme 49**). Three component reaction between arylidenemalononitriles **97**, 1,3-indanedione and thioglycolic acid under microwave irradiation afforded indeno[1,2-*b*]pyridine derivatives **104** [301] (**Scheme 49**). The analogous three-component reaction of **97** with 1,3-indanedione and 4-methylbenzenethiol, or aromatic amine under microwave irradiation afforded indeno[1,2-*b*]pyridines **105** [301] (**Scheme 49**). A series of dihydropyrrolo[1,2-*f*]phenanthridines **106** was prepared *via* reaction between **97**, isocyanides and phenanthridine in dry diethyl ether [302]

(**Scheme 49**). Condensation of **97** with 5-methylresorcinol monohydrate in ethanol afforded the chromene derivatives **107** [303] (**Scheme 49**). Hexahydroquinolin-5-ones **108** were synthesized through reaction of 3-aminocyclohex-2-en-1-ones with **97** [304] (**Scheme 49**). The thiazolopyridine derivatives **109** were obtained using 2:1 molar ratio of **97** and 2-(4,5-dihydro-4-oxothiazol-2-yl)acetonitrile or ethyl 2-(4,5-dihydro-4-oxothiazol-2-yl)acetate [305] [306] (**Scheme 49**). Reaction of 2-isoxazolin-5-ones with **97** was reported to yield pyrazolo[2,3-*c*]isoxazole derivatives **110** [307] (**Scheme 49**). Condensation of **97** with 3-methylpyrazol-5-ones gave the pyranopyrazole derivatives **111** [292] (**Scheme 49**). Whereas, pyrano[2,3-*d*]thiazole derivatives **112** were prepared through reaction of **97** with *α*-(4-oxothiazolin-2-yl)-*α*-phenylhydrazonoacetamide [308] (**Scheme 49**). Condensation of **97** with barbituric or thiobarbituric acid afforded the corresponding pyrano[2,3-*d*]pyrimidine derivatives **113** [308] (**Scheme 49**). In addition, the pyrido[1,2-*a*]quinazoline derivatives **114** were synthesized *via* condensation of **97** with 2-(2-cyanoacetamido)benzoic acid [309] (**Scheme 49**). It was found that 3-phenyl-2-thiohydantoin reacts with **97** to give the corresponding pyrrolo[1,2-*c*]imidazole derivatives **115** [310] (**Scheme 49**). Reaction of 3-oxo-3-phenyl-*N*-(pyridin-3-yl)propanamide with **97** in ethanol gave tetrahydropyridines **116** [311] (**Scheme 49**). In addition, 6-acetyl-3-amino-2,5-diphenyl-2,3,4,5-tetrahydropyridazine-4-carbonitrile derivatives **117** were obtained through reaction of **97** with 1-(phenylhydrazono)propan-2-one in pyridine [312]



Scheme 48. Preparation of compounds **97**.





Scheme 49. Preparation of compounds **98-100** and **103-125**.

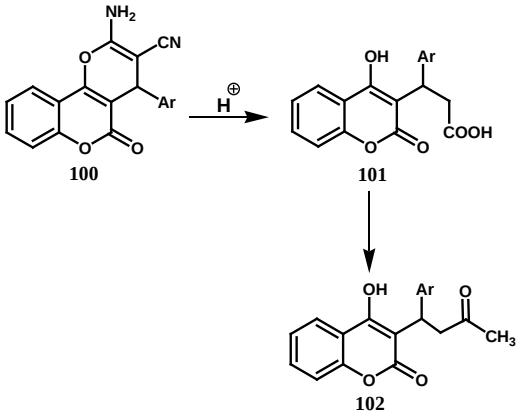
(**Scheme 49**). Reaction of **97**, ethyl acetoacetate and aqueous solution of ammonium hydroxide afforded 4*H*-pyran derivatives **118** [313] (**Scheme 49**). Tetrazoles **119** have been prepared through reaction of **97** with sodium azide in water [314] (**Scheme 49**). Micheal additions of arylidenemalononitriles **97** with amidines in refluxing acetonitrile and in the presence of a catalytic amount of magnesium oxide produced the pyrimidine derivatives **120** [289] (**Scheme 49**). Reaction of **97** with hydrazide-hydrazone derivatives in dioxane and in the presence of triethylamine as a catalyst gave 6-amino-2-oxopyridine-3,5-dicarbonitriles **121** [315] (**Scheme 49**). The synthesis of thiophene derivatives **122** was accomplished through the condensation of **97** with thioglycolic acid [316] [317] (**Scheme 49**). 2-Amino-1,1,3-tricyanoprop-2-ene reacts with **97** to yield the pyridine derivatives **123** [318] [319] (**Scheme 49**). Similary, diethyl 3-amino-2-cyanopent-2-ene-1,5-dicarboxylate was reacted with **97** to yield the pyridine derivatives **124** [319] [320] (**Scheme 49**). Treatment of **97** with thiourea in DMF and in the presence of a catalytic amount of piperidine resulted in the formation of the thioxopyrimidine derivatives **125** [321] (**Scheme 49**).

Reaction of arylidenemalononitriles **97** with 2-cyanothioacetamide in ethanol and in the presence of piperidine as a basic catalyst afforded 1,6-dihydro-6-thioxopyridine-2,3,5-tricarbonitriles **126**. Reaction of thioxopyridines **126** with (2-acetoxyethoxy)methyl bromide in DMF and in the presence of sodium hydride afforded 6-[(2-acetoxyethoxy)methylthio]pyridine-2,3,5-tricarbonitriles **127** that is further reacted with ammonia in methanol to produce 6-[(2-hydroxyethoxy)methylthio]pyridine-2,3,5-tricarbonitriles **128**. Reaction of **126** with ethoxymethyl chloride in DMF and in the presence of sodium hydride gave 6-(ethoxymethylthio)pyridine-2,3,5-tricarbonitriles **129** [322] (**Scheme 51**).

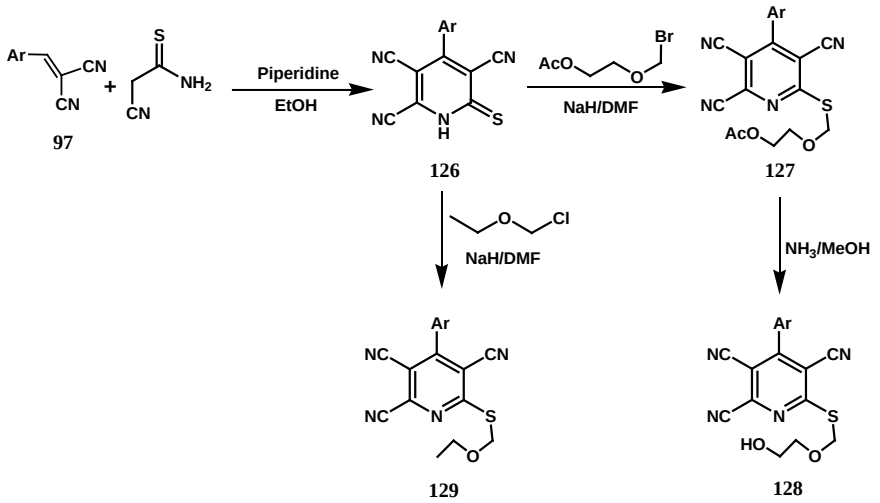
It was reported that thermal Michael addition reaction takes place when 6,7-dimethoxyisochromanone **130** was treated with arylidenemalononitriles **97** at 190˚C to afford **131** which underwent elimination of malononitrile producing **132** [323] (**Scheme 52**).

2.14. Synthesis of Diethyl Arylidenemalonate Derivatives

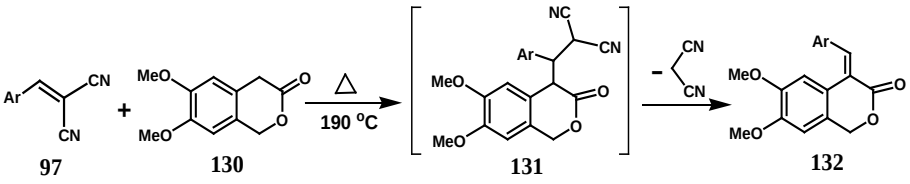
Diethyl arylidenemalonates **133** are easily accessible by Knoevenagel condensation [324] [325] (**Scheme 53**).



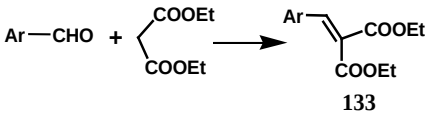
Scheme 50. Preparation of compounds **101** and **102**.



Scheme 51. Preparation of compounds **126-129**.



Scheme 52. Preparation of compounds **132**.



Scheme 53. Preparation of compounds **133**.

Various reaction conditions were reported for the preparation of diethyl arylidenemalonates **133**. The reaction was performed in water or ethanol without catalyst [326], in refluxing dry xylene and in the presence of a catalytic amount of piperidine/glacial acetic acid (3:1) [327], in refluxing ethanol and in the presence of a catalytic amount of piperidine/glacial acetic acid (2:1) [328] or in refluxing pyridine [329]. In addition, Knoevenagel condensations of benzaldehyde or substituted benzaldehydes with diethyl malonate was carried out in Lewis acidic 1-butyl-3-methylimidazolium chloroaluminate, [bmim]Cl·x$AlCl_3$ and 1-butylpyridinium chloroaluminate,

[bpy]Cl·xAlCl$_3$ ionic liquids [330].

Reactivity of Diethyl Arylidenemalonate Derivatives

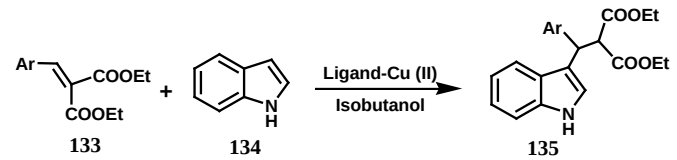
Reaction of diethyl arylidenemalonates **133** with indole **134** in isobutanol at room temperature, employing Cu(OTf)$_2$-bis(oxazoline) complexes under nitrogen afforded ethyl 3-aryl-2-ethoxycarbonyl-3-(3-indolyl) propanoates **135** [331] (**Scheme 54**). Reaction of diethyl 4-methoxybenzylidenemalonate **136** with but-2-yne-1,4-diol **137** in the presence of sodium hydride (NaH) in THF at room temperature for five minutes afforded ethyl 3-(4-methoxyphenyl)-4-oxo-3,3a,4,6-tetrahydro-1*H*-furo[3,4-*c*]pyran-3a-carboxylate **138** [332]-[335] (**Scheme 55**).

2.15. Synthesis of Ethyl Arylidenecyanoacetate Derivatives

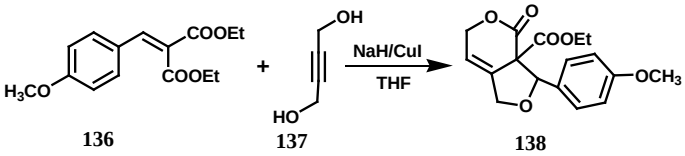
Ethyl arylidenecyanoacetates **139** are prepared *via* Knoevenagel condensation of aldehydes with ethyl cyanoacetate (**Scheme 56**). Several publications were reported for the synthesis of ethyl arylidenecyanoacetates. The reaction was carried out in aqueous medium at room temperature [336], in refluxing ethanol and in the presence of piperidine/glacial acetic acid (2:1) [328], in refluxing ethanol and in the presence of Trizma [337]. Also, this condensation was performed in ethanol/water mixture and in the presence of sodium or potassium hydroxide at 50˚C - 60˚C [279]. The same reaction was carried out using magnesium bromide diethyl etherate (MgBr$_2$·OEt$_2$) as Lewis acid in the presence of triethylamine [338]. In addition, poly(4-methyl vinylpyridinium hydroxide)/SBA-15, a novel basic polymeric composite was applied as a recyclable catalyst for the Knoevenagel condensation reaction of aromatic aldehydes with ethyl cyanoacetate in water at 95˚C [339]. Furthermore, they were prepared *via* heating a mixture of aldehyde and ethyl cyanoacetate at 80˚C - 85˚C in an oil bath and in the presence of rare earth triflates as Yb(OTf)$_3$ [340]. The same reaction was performed in distilled water and in the presence of hydroxyapatite supported caesium carbonate as a recyclable solid base catalyst (HAP-Cs$_2$CO$_3$) [278] or under microwave irradiation [341].

Reactivity of Ethyl Arylidenecyanoacetate Derivatives

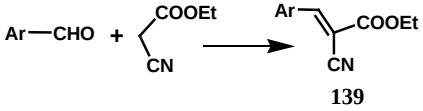
The thiazolopyridine derivatives **109** were also obtained using 2:1 molar ratio of ethyl arylidenecyanoacetates **139** and 2-(4,5-dihydro-4-oxothiazol-2-yl)acetonitrile or ethyl 2-(4,5-dihydro-4-oxothiazol-2-yl)acetate [305] [306] (**Scheme 57**). Reaction of ethyl arylidenecyanoacetates **139** with hydrazide-hydrazone derivatives in dioxane and in the presence of a catalytic amount of triethylamine produced ethyl 2-amino-5-cyano-6-oxopyridine-3-carboxylates **140** [298] (**Scheme 57**). The synthesis of thiophene derivatives **141** was accomplished through the condensation of **139** with thioglycolic acid [316] [317] (**Scheme 57**). Reaction of **139** with 2-cyanoacetic



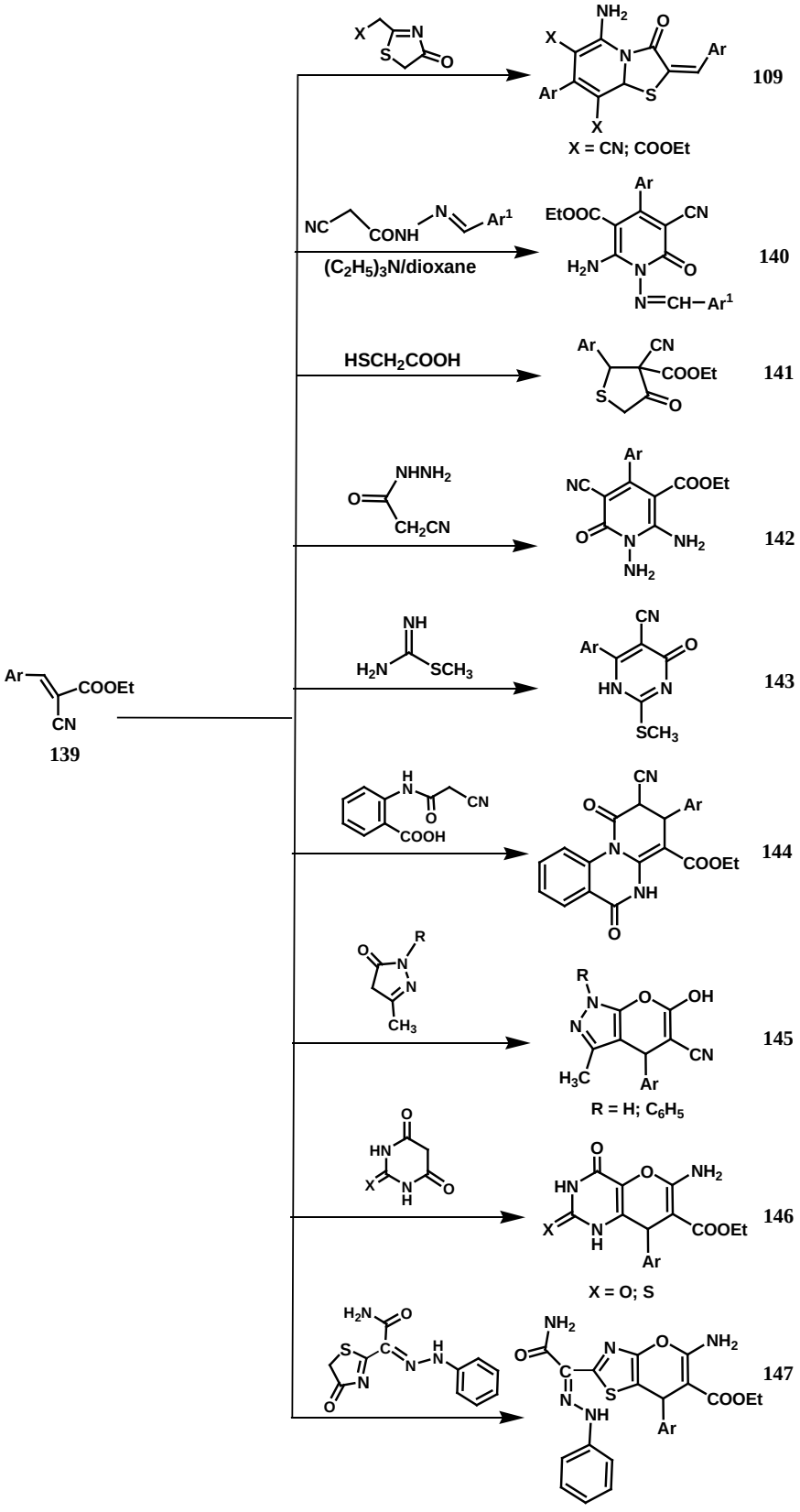
Scheme 54. Preparation of compounds **135**.



Scheme 55. Preparation of compounds **138**.



Scheme 56. Preparation of compounds **139**.



Scheme 57. Preparation of compounds **109** and **140-147**.

acid hydrazide gave the pyridine derivatives **142** [342] (**Scheme 57**). The reaction of **139** with *S*-methylthiourea in pyridine produced the pyrimidine derivatives **143** [343]. In addition, the pyrido[1,2-*a*]quinazoline derivatives **144** were synthesized *via* condensation of **139** with 2-(2-cyanoacetamido)benzoic acid [309] (**Scheme 57**). Moreover, the pyrano[2,3-*c*]pyrazole derivatives **145** were obtained through condensation of **139** with 3-methylpyrazolone derivatives [344] [345] (**Scheme 57**). Condensation of **139** with barbituric or thiobarbituric acids afforded the corresponding pyrano[3,2-*d*]pyrimidine derivatives **146** [308] (**Scheme 57**). Whereas, pyrano[2,3-*d*]thiazole derivatives **147** were prepared through reaction of **139** with *α*-(4-oxothiazolin-2-yl)-*α*-phenylhydrazonoacetamide [308] (**Scheme 57**).

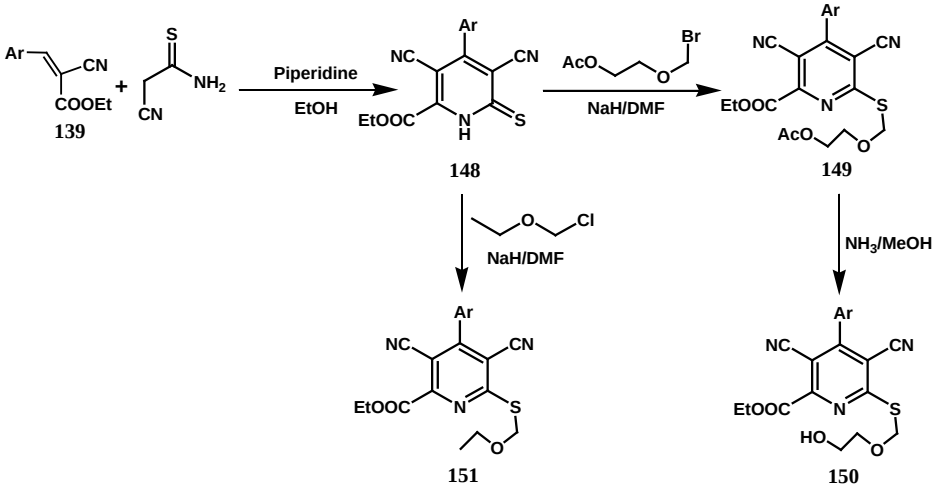
Reaction of **139** with 2-cyanothioacetamide in ethanol and in the presence of a catalytic amount of piperidine afforded ethyl 3,5-dicyano-1,6-dihydro-6-thioxopyridine-2-carboxylates **148**. Reaction of 6-thioxopyridines **148** with (2-acetoxyethoxy)methyl bromide in DMF and in the presence of sodium hydride afforded ethyl 3,5-dicyano-6-[(2-acetoxyethoxy)methylthio]pyridine-2-carboxylates **149** that is further reacted with ammonia in methanol to produce ethyl 3,5-dicyano-6-[(2-hydroxyethoxy)methylthio]pyridine-2-carboxylates **150**. Reaction of **148** with ethoxymethyl chloride in DMF and in the presence of sodium hydride gave ethyl 3,5-dicyano-6-(ethoxymethylthio)pyridine-2-carboxylates **151** [322] (**Scheme 58**).

2.16. Synthesis of Arylidenecyanoacetamide Derivatives

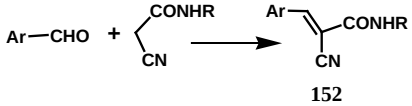
Arylidenecyanoacetamides **152** are prepared *via* Knoevenagel condensation of aldehydes with cyanoacetamide derivatives (**Scheme 59**). The reaction was carried out under solvent-free conditions [346], in water and in the presence of triethylbenzylammonium chloride (TEBA) [347], in aqueous medium at room temperature [326], or *via* grinding of aldehydes with cyanoacetamide derivatives at room temperature and in the presence of a catalytic amount of 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU) [348].

Reactivity of Arylidenecyanoacetamides

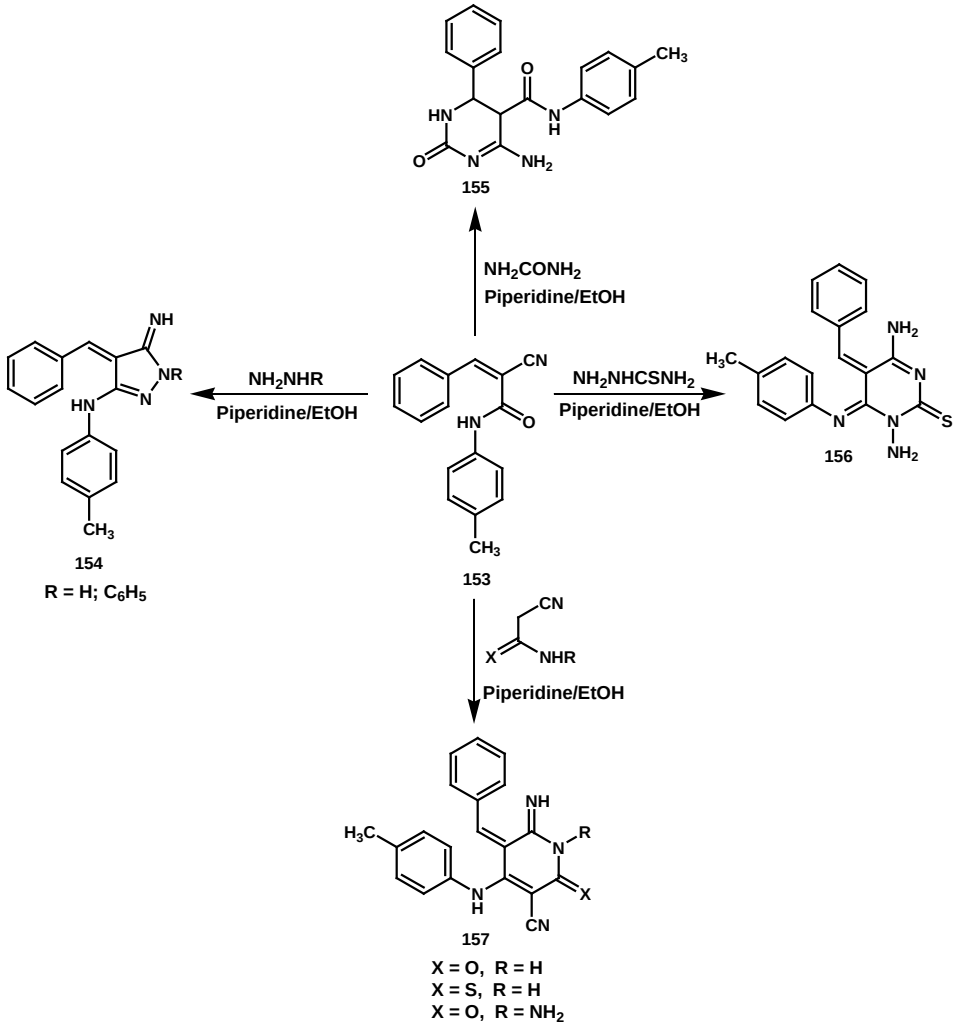
The benzylidenecyanoacetamide derivative **153** has been extensively utilized in heterocyclic synthesis. Reaction of **153** with hydrazine hydrate or phenylhydrazine in refluxing ethanol and in the presence of a catalytic amount of piperidine produced the pyrazole derivatives **154** (**Scheme 60**) [349]. The 2-oxopyrimidine derivative **155** was prepared through condensation of **153** with urea in refluxing ethanol and in the presence of a catalytic amount of piperidine (**Scheme 60**) [349]. In addition, the 2-thioxopyrimidine derivative **156** was obtained through



Scheme 58. Preparation of compounds **148-151**.



Scheme 59. Preparation of compounds **152**.



Scheme 60. Preparation of compounds **154-157**.

condensation of **153** with thiosemicarbazide in refluxing ethanol and in the presence of a catalytic amount of piperidine (**Scheme 60**) [349]. Moreover, reaction of **153** with cyanoacetamide, cyanothioacetamide or cyanoacetic acid hydrazide afforded the cyanopyridine derivatives **157** (**Scheme 60**) [349].

2.17. Synthesis of 5-Arylidene Derivatives of Barbituric and Thiobarbituric Acids

Barbituric and thiobarbituric acids have attracted the attention of medicinal chemists for over hundred years due to their therapeutic values [350] [351]. 5-Arylidenebarbiturate/thiobarbiturate derivatives are important members of the pyrimidine family. The major importance of these compounds has been centered on their application as useful precursors in the preparation of new heterocyclic compounds [293] and as selective oxidizing agents [352]-[354]. Barbituric acid and its derivatives exhibited different biological activities such as antibacterial, hypotensive and tranquilizing activities [355]. The clinical use of barbiturates in neurological disorders has also been investigated [356]. 5-Arylidenebarbiturates/thiobarbiturates **158** (**Scheme 61**) have been synthesized by Knoevenagel reaction of barbituric acid with different aldehydes under various conditions. The reaction was carried out under aqueous reflux using acetic acid as a catalyst [357]. Villemin and Labiad [358] synthesized 5-arylidenebarbiturates under microwave irradiation and in the presence of montmorillonite KSF clay. Dewan and Singh [359] reported various catalysts like NH_4OAc/AcOH, montmorillonite K-10, silica gel, basic alumina, NaCl, montmorillonite KSF, and KSF/NaCl for the synthesis of 5-arylidenebarbiturates/thiobarbiturates **158**. A grinding method has also been employed for the synthesis of **158** [360]. The same reaction was promoted by

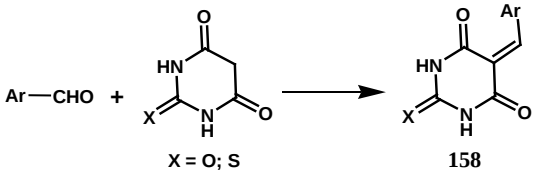
infrared irradiation in absence of solvent [361]. Also, it was carried out on basic alumina in a conventional microwave oven in the absence of solvent [362]. In addition, the same reaction has been achieved by employing bismuth chloride under solvent-free conditions [363]. Reddy *et al.* [364] reported the same reaction under microwave irradiation in absence of solvent and catalyst. In addition, Khan *et al.* [365] reported an improved, rapid and convenient method under eco-benign conditions *i.e.*, using water as a solvent and bismuth chloride as a catalyst at room temperature. Recently, 5-arylidenebarbiturate/thiobarbiturate derivatives **158** were obtained in excellent yields and high purity through condensation of barbituric or thiobarbituric acid with aromatic aldehydes in distilled water at room temperature and in the presence of a catalytic amount of ethanolamine [366] or L-tyrosine [367].

Reactivity of Arylidenebarbiturates and Thiobarbiturates

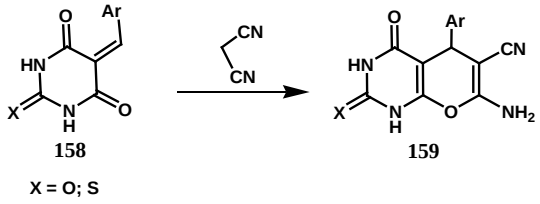
Reaction of **158** with malononitrile in ethanol and in the presence of a catalytic amount of piperidine afforded 7-amino-5-aryl-2-oxo(thioxo)-4-oxo-2,3,4,5-tetrahydro-1*H*-pyrano[2,3-*d*]pyrimidine-6-carbonitriles **159** [366] (**Scheme 62**). Cycloaddition reactions of 5-arylidenebarbiturate derivatives **158** with a tenfold excess of ethyl vinyl ether in methylene chloride at room temperature afforded 2*H*-pyrano[2,3-*d*]pyrimidine-2,4-(3*H*)-diones **160** [368] (**Scheme 63**).

2.18. Synthesis of Arylidene Derivatives of Meldrum's Acid

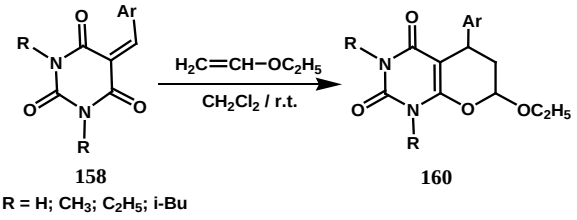
2,2-Dimethyl-1,3-dioxane-4,6-dione (Meldrum's acid) undergoes standard Knoevenagel condensation with aromatic and heteroaromatic aldehydes furnishing the corresponding arylidene derivatives **161** (**Scheme 64**), which are versatile substrates for different kinds of reactions [369]. In addition, they are useful intermediates for cycloaddition reaction and for the synthesis of heterocyclic compounds with potential pharmacological activity [370]. The Knoevenagel condensation of aldehydes with Meldrum's acid is generally catalyzed by bases, such as pyridine [371] or by piperidine/glacial acetic acid in benzene [372]. Uncatalyzed reaction was reported in literature using DMF or DMSO as solvent [373]. In addition, anhydrous zinc chloride was reported to promote the reaction in absence of any solvent [374]. The same reaction was also carried out in water [375]. In addition, the Knoevenagel condensation of aromatic aldehydes with Meldrum's acid proceeded efficiently in the recyclable



Scheme 61. Preparation of compounds **158**.



Scheme 62. Preparation of compounds **159**.



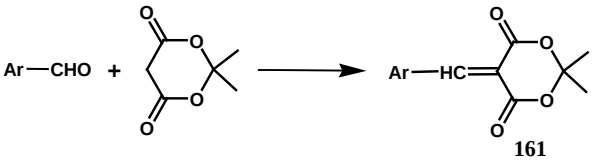
Scheme 63. Preparation of compounds **160**.

ionic liquid [bmim]BF_4 at room temperature and in the presence of a catalytic amount of piperidine [376]. Also, the same condensation was carried out in methanol at room temperature [377].

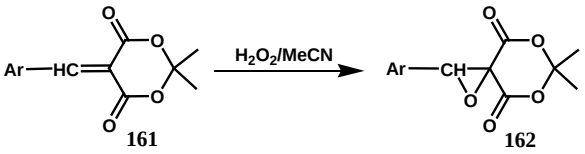
Reactivity of Arylidene Derivatives of Meldrum's Acid

The epoxide analogues of arylidene Meldrum's acid **162** were prepared through reaction of **161** with hydrogen peroxide in acetonitrile at room temperature [378] [379] (**Scheme 65**).

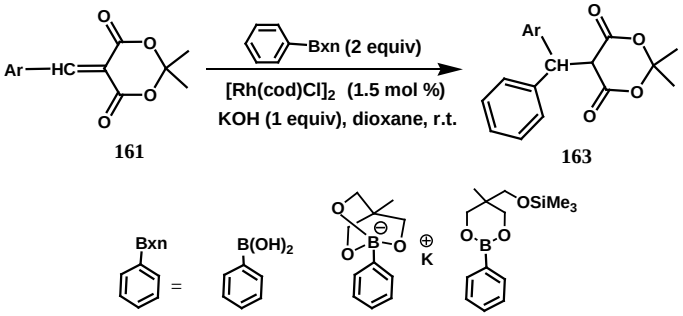
Rhodium-catalyzed additions of arylboron reagents to **161** in dioxane at room temperature gave compounds **163** [380] (**Scheme 66**). The condensation of **161** with 3-amino-1,2,4-triazole in nitrobenzene afforded 4,5,6,7-tetrahydro-1,2,4-triazolo[1,5-*a*]pyrimidin-5-ones **164**. In DMF, the reaction proceeds with the formation of arylsubstituted *N*-(2*H*-1,2,4-triazol-3-yl)-3-(2*H*-1,2,4-triazol-3-ylamino)propionamides **165**, in addition, the amide **166** and the aldehydes were present in the reaction mixture [381] (**Scheme 67**).



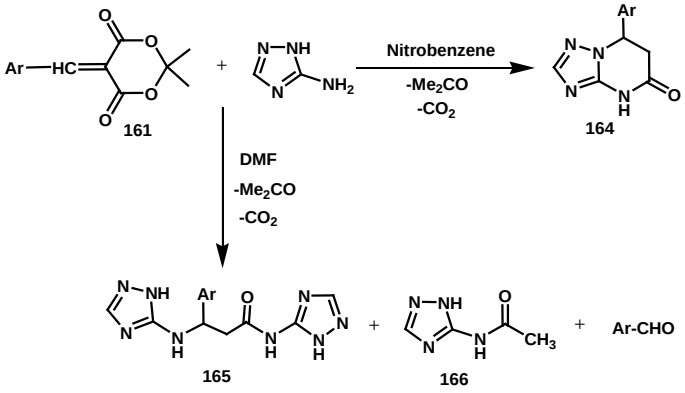
Scheme 64. Preparation of compounds **161**.



Scheme 65. Preparation of compounds **162**.



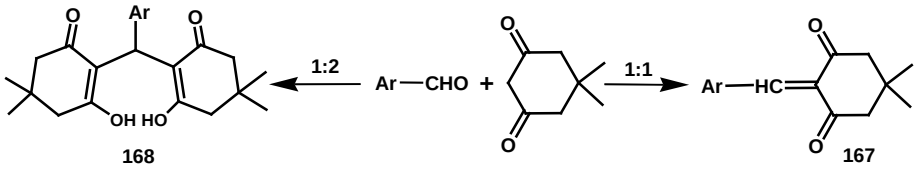
Scheme 66. Preparation of compounds **163**.



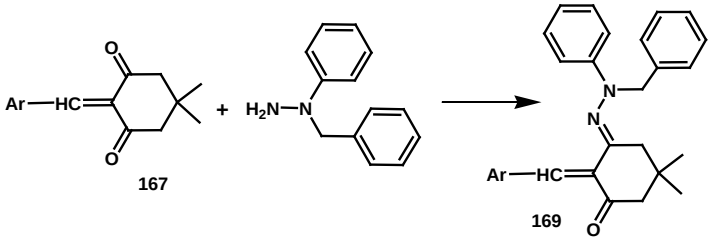
Scheme 67. Preparation of compounds **164** and **165**.

2.19. Synthesis of 2-Arylidene Dimedone and Bisdimedone Derivatives

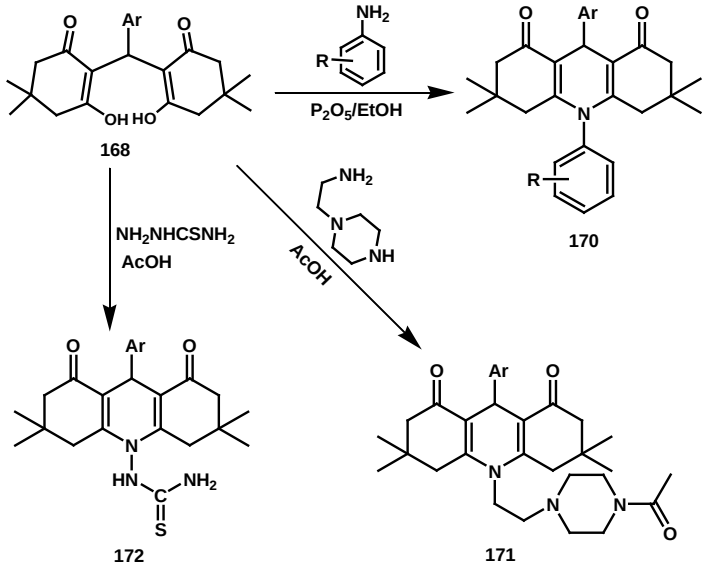
5,5-Dimethylcyclohexane-1,3-dione (dimedone) was condensed with aromatic aldehydes in equimolar ratio and in presence of bases such as potassium hydroxide [382] [383] or piperidine [383] to give 2-arylidene-5,5-dimethylcyclohexane-1,3-dione derivatives **167** (**Scheme 68**). The same products were obtained through fusion of equimolar amounts of the aromatic aldehydes and dimedone in an oil bath at 150˚C [384]. On the other hand, reaction of dimedone with aromatic aldehydes in 2:1 molar ratio afforded the bisdimedone derivatives **168** (**Scheme 68**), several reaction conditions were reported for this reaction. The reaction was performed in refluxing aqueous ethanol and in the presence of a catalytic amount of piperidine [385], under solvent-free conditions [346], in aqueous ethanol at room temperature and in the presence of a catalytic amount of piperidine [386], in water at 100˚C and in the presence of a catalytic amount of iodine [387], $HClO_4$-SiO_2 or PPA-SiO_2 [388], in refluxing aqueous methanol [389], in aqueous media at room temperature [390] or in refluxing acetonitrile and in the presence of of zinc oxide as a catalyst [391], also it was carried out in dry methylene chloride and in the presence of silica chloride nano particle (nano SiO_2-Cl) [392] to afford the corresponding 2,2'-(arylmethylene)*bis*(3-hydroxy-5,5-dimethyl-2-cyclohexen-1-one) (bisdimedone derivatives) **168**. The same reaction was performed using ytterbium triflate [$Yb(OTf)_3$-SiO_2] and amine as a catalytic system under solvent-free conditions



Scheme 68. Preparation of compounds **167** and **168**.



Scheme 69. Preparation of compounds **169**.



Scheme 70. Preparation of compounds **170-172**.

[393]. This method is advantageous as being eco-friendly, non-corrosive, and allows reutilization of the catalytic system. Recently, bisdimedone derivatives **168** were obtained through reaction of dimedone with aromatic aldehydes in 2:1 molar ratio in ethylene glycol and in presence of nickel nanoparticles [394].

Reactivity of 2-Arylidene Dimedone and Bisdimedone Derivatives

Reaction of arylidenedimedone derivatives **167** with *N*-benzyl-*N*-phenylhydrazine in 50% acetic acid produced compounds **169** [384] (**Scheme 69**).

When bisdimedone derivatives **168** were reacted with different amines in ethanol and in the presence of a catalytic amount of P_2O_5, 10-(substituted phenyl)-3,4,6,7,9,10-hexahydro-1,8(2*H*, 5*H*)-acridinedione derivatives **170** were obtained. The condensation of *N*-(2-aminoethyl)piperazine with bisdimedones **168** in acetic acid afforded the acridinediones **171** in which the *N*-acetylation of the piperazine ring has also occurred. The reaction of bisdimedones **168** with thiosemicarbazide gave *N*-(3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-1,8-dioxo-(2*H*, 5*H*)-acridin-10-yl)thiourea derivatives **172** [395] (**Scheme 70**).

3. Conclusion

Literature data have been summarized to help chemists to find information appropriate for the high synthetic potential of different arylidene derivatives. Syntheses of many biologically active heterocyclic compounds belonging to these compounds have also been reported.

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