Review Paper:

Ultrasound assisted Heterocycles Synthesis

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Abstract

This current review focuses on ultrasound irradiation literature published between 2008– 2015 and is a continuation of the previous published work[2] which also reviewed the literature published from 1980–2007. This review revives the effect of ultrasound on the development of chemical reaction and to highlight some applications of sonochemistry in various organic synthesis such as alkylation, condensation and cycloaddition.

Keywords: Ultrasound, Heterocycles, Synthesis.

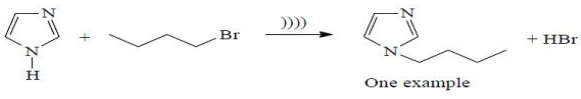
Introduction

Sonochemistry can be defined as a technique to promote chemical reactions under the influence of ultrasound irradiation which is a part of the sonic spectrum that ranges from 20 kHz to 100 kHz[2]. Moreover, sonochemistry is incorporated in several fields including clinical, industrial and research applications in various branches of chemistry as well as in the synthesis of micro and nanomaterials which have established sonochemistry as an important tool in environmentally clean technology[3].

The chemical effects of ultrasound have three classes of reactions[4]. Homogeneous systems that are done by means of radical or radical-ion intermediates are defined under homogeneous sonochemistry. On the other hand, heterogeneous sonochemistry (liquid-liquid or solid-liquid systems): heterogeneous systems are done by means of ionic intermediates heterogeneous reactions that include a radical and ionic mechanism are so-called sonocatalysis (overlap homogeneous and heterogeneous sono-chemistry). There are three classes of reactions influenced by the chemical effects of ultrasound.

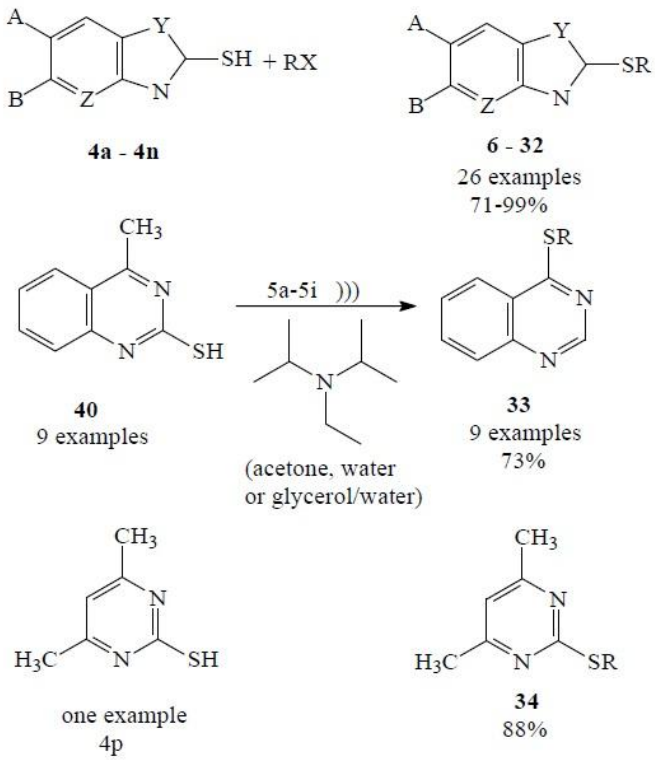
Alkylation

In 2010, Ferrera-Escudero et al[12] reported synthesis of N-alkylimidazoles (3) from imidazole (1) and 1-bromobutane under sonochemical irradiation (15–120 min) and conventional thermal conditions (293 K, 3113 K, 333 K) using different amounts of catalysts (Scheme 1). Reactions performed with a combination of ultrasound irradiation with alkaline-promoted carbons afforded excellent yields under very mild conditions. Unfortunately, the yields obtained by this methodology were not specified by the authors.



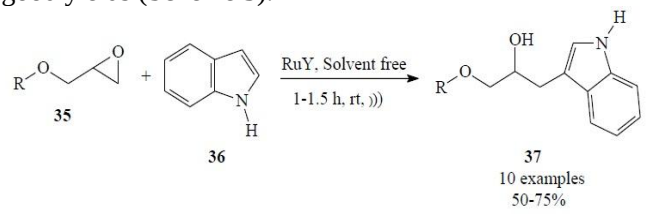
Scheme 1

Deligeorgiev et al[8] described the synthesis of 2-alkylthio derivatives of hetaryl thiols (6–34) via selective S-alkylation with alkyl halides (bromides and iodides) under ultrasonic irradiation at room temperature. The products were obtained in high to excellent yields and in high purity (Scheme 2).



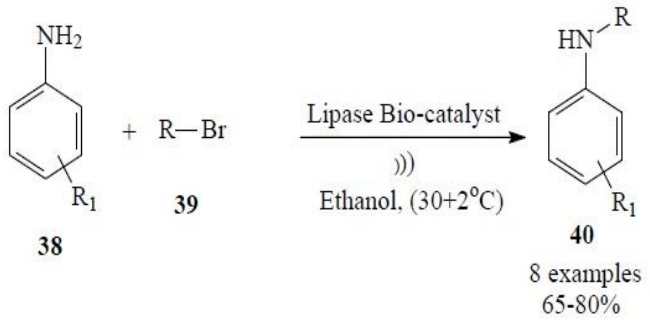
Scheme 2

Khorshidi[22] explored the ultrasonic irradiation assisted synthesis of 3-alkylated indole derivatives (37) from epoxides (35) and indoles (36) catalyzed by ruthenium-exchanged FAU-Y zeolite. The compounds were isolated in good yields (Scheme 3).

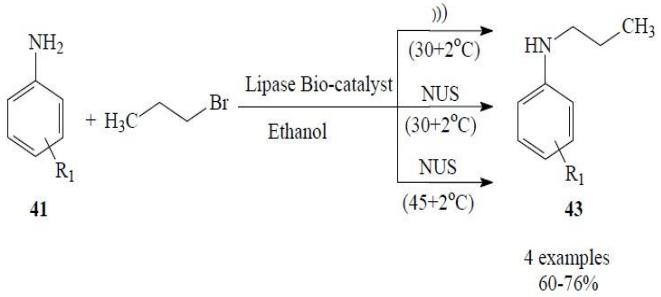


Scheme 3

In 2013, Lobo et al[29] described the synthesis of mono-N-alkyl aromatic amines in the presence of bio-catalyst under sonochemical irradiation at room temperature (Scheme 4). The comparison evidence that the ultrasonic irradiation enhances mono-N-alkyl of aromatic amines is better than the non-sonochemical method (Scheme 5).

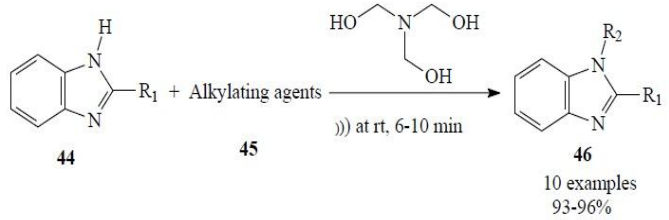


Scheme 4



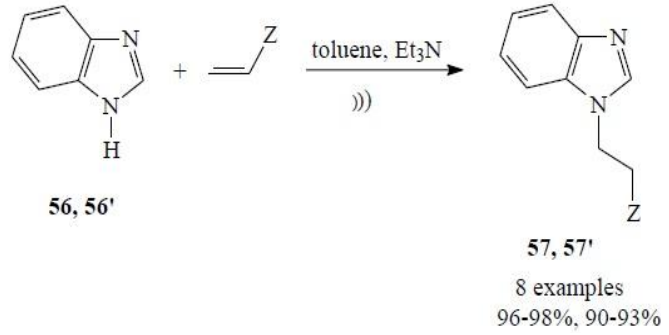
Scheme 5

Srinivas et al[42] synthesized a series of N alkyl benzimidazole (46) via the reaction of 2- substituted 1*H*-benzimidazoles (44) with different alkylating agents (45) using triethanol amine as solvent under ultrasonic irradiation (Scheme 6).

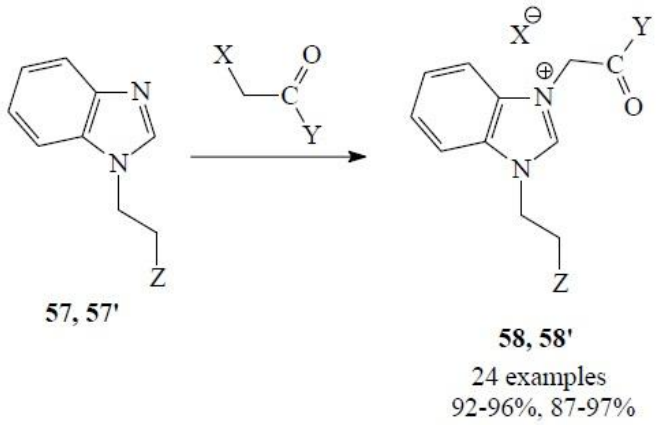


Scheme 6

Zbancioc et al[47] synthesized a series of 1, 3-diazole derivatives under sonochemical irradiation through sequential reactions of imidazole and benzimidazole (56, 56') which react with acrylic acid derivatives in the presence of trimethylamine to give the target N-alkylated compounds (57, 57') as shown in scheme 7 which react with activated halogen-derivatives in the presence of acetone as solvent to give the desired imidazolium salts and benzimidazolium salts (58, 58'). These derivatives were synthesized in excellent yield (Scheme 8).



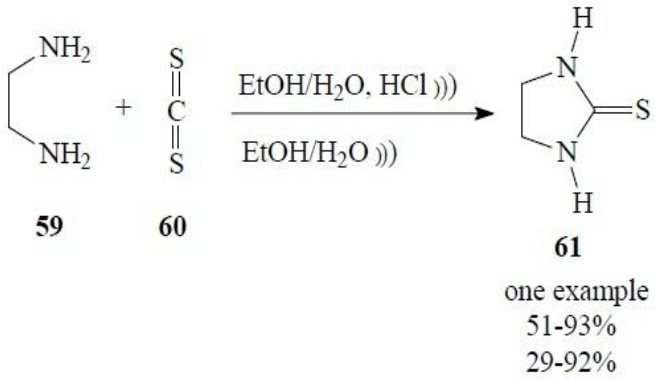
Scheme 7



Scheme 8

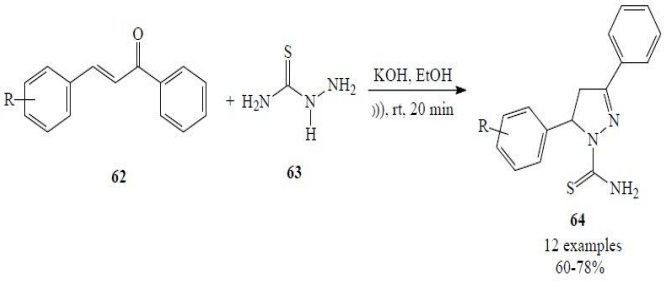
Condensation

Synthesis of imidazolidine-2-thione (61) was reported by Entezari et al[10] by the reaction between ethylene diamine (59) and carbon disulfide (60) under ultrasound irradiation in the presence of ethanol/water, HCl or ethanol/water as solvents (Scheme 9).



Scheme 9

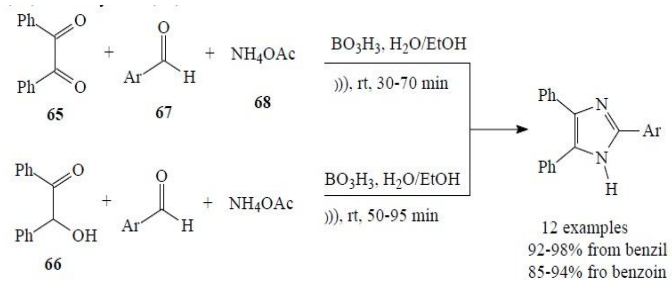
Pizzuti et al[34] reported an efficient and eco-friendly methodology for the synthesis of 1-thio-carbonyl-3,5-diaryl-4,5-dihydro-1*H*-pyrazoles (64) from chalcones (62) and thiosemi-carbazide (63) in ethanol and KOH under sonication. The desired products were isolated in good yields (Scheme 10).



Scheme 10

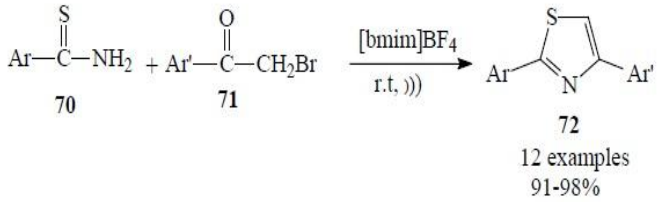
Shelke et al[41] described the synthesis of 2,4,5-triaryl-1*H*-imidazoles (69) from the three component one-pot condensation of benzil (65)/ benzoin (66), aldehydes (67) and ammonium acetate (68) in aqueous media under

ultrasonic irradiation at room temperature in the presence of BO_3H_3 as catalyst (Scheme 11).

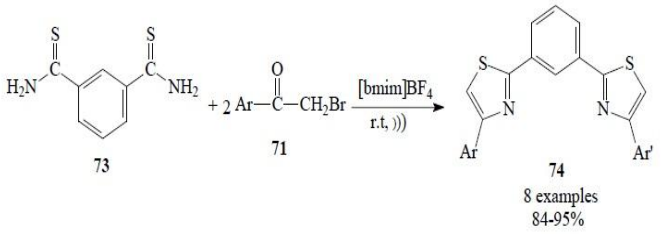


Scheme 11

Noei and Khosropour[33] reported the synthesis of 2,4-diarylthiazole derivatives (72 and 74) in high yields via the reaction of aryl thioamides (70 and 73) with α-bromo acetophenones (71) under sonication at room temperature in the ionic liquid [bmim]BF_4 (Schemes 12 and 13).

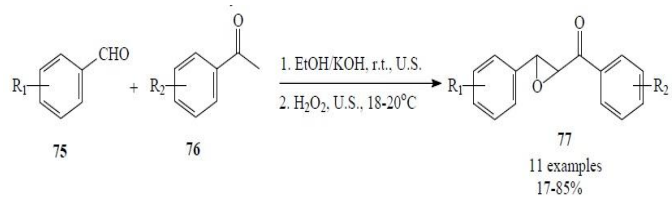


Scheme 12



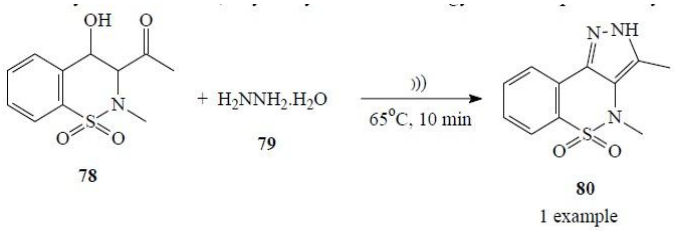
Scheme 13

In 2010, Li et al[27,28] reported the rapid and efficient preparation of 2,3-epoxyl-1,3-diaryl-1- propanone (77) from aromatic aldehydes (75) and acetophenones (76) under sonication at room temperature (Scheme 14).



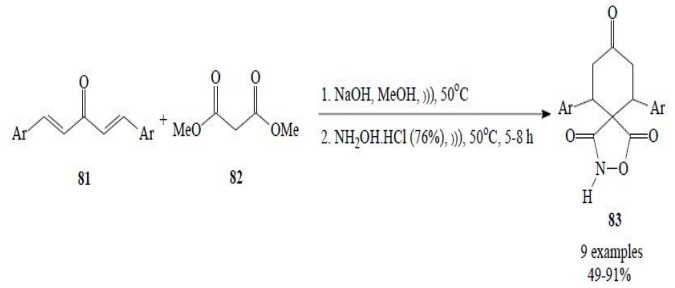
Scheme 14

Ahmad et al[1] explored the ultrasonic irradiation assisted synthesis of 3,4-dimethyl-2,4- dihydropyrazolo[4,3-c][1,2]benzothiazine-5,5- dioxide (80) via the cyclization of 1-(4-hydroxy- 2-methyl-1,1-dioxido-2*H*-1,2-benzothiazin-3-yl) ethanone (78) with hydrazine (79) (Scheme 15). Unfortunately, the yield obtained by this methodology was not specified by the authors.



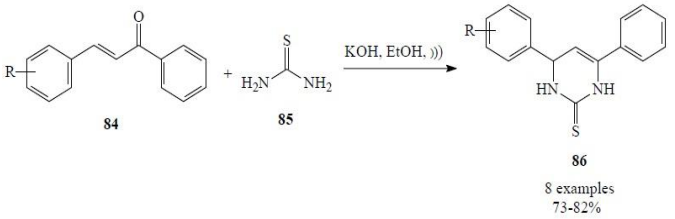
Scheme 15

Li et al[26] reported the synthesis of 3-aza- 6,10-diaryl-2-oxaspiro[4,5]decane-1,4,8-trione (83) from the three-component one-pot condensation of 1,5-diaryl-1,4-pentadien-3-one (81), dimethyl malonate (82) and hydroxylamine hydrochloride in the presence of sodium hydroxide under ultrasound irradiation at 50°C. The products were obtained in good yields (Scheme 16).



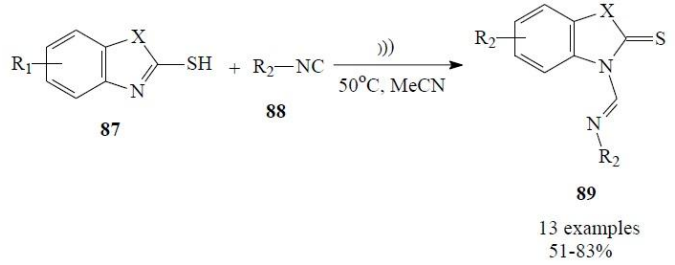
Scheme 16

In 2011, Ghomi and Ghasemzadeh[15] described the synthesis of pyrimidine-2-thion derivatives (86) under ultrasonic irradiation via the reaction of chalcones (84) and thiourea (85). The reactions afforded the desired products in good to very good yields (Scheme 17).



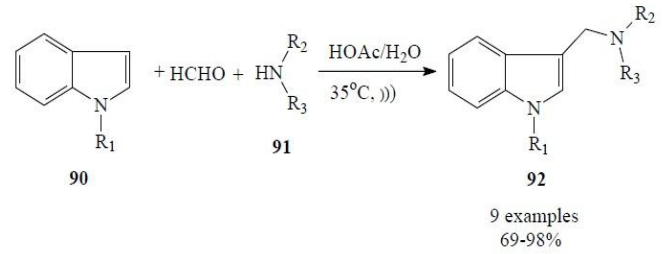
Scheme 17

Zhu et al[48] reported the synthesis of a series of formamidine framework (89) via direct reactions of 2-mercaptobenzthiazole/2-mercapto-benzoxazole (87) with isocyanides (88) under ultrasonic irradiation. The compounds were isolated in moderate to high yields (Scheme 18).



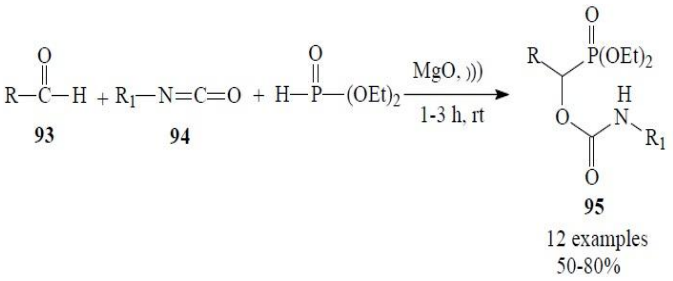
Scheme 18

Li et al[27,28] explored the synthesis of 3- (dialkyl aminoethyl)-indole (92) via Mannnich reaction of secondary amine (91), formaldehyde and indole or N-methyl indole (90) in the presence of acetic acid/water at 35°C under sonication (Scheme 19).



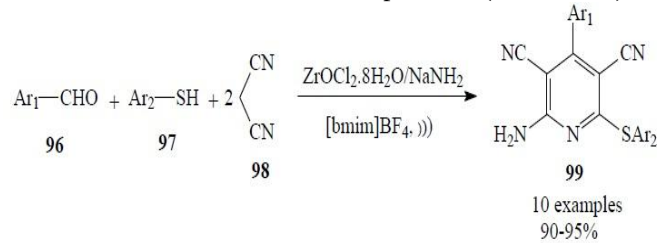
Scheme 19

Kaboudin and Fallahi[21] presented a green synthesis of α-oxycarbanilino phosphonates (95) via the three-component condensation consisted of the reaction of aldehyde (93), diethyl phosphite and isocyanate (94) in the presence of magnesium oxide under solvent-free conditions and under ultrasonic irradiation. The desired products were isolated in moderate to good yields (Scheme 20).



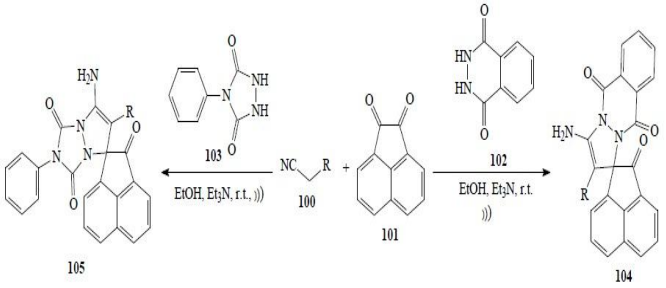
Scheme 20

In 2011, Poor Heravi and Fakhr[35] reported an efficient and eco-friendly methodology for the synthesis of 2-amino-6-(arylthio)-4-aryl pyridine-3,5dicarbonitrile derivatives (99) in high yields by the one-pot condensation of aldehyde (96), aryl thiol (97) and malononitrile (98) catalyzed by $ZrOCl_2.7H_2O/NaNH_2$ in the ionic liquid [bmim] BF_4 under ultrasound irradiation at room temperature (Scheme 21).



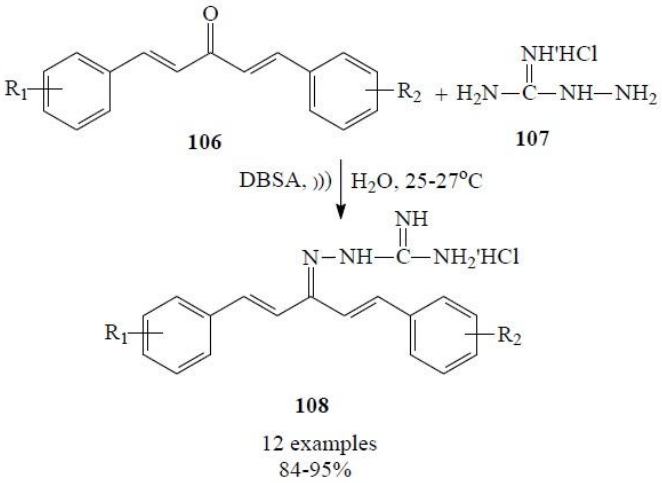
Scheme 21

Rezaei et al[38] presented a green synthesis of spiroacenaphthylene-1,1'-pyrazolo- [1,2-b]phthalazin and spiroacenaphthylene-1,5'- pyrazolo[1,2-a][1,2,4]triazole derivatives (104, 105) in high yields via the three component condensation of nitrile (100), acenaphthylene-1,2-dione (101) and phthalhydrazide (102) or 4- phenyl-1,2,4-triazolidine-3,5-dione (103) in the presence of Et_3N in ethanol as catalyst under sonication (Scheme 22).



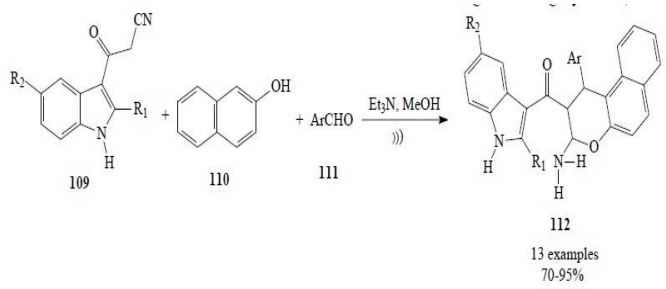
Scheme 22

Li et al[27,28] explored an efficient and environmental friendly protocol for the synthesis of 2-(1,5-diaryl-1,4-pentadien-3-ylidene)- hydrazine carboximidamide hydrochloride (108) by the condensation of 1,5-diaryl-1,4-pentadiene- 3-one (106) with aminoguanidine hydrochloride (107) catalyzed by DBSA in water under ultrasonic irradiation (Scheme 23).



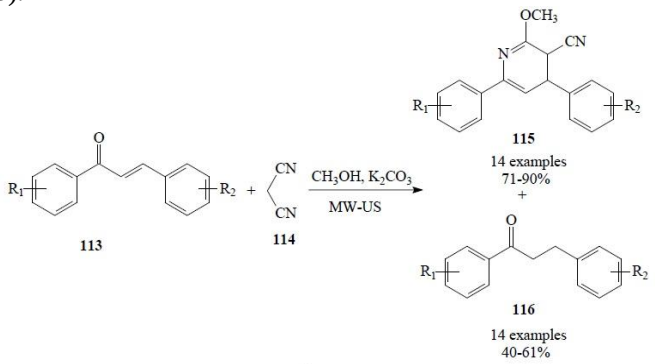
Scheme 23

Hossein et al[19] described the efficient synthesis of novel 1*H*-benzol[f] chromen-indole derivatives (112) under sonocation via one-pot three-component reaction of 3-cyanoacetyl indoles (109), β-naphthol (110) and aryl aldehydes (111) in the presence of methanol and triethyl amine as a catalyst. The desired products were isolated in good to high yields (Scheme 24).



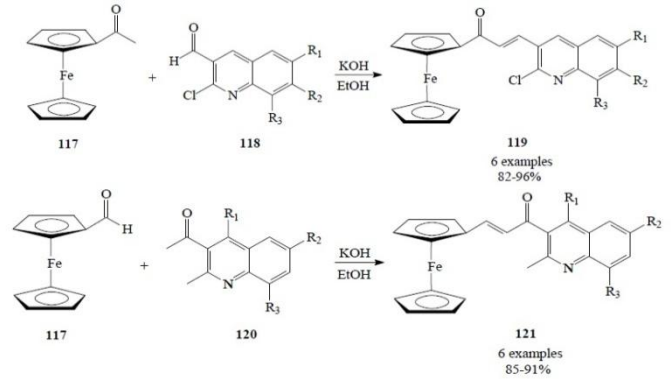
Scheme 24

Feng et al[11] have developed an efficient methodology for the synthesis of polysubstituted pyrydines (115, 116) in good yield by a K_2CO_3- promoted condensation of chalcone (113), malonitrile (114) and methanol under combined microwave and ultrasound irradiation (Scheme 25).



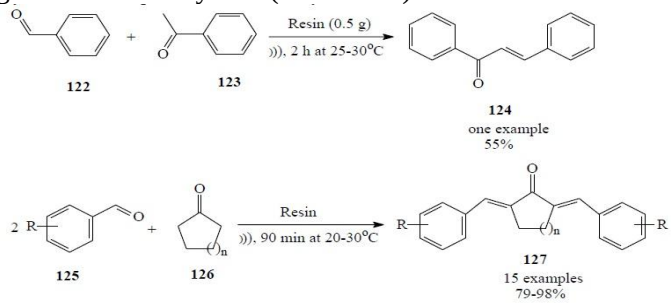
Scheme 25

Prasath et al[36] explored the facile and efficient synthesis of quinoline-appended ferrocenyl chalcones (119 and 121) under ultrasound irradiation via condensation of 2- chloro-3-acetyl quinoline (118) with 2-methyl-3- formyl quinoline (120) in the presence of KOH and ethanol. The products were obtained in good to excellent yields in short reaction times (Scheme 26).



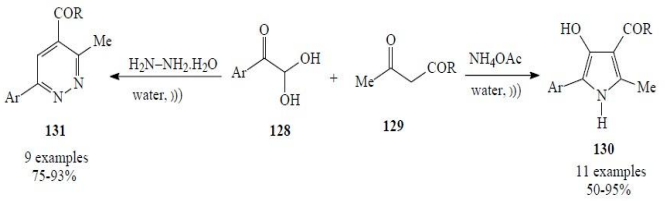
Scheme 26

Lahyani et al[24] synthesized trans-chalcones (124) and α,α'-bis(arylmethylidene) cycloalkanones (127) derivatives by aldol and cross-aldol condensation reaction. The reaction was carried out by the condensation of benzaldehyde (122), acetophenone (123) and aryl aldehyde (125), cyclopentanone (126) respectively in the presence of acid-resins as catalysts in solvent free conditions under ultrasonic irradiation. All products were synthesized in good to excellent yields (Scheme 27).



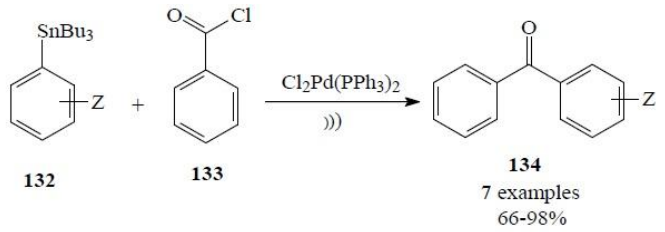
Scheme 27

Efetekhari-Sis and Vahdati-Khajch[9] reported an efficient and environmental friendly method for the synthesis of 5-aryl-4-hydroxy-1*H*-pyrrole-3-carboxylic acid esters (130) and 6-aryl- 3-methylpyridazine-4-carboxylic acid esters (131) under ultrasound irradiation via one-pot three component condensation of aryl glyoxal hydrates (128) with β-dicarbonyl compounds (129) in the presence of ammonium acetate, hydrazine hydrate and water as solvent. The products were obtained in good to high yields (Scheme 28).



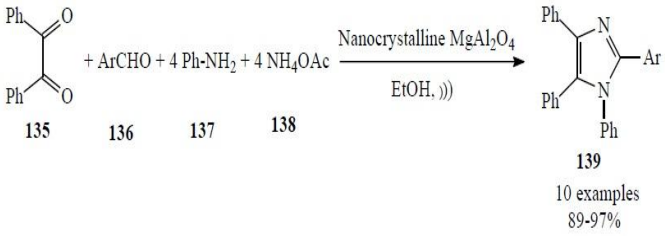
Scheme 28

Luong et al[30] synthesized a series of diaryl ketones (134) in moderate to excellent yields by means of still cross-coupling reaction of substituted aryl tributyl stannanes (132) with benzoyl chlorides (133) in the presence of $Cl_2Pd(PPh_3)_2$ as catalyst under ultrasonic irradiation (Scheme 29).



Scheme 29

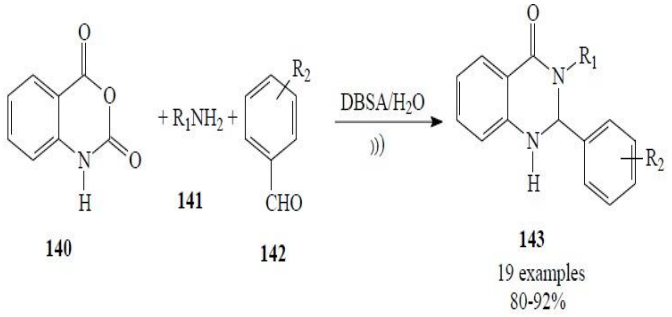
Safari et al[40] explored green and efficient protocol for the synthesis of 1,2,4,5-tetra-substituted imidazoles (139) via four-component condensation of benzil (135), aldehyde (136), primary aromatic amine (137) and ammonium acetate (138) in presence of nanocrystalline magnesium aluminate as catalyst and ethanol under ultrasound irradiation. The reactions proceeded easy and afforded the products in high yields (Scheme 30).



Scheme 30

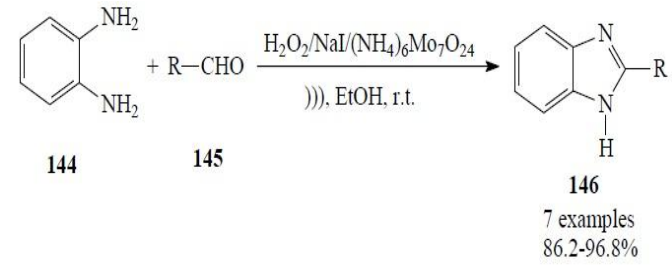
Chen et al[7] worked on synthesis of 2,3- disubstituted-2,3-dihydroquinazolin-4(1*H*)-one derivatives (143) through the one-pot three-component condensation of isatoic anhydride (140), amine (141) and aromatic aldehyde (142) using a catalytic amount of dodecyl benzene sulfonic acid in aqueous media at 40-42°C under ultrasonic irradiation. This methodology offers several advantages including mild

reaction conditions, short reaction times, high yields and being eco-friendly (Scheme 31).



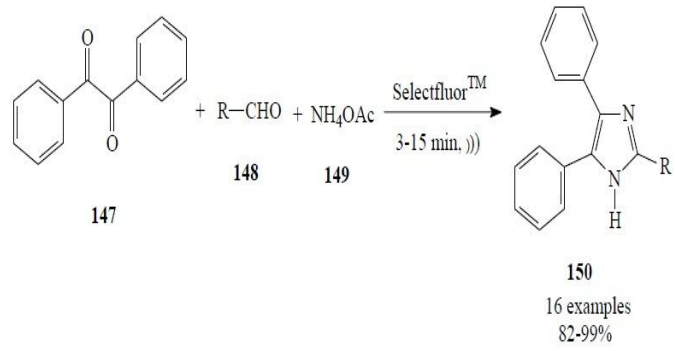
Scheme 31

Bai et al[4] developed a novel sodium iodide and ammonium molybdate co-catalytic system for the aqueous-mediated ultrasound-promoted synthesis of 2-benzimidazoles (146) by the reaction of *o*-phenylene diamine (144) and aldehyde (145) at room temperature. The desired products were isolated in high yields (Scheme 32).



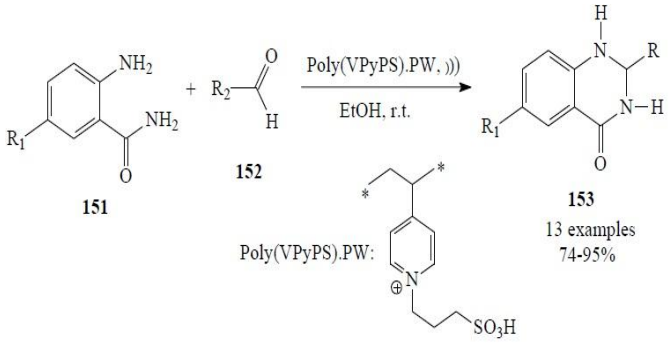
Scheme 32

Heravi et al[18] developed novel selectfluorTM efficiently catalyzed the reaction of benzil (147), aryl aldehydes (148) and ammonium acetate (149) under ultrasonic irradiation and solvent-free to afford 2,4,5-tri-substituted imidazoles (150) in excellent yields (Scheme 33).



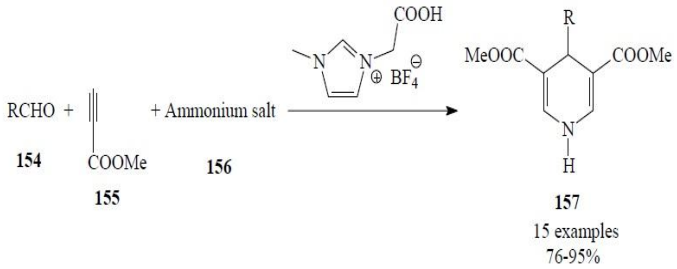
Scheme 33

Wang et al[43] designed poly(4-vinyl pyridine) supported acidic ionic liquid as novel solid catalysts and used it into the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones (153) via cycloaddition reaction of an anthranilamide (151) with aldehydes (152) under ultrasound irradiation. This method provides good to excellent yields of the desired products (Scheme 34).



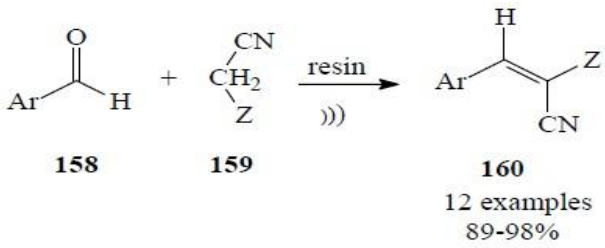
Scheme 34

He et al[17] presented ultrasound-promoted synthesis of 4-substituted 1,4-dihydropyridine- 3,5-dicarboxylates (157) via the reaction of aldehyde (154), methyl propiolate (155) and ammonium carbonate (156) in the presence of ionic liquid 1-carboxy methyl-3-methyl imidazolium tetrafluoroborate as an efficient catalyst under ultrasonic irradiation at room temperature. The products are in moderate to high yields (Scheme 35).



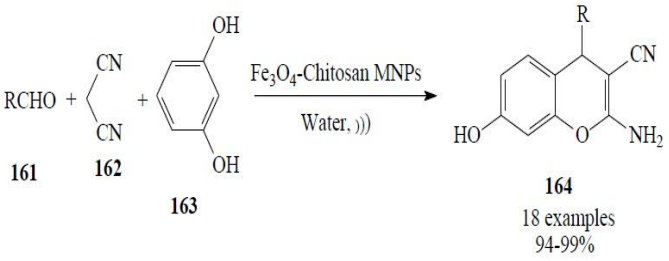
Scheme 35

Ammar et al[3] carried out the Knoevenagel condensation of aromatic aldehydes (158) with active methylene groups (159) in the presence of anion-exchange resins as an efficient and green catalyst under ultrasonic irradiation. This method provides high yields of derivatives (160) in short time under mild conditions (Scheme 36).



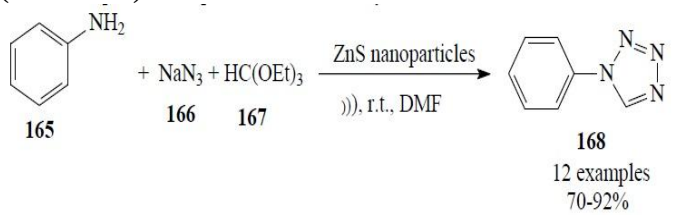
Scheme 36

Safari and Javadian[39] presented ultrasound-promoted synthesis of 2-amino-4*H*-chromene derivatives (164) via one-pot condensation of aldehydes (161) with malonitrile (162) and resorcinol (163) using Fe_3O_4-chitosan nanoparticles as a magnetic heterogeneous catalyst. This novel methodology provides several advantages including higher yields of products and mild reaction conditions as well as a simple experimental and reusability of the magnetic nanocatalyst (Scheme 37).



Scheme 37

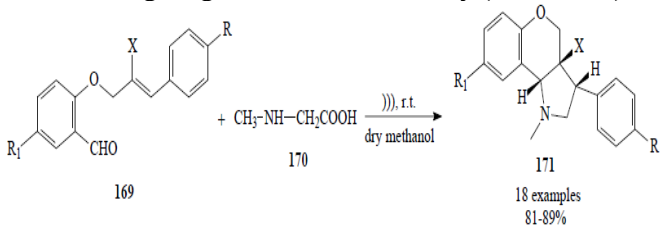
Naeimi and Kiani[32] synthesized three-component, one-pot condensation reaction of various primary amines (165), sodium azide (166) and triethyl orthoformate (167) in the presence of zinc sulphide nanoparticles as a catalyst at room temperature under ultrasonic irradiation to afford 1-substituted tetrazoles (168). This novel method offers high yields shorter reaction time and catalyst can be recyclable (Scheme 38).



Scheme 38

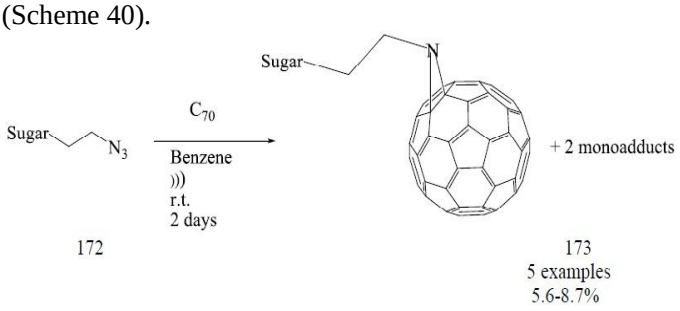
Cycloaddition

Ramesh et al[37] developed a simple and efficient method for the synthesis of a variety of chromeno[4,3-b] pyrroles (171) by intramolecular 1,3-dipolar cycloaddition reaction of an O-allyl salicylaldehyde derivative (169) which derived from Baylis-Hillman adducts as dipolarophiles with sarcosine (170) in anhydrous methanol under ultrasonic irradiation at room temperature. This method offers good yields with high regio-and stereoselectivity (Scheme 39).



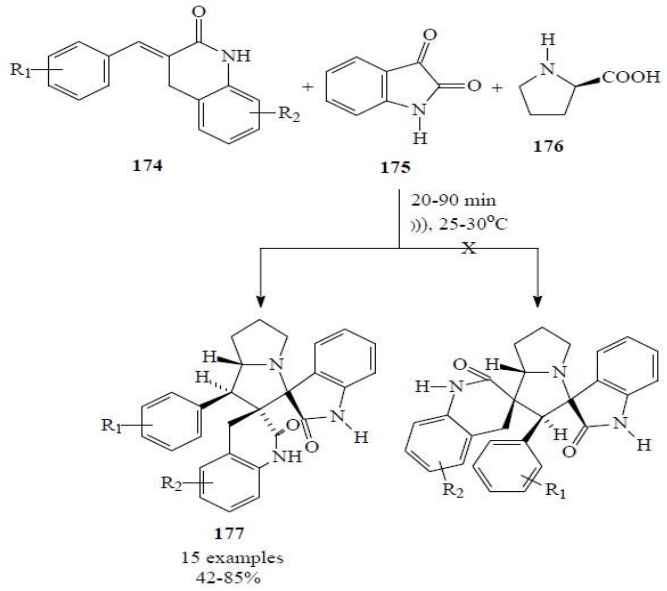
Scheme 39

Yoon et al[45] carried out the synthesis of glycosyl [70] fullerene derivatives (173) via cycloaddition reaction of [70] fullerene with 2- azidoethyl glycosides (172) under ultrasound irradiation for 2 days at room temperature (Scheme 40).



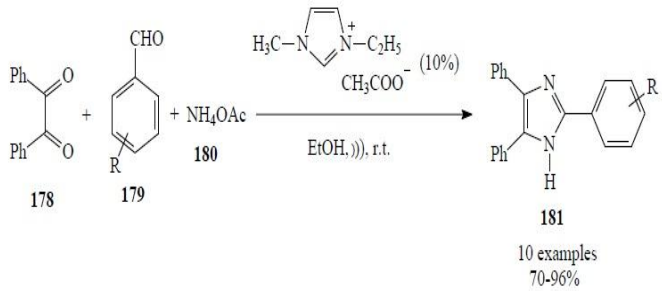
Scheme 40

Ge et al[13] synthesized a novel tetracyclic frameworks of dispiropyrrolizidines (177) under ultrasonication irradiation via the 1,3-dipolarcycloaddition of azomethine ylides using isatin (175) and L-proline (176) with 3- benzylidene-3,4-dihydroquinolin-2-(1*H*)-one (174) as dipolarophiles which is derived from aza-Claisen rearrangement of Baylis-Hillman amines. This method offers moderate to good yields (Scheme 41).



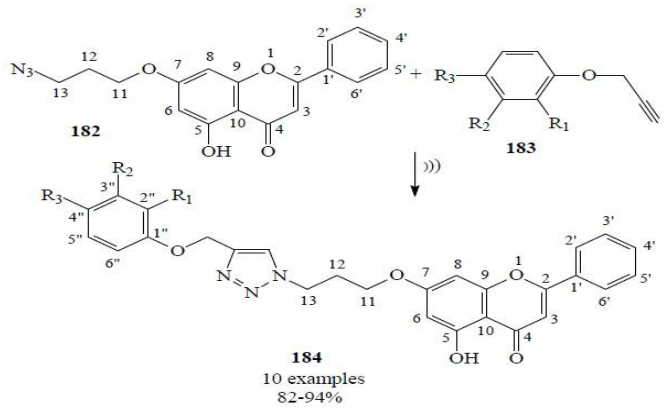
Scheme 41

Zang et al[46] performed one-pot synthesis of 2-aryl-4,5-diphenyl imidazoles (181) via the three-component reaction of benzil (178), aromatic aldehyde (179) and ammonium acetate (180) using 1-ethyl-3-methyl imidazole acetate as catalyst under ultrasonic irradiation at room temperature. This method provides good to excellent yields of the desired products (Scheme 42).



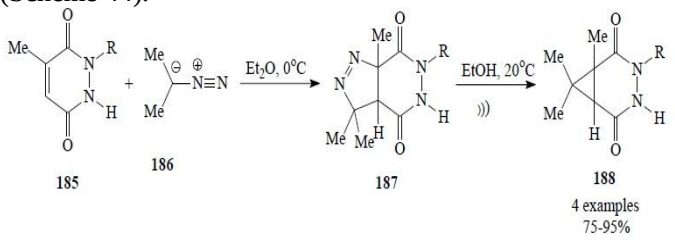
Scheme 42

Jiang et al[20] prepared a series of 7-(3-(4- phenyl-1,2,3-triazol-1-yl)propoxy)-5-hydroxy flavone derivatives (184) via ultrasound-promoted 1,3-dipolar Huisgen cycloaddition reaction between 7-(3-azido-propoxy)-5-hydroxy flavone (182) and substituted terminal alkyne (183) in the presence of t-BuOH/H2O (1:1 v/v) as solvent and CuSO4.5H2O/sodium ascorbate as the catalyst at room temperature. This methodology provides good to excellent yields of the desired products (Scheme 43).



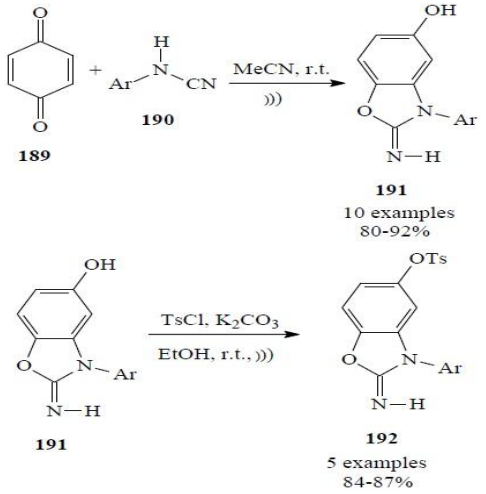
Scheme 43

Hamadi and Msaddek[5] designed a series of pyrazolines (187) via the 1,3-dipolar cycloaddition of 2-diazopropane (186) to pyridazine-3,6-dione derivatives (185). Moreover, ultrasound-assisted stereospecific preparation of bicyclo-cyclopropanes (188) through the irradiation of pyrazolines derivatives was made in the presence of ethanol as a solvent. This method provides good to excellent yields (Scheme 44).



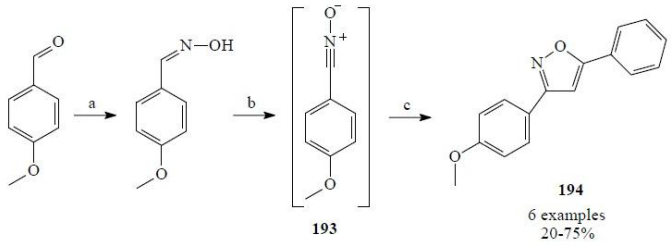
Scheme 44

Habibi et al[16] synthesized a class of novel 2-iminooxazolidines (191) via the reaction of arylcyanamides (190) with *p*-benzoquinone (189) at room temperature under ultrasonic irradiation in excellent yields. They also reported the chemoselective tosylation of the products which afforded the O-tosylated compounds (192) in good yields (Scheme 45).



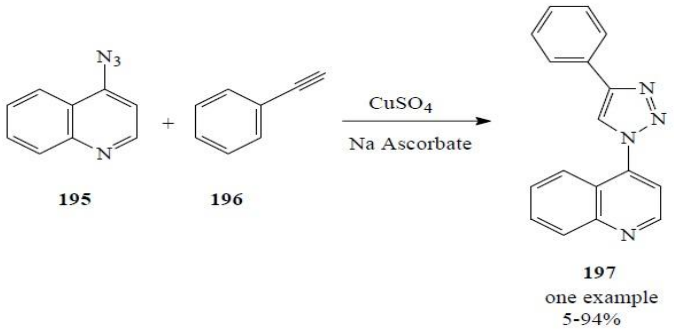
Scheme 45

Koufaki et al[23] designed one-pot three-step process for the synthesis of 3,5-disubstituted isoxazoles (194) via cycloaddition reaction between aromatic nitrile oxides (193) and alkynes in the presence of copper(I) as a catalyst under ultrasound irradiation. This methodology provides low to moderate yields of the desired products (Scheme 46).



Scheme 46

Marullo et al[31] worked on synthesis of 4- (4-phenyl-1*H*-1,2,3-triazol-1-yl)quinoline (197) via cycloaddition reaction between 4-azido-quinoline (195) and phenyl acetylene (196) in the presence of copper as catalyst under ultrasonic irradiation. This method offers low to excellent yields of the desired products (Scheme 47).



Scheme 47

Conclusion

In conclusion, this survey shows that ultrasound-assisted synthesis is an important tool of green chemistry as illustrated by the reactions presented here. Moreover, it is a fairly new technique it is now increasingly applied in research such as organic synthesis and development of nanoparticles catalysts as the present review shows. The advantages of ultrasound in chemical reactions are milder conditions, shorter reaction times, higher yields and environmental friendly reaction conditions, but the increasing requirement for environmentally clean technology that minimizes the production of waste at source is an important factor.

Acknowledgement

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