Journal of the Serbian Chemical Society
JSCS-info@shd.org.rs • www.shd.org.rs/JSCS
J. Serb. Chem. Soc. *82* (7–8) 755–790 (2017) UDC 534–8:542.913+547+54–145.2:547.7
JSCS-5001 *Review*

REVIEW

Recent developments on ultrasound-assisted organic synthesis in aqueous medium•

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(Received 17 February, Revised 27 April, Accepted 3 May 2017)

Abstract: In the recent past, a number of methods were reported on the application of ultrasound in organic reactions for the synthesis of diverse organic scaffolds. On the other hand, as far as green chemistry is concerned, water is the safest of all solvents. Thus, a "strong collaboration" between ultrasonic irradiation and aqueous medium holds the key to the development of an environmentally sustainable protocol. The present review summarizes the latest developments in ultrasound-assisted and water-mediated organic synthesis reported to date.

Keywords: ultrasonic irradiation; sonochemistry; aqueous medium; organic synthesis; heterocyclic chemistry; sustainable chemistry.

CONTENTS

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• This work is dedicated to Prof. Dr. György Keglevich, Budapest University of Technology and Economics, Hungary.
https://doi.org/10.2298/JSC170217057B

1. INTRODUCTION

Sustainable organic methodologies are in high demand in the chemical industry to produce various organic scaffolds in the form of fine chemicals, medicinal and pharmaceutical agents, agrochemicals, and many others.[1–4] The people of the twenty-first century are well aware about the side effects of the hazardous substances used or generated in chemical processes. It is high time to take the steps necessary to protect our "Mother Nature" from ever increasing chemical pollution associated with synthetic organic processes. Thus, today's methodologists are trying to make their protocols more environmentally friendly and sus-

tainable by avoiding the extensive use of harsh reaction conditions, hazardous reagents and solvents.[5–8] As a result, the last decade has seen a tremendous outburst in the modification of chemical processes to make them 'sustainable' for the betterment of the environment.

The involvement of ultrasound in organic synthesis sometimes fulfills this goal. Ultrasound irradiated reactions are much more advantageous over traditional thermal methods in terms of reaction rates, yields, purity of the products, product selectivity, *etc*.[9–14] Ultrasonic waves cause molecules to oscillate around their mean position, which increases the average distance between the molecules.[15] Thus, under suitable conditions, a huge number of cavitation bubbles are generated in reaction mixture, which grow rapidly and subsequently undergo vigorous collapses that results in the formation of micro-jets that can produce fine emulsions between the reactants.[16] In addition, the local temperature within the reaction mixture is also increased by the violent collapse of the cavitation bubbles, which eventually lead to the activation energy barrier being crossed.[17] Sometimes the application of ultrasound can avoid the use of catalysts in organic reactions.[18–22] On the other hand, last decades have shown a tremendous outburst of chemical reactions performed under "in-water" conditions[23] to make them "sustainable" for the betterment of the environment. Scientists are choosing water as a solvent not only because of its environmental friendliness but also because it is cheap, non-flammable, and abundantly available. Water as a solvent activates the functional groups by forming hydrogen bonds. Due to high surface tension and hydrophobic nature,[24] the reactants in aqueous medium are bound to form aggregates in order to decrease the exposed organic surface area,[25–27] which increases the rate of the reaction. Thus, a "strong collaboration" between ultrasonic irradiation and aqueous media holds the key to the development of an environmentally sustainable protocol.

Such beneficial features in terms of sustainability have motivated organic chemists to explore aqueous-mediated organic synthesis employing ultrasonic irradiation in more depth and as a result, in the recent past, there were immense applications of ultrasound in organic reactions for the synthesis of various heterocyclic as well as non-heterocyclic scaffolds in aqueous media. The present review summarizes the latest developments in ultrasound-assisted and water-mediated organic synthesis reported to date.

2. ULTRASOUND-ASSISTED SYNTHESIS OF HETEROCYCLES IN AQUEOUS MEDIUM

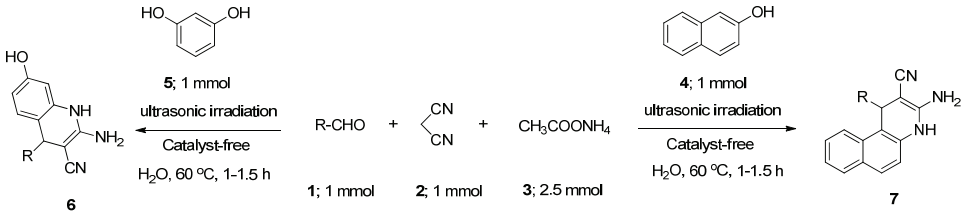
More than half of the known organic compounds are containing heterocyclic ring.[28–33] These spectacular classes of compounds are important because they represent a "privileged" structural subunits well distributed in naturally occurring compounds with immense biological activities such as anticancer,[34] cytotocxic,[35] anti-HIV,[36] anti-malarial,[37] anti-inflammatory,[38] anti-microbial[39] and many

more.[40,41] To realize the importance of heterocycles, in the recent past, a number of synthetic protocols are being reported in the literature almost in every month. But in generally these protocols are not satisfying the sustainability issues as in many occasions toxic solvents, harsh reaction conditions are being used. Therefore, now-a-days it is the thrusting area of research to synthesize biologically relevant heterocycles using sustainable pathways.

2.1. Ultrasound-assisted synthesis of N*-heterocycles in aqueous medium*

2.1.1. Synthesis of dihydroquinolines

Quinolines are very common naturally occurring compounds.[42] Many synthetic scaffolds containing the quinoline moiety possess significant biological efficacies that include anti-fungal,[43] anti-malarial,[44] anti-protozoal,[45] analgesic[46] activity. Therefore a large numbers of methods employing various catalysts are already available in the literature for the synthesis of quinolines and dihydroquinolines.[47–49] Although these reported methods have their own merits, they still suffer from some disadvantage, such as the use of toxic solvents, drastic reaction conditions, long reaction times, *etc*. Pagadala *et al.* (Scheme 1)[50] developed a simple, efficient, ultrasound-assisted, catalyst-free, one-pot, four-component protocol for the synthesis of a series of biologically promising dihydroquinoline derivatives (**6** and **7**) from the reactions of various aldehydes (**1**), malononitrile (**2**), ammonium acetate (**3**) and 2-naphthol (**4**)/resorcinol (**5**) in aqueous medium at 60 °C. The ultrasonic-irradiated method was found to be superior as compared to the conventional method with respect to reaction time and yields.

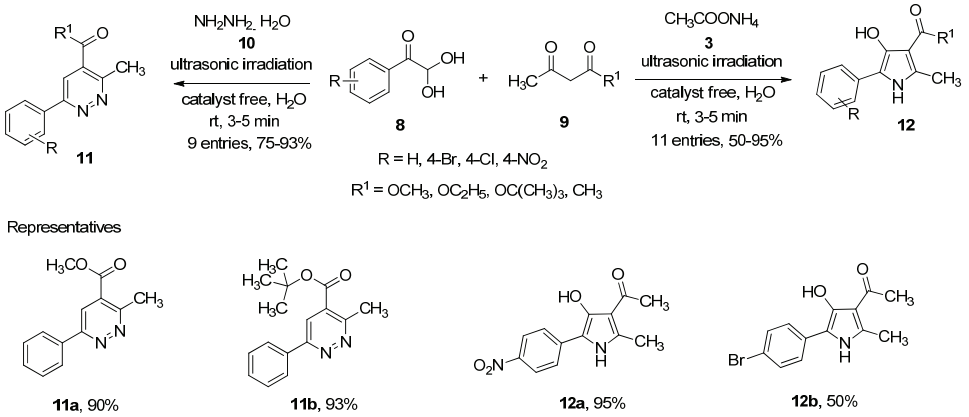


Entry	Scaffolds	R	Conventional stirring		With sonication	
			Time, h	Yield, %	Time, h	Yield, %
1	**6a**	C_6H_5	4.0	80	1.0	96
2	**6b**	4-Br–C_6H_4	3.5	74	1.5	94
3	**6c**	4-OH–C_6H_4	4.5	70	1.0	90
4	**6d**	4-Cl–C_6H_4	3.0	76	1.0	92
5	**7a**	C_6H_5	3.0	82	1.0	97
6	**7b**	4-Br–C_6H_4	4.0	80	1.0	95
7	**7c**	2-Cl–C_6H_4	4.0	70	1.5	90
8	**7d**	4-Cl–C_6H_4	3.5	78	1.0	94
9	**7e**	4-OH–C_6H_4	4.0	74	1.5	90

Scheme 1. Ultrasound-promoted catalyst-free synthesis of dihydroquinolines in water.

2.1.2. Synthesis of pyrroles and pyridazines

Pyrroles and pyridazines are very common in natural products, pharmaceuticals, and various bioactive molecules.[45,46] Thus, a number of methods are already available in the literature for the synthesis of these important classes of heterocyclics employing various reaction conditions.[51–56] However these reported methods suffer due to the use of hazardous solvents, harsh reaction conditions, long reaction times, *etc*. In this context, a facile, straightforward, efficient, ultrasound-assisted, catalyst-free protocol was developed by Eftekhari-Sis and Vahdati-Khajeh (Scheme 2)[57] for the synthesis of 6-aryl-3-methylpyridazine-4-carboxylic acid esters (**11**) and 5-aryl-4-hydroxy-2-methyl-1*H*-pyrrole-3--carboxylic acid esters (**12**) *via* three-component reactions of arylglyoxal hydrates (**8**), *β*-dicarbonyl compounds (**9**) and hydrazine hydrate (**10**) or ammonium acetate (**3**), respectively, in aqueous media at room temperature.

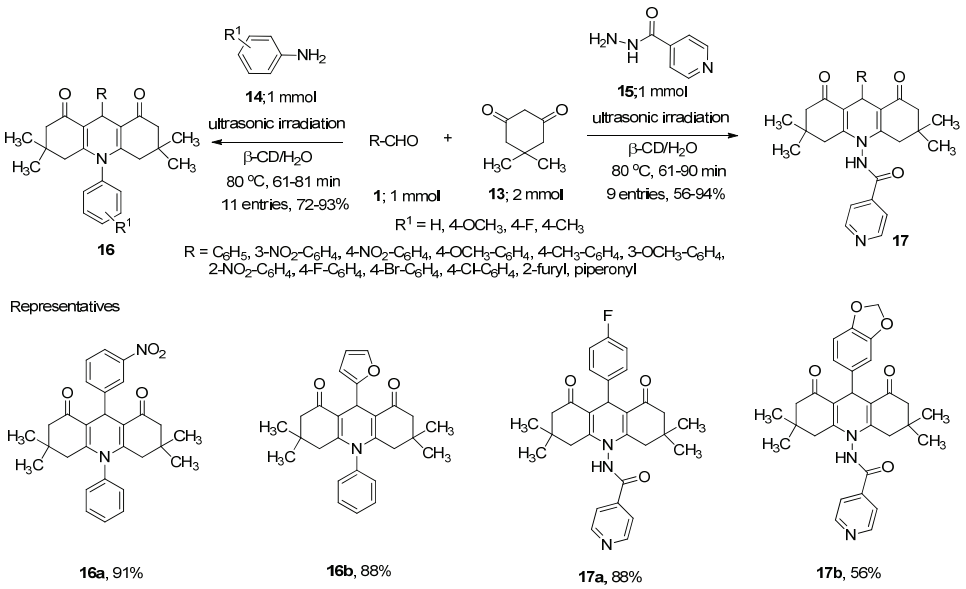


Scheme 2. Ultrasound promoted, catalyst-free synthesis of pyrroles and pyridazines in water.

2.1.3. Synthesis of *N*-substituted 1,8-dioxodecahydroacridines

N-Aryl-1,8-dioxodecahydroacridine moieties are important in drug design and discovery as they possesses potential pharmacological activities, such as antimicrobial, anticancer and enzyme inhibitory properties.[58–63] A large number of methods are already available in the literature for the synthesis of these important scaffolds employing various catalysts such as 1-methylimidazolium trifluoroacetate,[58] silica-supported *N*-propylsulfamic acid,[59] amberlyst-15,[60] ZnO nanoparticles,[61] LiBr[62] and NH_4Cl.[63] Although these available methods possess notable merits they still suffer from some drawbacks, such as, use of organic solvents, longer reaction times, *etc*. In this context, Chate *et al*. (Scheme 3)[64] developed a practical ultrasound-assisted, one-pot method for the synthesis of *N*-substituted 1,8-dioxodecahydroacridines (**16** and **17**) *via* three-component reactions between aldehydes (**1**), dimedone (**13**) and isoniazid (**15**)/aromatic amines

(**14**) in the presence of *β*-cyclodextrin as a supramolecular, biodegradable, reusable catalyst in aqueous medium. Use of organic solvents such as CH_3CN, THF, DMF and EtOH gave inferior results in terms of both reaction time and yield.



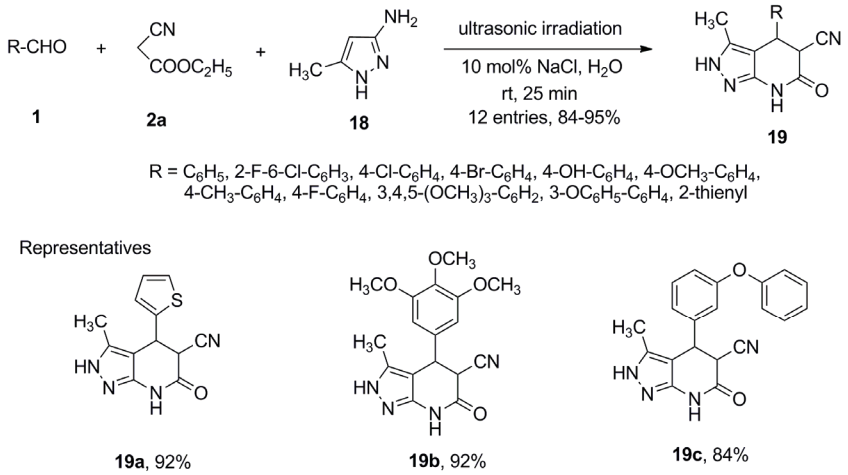
Scheme 3. Ultrasound-promoted synthesis of *N*-substituted 1,8-dioxodecahydroacridines in water.

2.1.4. Synthesis of pyrazolo[3,4-*b*]pyridine derivatives

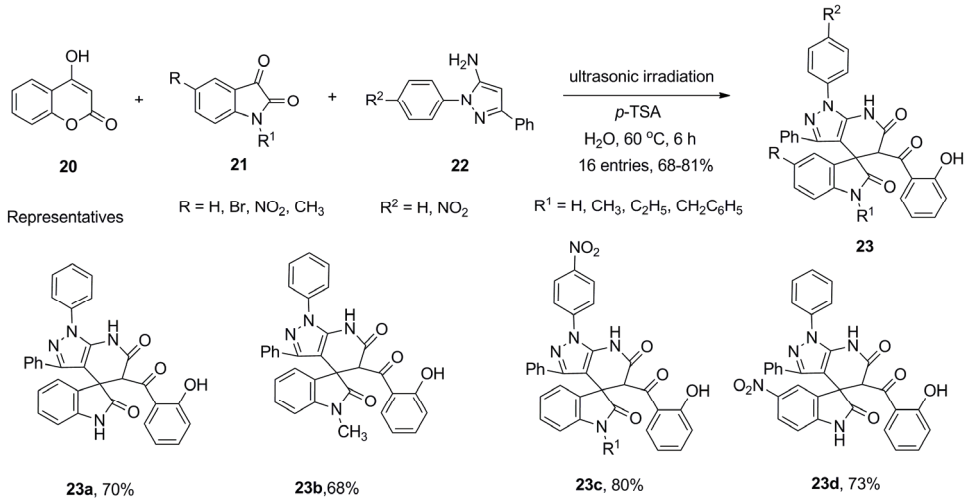
Many anxiolytic drugs, such as cartazolate, etazolate and tracazolate, contain the pyrazolopyridine moiety.[65–68] A simple, ultrasound-assisted, high-yielding, environmentally benign protocol was developed for the synthesis of pyrazolo-[3,4-*b*]pyridine derivatives (**19**) *via* one-pot, three-component reactions of aldehydes (**1**), ethyl cyanoacetate (**2a**), and 3-amino-5-methylpyrazole (**18**) using sodium chloride as the catalyst in aqueous media at room temperature (Scheme 4).[69] The ultrasonic-irradiated method was found to be advantageous as compared to the conventional heating method. The use of organic solvents, such as acetonitrile, toluene, dichloromethane, tetrahydrofuran, ethanol and methanol, gave inferior results in terms of both reaction times and yields.

2.1.5. Synthesis of spiro[indoline-3,4′-pyrazolo[3,4-b]pyridine]-2,6′(1′H)--diones

A simple, facile, environmentally benign, aqueous-mediated, ultrasound--assisted, three-component protocol was developed for the synthesis of medicinally important spiro[indoline-3,4′-pyrazolo[3,4-*b*]pyridine]-2,6′(1′*H*)-diones (**23**) from the reaction of 4-hydroxycoumarin (**20**), isatins (**21**) and 1*H*-pyrazol-5-amines (**22**) using *p*-toluenesulfonic acid (*p*-TSA) as catalyst at 60 °C (Scheme 5).[70]



Scheme 4. Ultrasound-promoted synthesis of pyrazolo[3,4-*b*]pyridine derivatives in water.

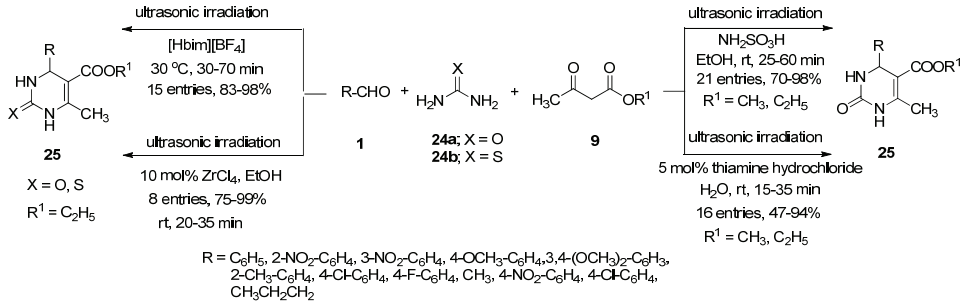


Scheme 5. Ultrasound-promoted synthesis of spiro[indoline-3,4′-pyrazolo[3,4-*b*]pyridine]--2,6′(1′*H*)-diones in water.

2.1.6. Synthesis of highly substituted pyrimidine-5-carboxylic acid derivatives

Dihydropyrimidinones (**25**) possess immense biological efficacies, such as antiviral, antibacterial, antihypertensive, antitumor and many more activities.[71] In 1893, Biginelli[72] first reported the synthesis of dihydropyrimidinones (**25**) with only 20–50 % yields. In 2003, Li *et al.* (Scheme 6)[73] employed ultrasound for the synthesis of bioactive dihydropyrimidinones (**25**) by one-pot, three-component reactions between various aldehydes (**1**), urea (**24a**) and ethyl acetoace-

tate (**9**) using sulfamic acid as catalyst in ethanol at room temperature. Later, Gholap *et al*. (Scheme 6)[74] used 1-butylimidazolium tetrafluoroborate [Hbim][BF_4] as an efficient ionic liquid for the synthesis of dihydropyrimidinones (**25**) under the influence of ultrasound at ambient temperature. Zirconium chloride (10 mol %) was used by Kumar and Parmar (Scheme 6)[75] as catalyst in ethanol for the same batch of the reactions under ultrasonic irradiation at room temperature.

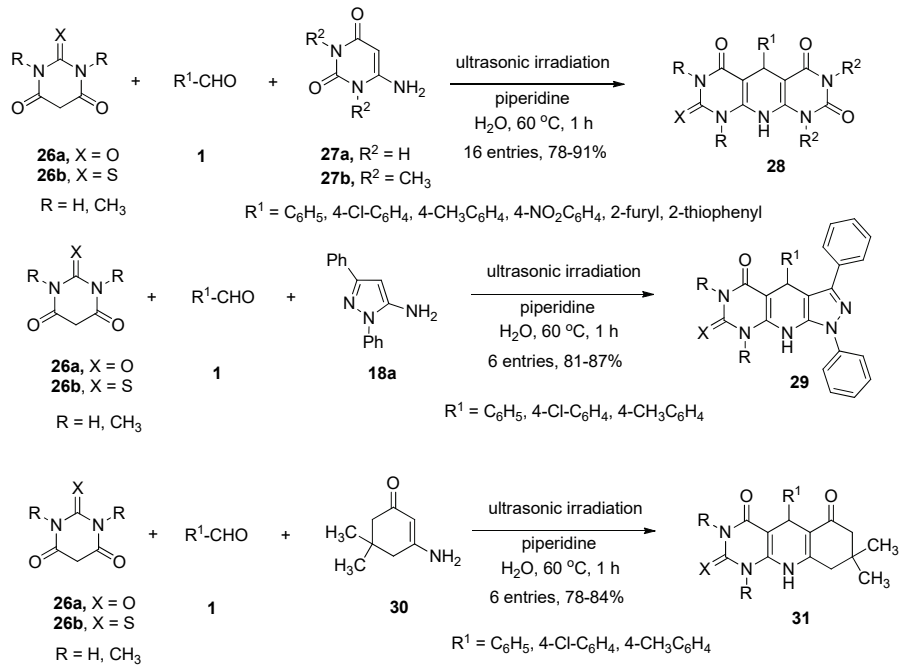


Scheme 6. Ultrasound-promoted synthesis of pyrimidine-5-carboxylic acid derivatives.

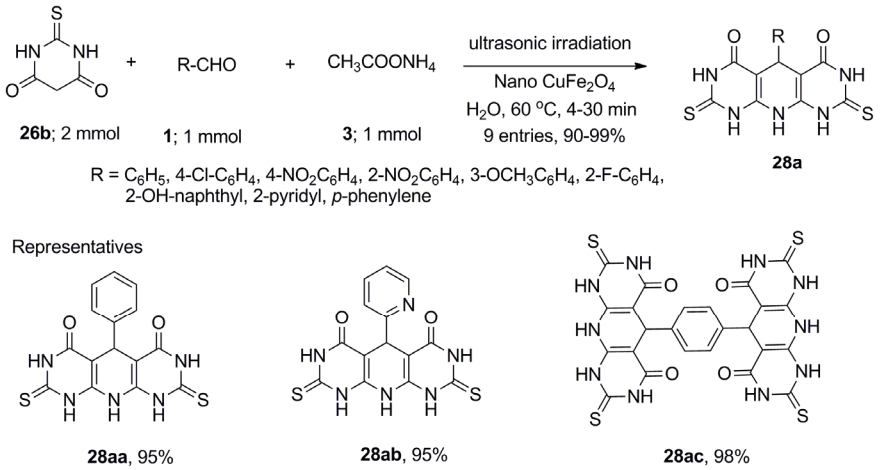
Recently, in 2010, Mandhane *et al*. (Scheme 6)[76] developed a simple, efficient, environmentally friendly, aqueous-mediated, ultrasound-assisted method for the synthesis of dihydropyrimidinones (**25**) at room temperature using a catalytic amount of thiamine hydrochloride as an efficient organo-catalyst.

2.1.7. Synthesis of pyrimidine fused heterocycles

Heterocycles containing a pyrimidine moiety are very common in naturally occurring compounds. They possess numerous biological efficacies that include antitumor,[77] antiviral[78] and antioxidant[79] activity. Owing to the attractive pharmacological properties of these scaffolds, several methods are available for the synthesis of various pyrimidine fused heterocycles.[80–86] Again these methods suffer disadvantages due to the use of hazardous solvents, long reaction times and harsh reaction condition. Recently, in 2010, Mosslemin and Nateghi (Scheme 7)[87] synthesized a series of pyrimidine annulated fused heterocycles (**28**, **29** and **31**) in high yields *via* an ultrasound-assisted, one-pot, three-component condensation reaction of barbituric acid (**26a**) or 2-thiobarbituric acid (**26b**), aldehydes (**1**) and a series of enamines (**27a**, **27b**, **18a** and **30**) using piperidine as catalyst in water at 60 °C. Another simple, convenient, environmentally friendly, ultrasound-assisted, facile one-pot, four-component protocol was developed by Naeimi and Didar (Scheme 8)[88] for the synthesis of pyrido[2,3-*d*:6,5-*d'*]dipyrimidines (**28a**) from the reactions of aldehydes (**1**), 2-thiobarbituric acid (**26b**) and ammonium acetate (**3**) using magnetically separable nano copper ferrite as a heterogeneous catalyst in water.



Scheme 7. Ultrasound-promoted synthesis of pyrimidine fused heterocycles in water.

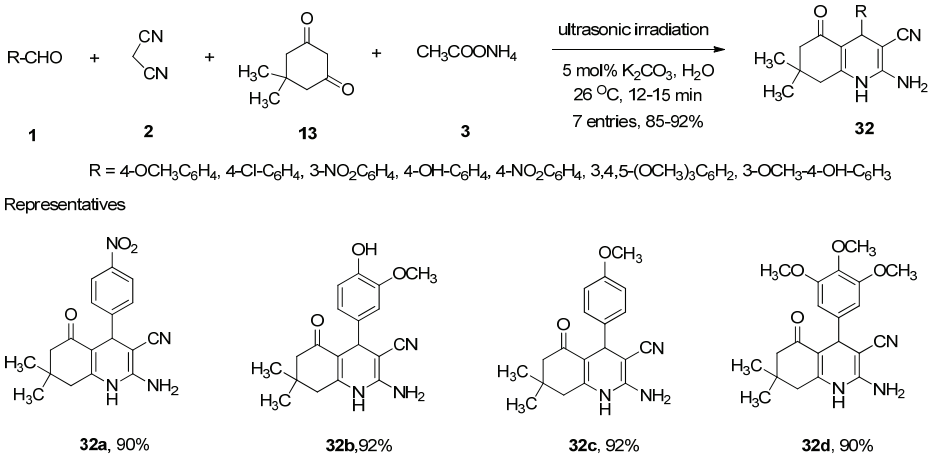


Scheme 8. Ultrasound promoted synthesis of pyrido[2,3-*d*:6,5-*d*]dipyrimidines in water.

2.1.8. Synthesis of 2-amino-1,4,5,6,7,8-hexahydroquinoline-3-carbonitriles

Quinoline and its derivatives are an important class of pharmaceutical compounds that occur predominately in natural products, exhibit a broad spectrum of biological activities, and act as antivirals,[89,90] antiherpetics,[91] antidepressants,[92] anti-

-oxidants,[93] and anti-inflammatory,[94] antiproliferation[95] and anticancer[96] agents. In particular, hexahydroquinolines also possess antioxidant[97] and cytotoxic[98] activities. A series of highly substituted 2-amino-1,4,5,6,7,8-hexahydroquinolines-3-carbonitriles (**32**) were synthesized *via* ultrasound-assisted, one-pot, four-component condensations of aromatic aldehydes (**1**), dimedone (**13**), malononitrile (**2**) and ammonium acetate (**3**) using K_2CO_3 as a catalyst in aqueous medium at ambient temperature (Scheme 9).[99]



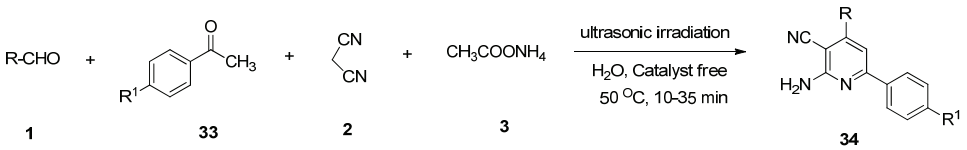
Scheme 9. Ultrasound-promoted synthesis of 2-amino-1,4,5,6,7,8-hexahydroquinoline-3--carbonitriles in water.

2.1.9. Synthesis of 2-amino-4,6-diphenylnicotinonitriles

Safari *et al*. (Scheme 10)[100] reported a simple ultrasound-promoted catalyst--free, high yielding, convenient, environmentally benign approach to the synthesis of 2-amino-4,6-diphenylnicotinonitriles (**34**) *via* four-component reactions of aromatic aldehydes (**1**), malononitrile (**2**), acetophenones (**33**) and ammonium acetate (**3**) in water. The ultrasonic-irradiated method was found to be advantageous as compared to the conventional stirring method at 50 °C. The use of organic solvents, such as ethanol, methanol, acetonitrile, dichloromethane or tetrahydrofuran, gave inferior results in terms of both reaction times and yields.

2.1.10. Synthesis of tetrahydrodipyrazolopyridines

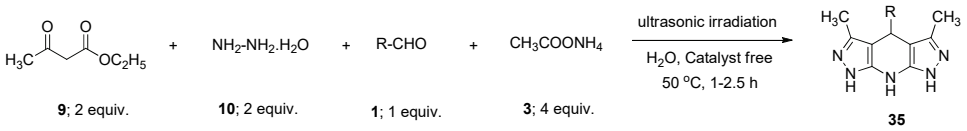
Shabalala *et al*. (Scheme 11)[101] demonstrated a simple, convenient, ultrasound-promoted, catalyst-free, environmentally benign protocol for the one-pot, four-component syntheses of tetrahydrodipyrazolopyridines (**35**) from the sequential reactions of ethyl acetoacetate (**9**), hydrazine hydrate (**10**), aromatic aldehydes (**1**) and ammonium acetate (**3**) in water at 50 °C. The ultrasonic-irradiated method was found to be advantageous when compared to the conventional method with respect to reaction times and yields.



1 **33** **2** **3** **34**

Entry	R	R^1	With sonication		Conventional stirring	
			Time, min	Yield, %	Time, h	Yield, %
1	H	H	25	91	1.5	80
2	H	4-CH_3	20	89	2.5	76
3	OH	H	25	93	1.5	74
4	OH	4-OCH_3	23	85	2.5	68
5	OH	4-Cl	18	93	1.2	76
6	OH	3-NO_2	30	95	1.2	77
7	H	4-Cl	20	95	1.0	85
8	H	4-OCH_3	20	87	3.0	70
9	H	4-Br	22	98	1.5	85
10	H	4-pyridyl	15	95	1.2	75
11	OH	2-OCH_3	30	75	3.0	0
12	OH	2-F	35	80	3.0	0
13	OH	2-Cl	35	78	3.0	0
14	OH	3-F	15	96	1.5	82
15	OH	3-OCH_3	17	90	2.0	80
16	OH	3-Br	15	90	1.5	76
17	OH	3-Cl	15	95	1.5	82
18	OH	3-OH	20	97	2.0	82
19	OH	4-Br	18	97	1.5	84
20	OH	4-NO_2	10	99	1.5	85
21	OH	4-pyridyl	15	98	2.2	77
22	OH	4-F	15	97	1.3	82
23	OH	2-furyl	18	90	2.5	73
24	OH	2-thienyl	15	93	2.3	76

Scheme 10. Ultrasound-promoted synthesis of 2-amino-4,6-diphenylnicotinonitriles in water.



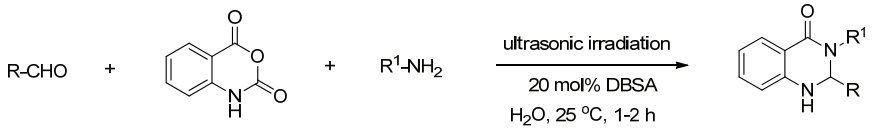
9; 2 equiv. **10**; 2 equiv. **1**; 1 equiv. **3**; 4 equiv. **35**

Entry	R	Scaffolds	Without sonication		With sonication	
			Time, h	Yield, %	Time, h	Yield, %
1	C_6H_5	**35a**	7.0	64	2.0	95
2	4-OCH_3–C_6H_4	**35b**	6.0	60	2.5	96
3	4-Br–C_6H_4	**35c**	6.0	58	2.0	94
4	4-$N(CH_3)_2$–C_6H_4	**35d**	6.5	61	2.5	92
5	4-OH–C_6H_4	**35e**	6.0	63	2.5	90
6	2-Br–C_6H_4	**35f**	6.0	55	2.0	92
7	2-OCH_3–C_6H_4	**35g**	5.5	60	1.5	91
8	2-Cl–C_6H_4	**35h**	6.0	64	2.0	94
9	2-NO_2–C_6H_4	**35i**	6.5	60	2.5	92

Scheme 11. Ultrasound-promoted synthesis of tetrahydrodipyrazolopyridines in aqueous medium.

2.1.11. Synthesis of 2,3-disubstituted 2,3-dihydroquinazolin-4(1H)-ones

A series of 2,3-disubstituted 2,3-dihydroquinazolin-4(1*H*)-one derivatives (**37**) were synthesized with excellent yields in the one-pot, three-component condensations between aromatic aldehydes (**1**), isatoic anhydride (**36**), and various amines (**14**) in the presence of a catalytic amount of *p*-dodecylbenzenesulfonic acid (DBSA) in water under the influence of ultrasound irradiation at 40––42 °C (Scheme 12).[102] The ultrasonic-irradiated method was found to be superior to the conventional method with respect to reaction times and yields.



1 **36** **14** **37**

Entry	R	R^1	Conventional stirring		With sonication	
			Time, h	Yield, %	Time, h	Yield, %
1	C_6H_5	C_6H_5	3	68	1.5	83
2	4-Cl–C_6H_4	C_6H_5	5	71	2	85
3	4-CH_3–C_6H_4	C_6H_5	4	73	2	88
4	4-NO_2–C_6H_4	C_6H_5	5	67	2	84
5	4-CH_3O–C_6H_4	C_6H_5	4	68	1.5	90
6	2-Cl–C_6H_4	C_6H_5	4	63	2	80
7	3,4-(OCH_2O)–C_6H_2	C_6H_5	3	76	1.5	89
8	C_6H_5	CH_3	2	70	1	86
9	4-Cl–C_6H_4	CH_3	3	72	1.5	88
10	4-CH_3–OC_6H_4	CH_3	3	68	1.5	89
11	4-CH_3–C_6H_4	CH_3	2	73	1	91
12	2-Cl–C_6H_4	CH_3	3	70	1.5	82
13	4-NO_2–C_6H_4	CH_3	3	67	1.5	85
14	2-NO_2–C_6H_4	CH_3	3	62	1.5	80
.15	2-CH_3O–C_6H_4	CH_3	2.5	70	1.5	85
16	3,4-(OCH_2O)–C_6H_2	CH_3	2	75	1	92
17	C_6H_5	4-CH_3–C_6H_4	2	73	1	91
18	C_6H_5	4-CH_3–C_6H_4	2	71	1	89
19	C_6H_5	C_2H_5	2	68	1	83

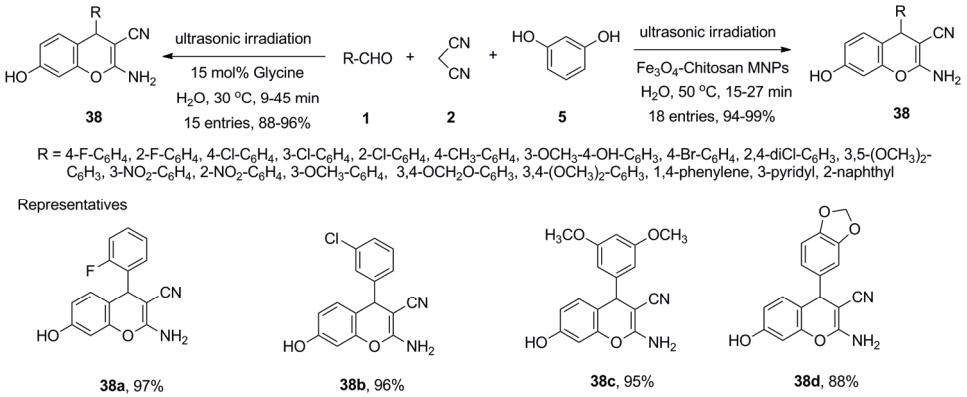
Scheme 12. Ultrasound-promoted synthesis of 2,3-disubstituted 2,3-dihydroquinazolin-4(1*H*)--ones in water.

2.2. Ultrasound-assisted synthesis of O*-heterocycles in aqueous medium*

2.2.1. Synthesis of 2-amino-4*H*-chromenes

2-Amino-4*H*-chromenes (**38**) belong to the class of privileged medicinal scaffolds having highly pronounced anticoagulant, diuretic, spasmolitic and anti-anaphylactic activities.[103–105] After realizing the importance of this moiety, Safari and Javadian (Scheme 13)[106] demonstrated an efficient, environmentally benign,

aqueous-mediated, ultrasound-assisted, one-pot, three-component condensation of aldehydes (**1**), malononitrile (**2**) and resorcinol (**5**) in the presence of a catalytic amount of magnetically-separable Fe_3O_4–chitosan nanoparticles to afford the corresponding 2-amino-4*H*-chromenes (**38**) in high yields. Later, Datta and Pasha (Scheme 13)[107] also developed an ultrasound-assisted protocol for the synthesis of 2-amino-4*H*-chromenes (**38**) by the same sequence of reactions using glycine as an efficient organo-catalyst in aqueous medium at ambient temperature.



Scheme 13. Ultrasound-promoted synthesis of 2-amino-4*H*-chromenes in aqueous media.

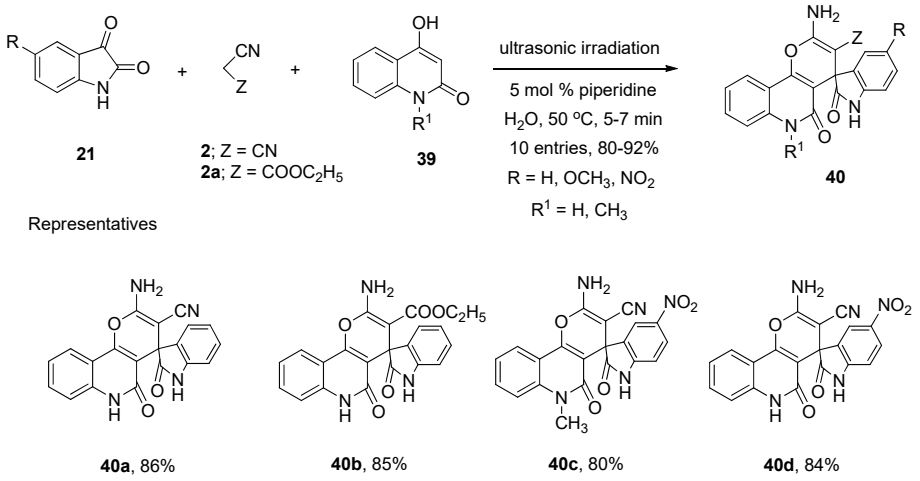
2.2.2. Synthesis of spiro[oxindoles]

A simple, efficient, environmentally benign, ultrasound-assisted, one-pot three-component protocol was developed for the synthesis of a series of medicinally important spiro[indoline-3,4′-pyrano[3,2-*c*]quinolines (**40**) in the reactions between isatins (**21**), malononitrile (**2**) or ethylcyanoacetate (**2a**) and 4-hydroxy-2*H*-quinolin-2-one (**39**) using a small amount of piperidine as a base catalyst in aqueous medium at 50 °C (Scheme 14).[108] Another novel, efficient, convenient, ultrasound-assisted, environmentally-friendly protocol was developed by Dandia *et al*. (Scheme 15)[109] for the synthesis of a series of biologically relevant spiro[chromene-4,3′-indolines] (**42**) and spiro[indoline-3,4′-pyrano[2,3-*c*]pyrazoles] (**43**) *via* one-pot, three-component reactions between substituted isatins (**21**), malononitrile (**2**) or ethyl cyanoacetate (**2a**), and dimedone (**13**) or 3-methyl-1--phenyl-2-pyrazolin-5-one (**41**) using sodium chloride as a catalyst in water.

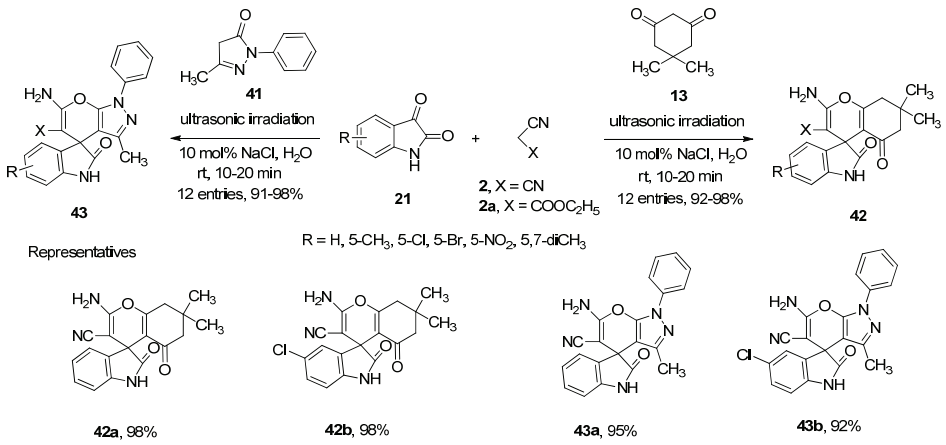
2.2.3. Synthesis of pyrano-chromenes and benzopyrano-chromenes

A simple, convenient, ultrasound-assisted, one-pot, three-component protocol was developed by Gohil *et al*. (Scheme 16)[110] for the synthesis of a series of biologically relevant 2-amino-3-cyano-pyrano[4,3-*b*]pyrans (**47** and **48**) and 2--amino-3-cyano-pyrano[3,2-*c*]chromenes (**49**,**50**) from the reaction between 2--(triazolium(tetrazolium-amino)quinoline-3-carbaldehydes (**44**/**45**), malononitrile

(**2**)/methyl cyanoacetate (**2a**) and 4-hydroxy-6-methyl-2*H*-pyran-2-one (**46**) or 4--hydroxy-coumarin (**20**), respectively, in the presence of a catalytic amount of L-proline in water at 50 °C.



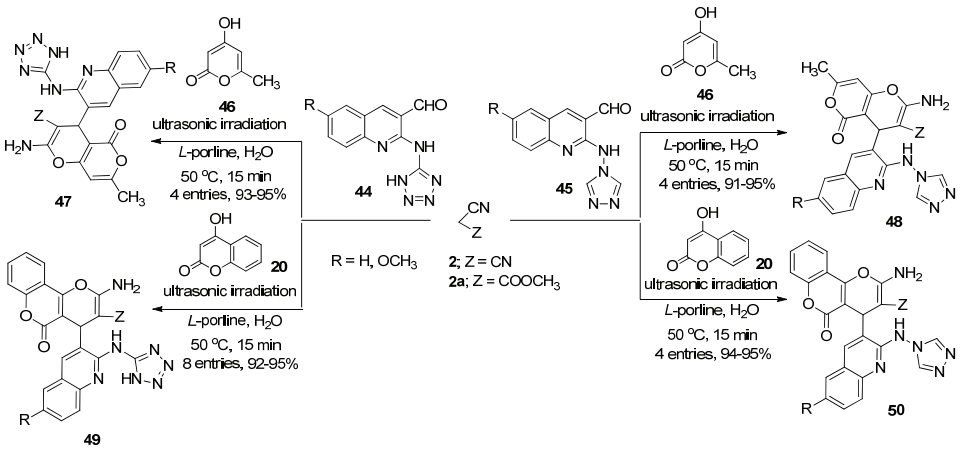
Scheme 14. Ultrasound-promoted synthesis of spiro[indoline-3,4′-pyrano[3,2-*c*]quinolone] in aqueous media.



Scheme 15. Ultrasound-promoted synthesis of spiro[chromene-4,3′-indolines] and spiro[indoline-3,4′-pyrano[2,3-*c*]pyrazoles] in aqueous media.

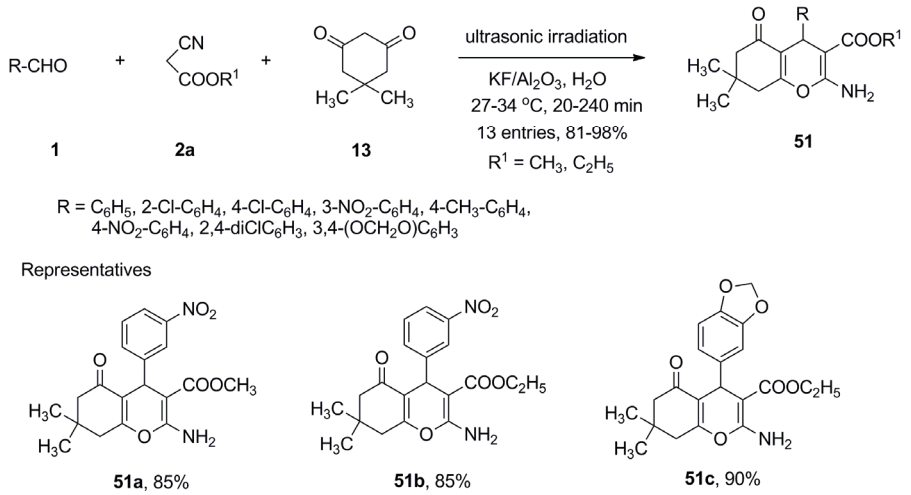
2.2.4. Synthesis of 2-aminobenzo[*b*]pyran

Recently, a series of synthetic 2-amino-4*H*-pyrans was evaluated to possess potent anticancer,[111,112] antibacterial, and antifungal[113,114] properties. Such a handful of diverse applications of 2-amino-4*H*-pyrans has resulted in a good number of synthesis methods being available in the literature using various catalysts



Scheme 16. Ultrasound-promoted synthesis of pyrano[4,3-*b*]pyrans and pyrano[3,2-*c*]chromenes in aqueous media.

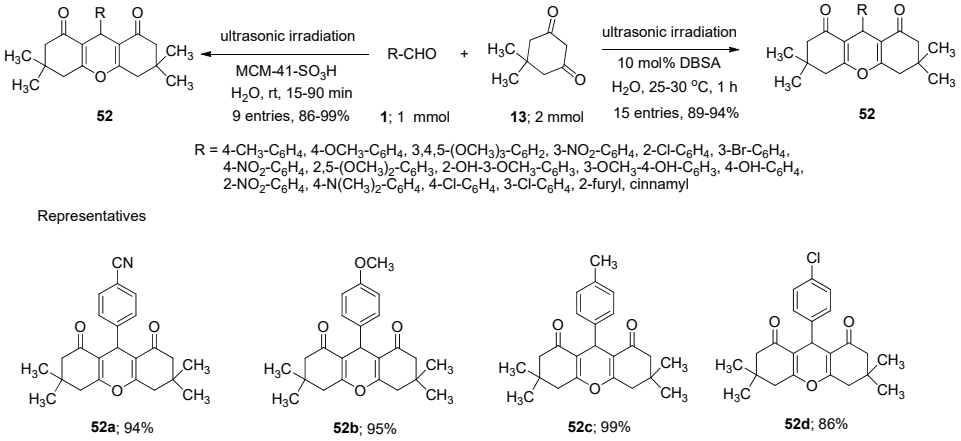
that include urea,[115] sodium formate,[116] trisodium citrate,[117] TBAB,[118] DBU,[119] nano ZnO[120] and MgO.[121] Although these reported protocols find certain merits of their own, they still suffer from a number of demerits such as the use of toxic organic solvents, long reaction times, heating and harsh reaction conditions. A simple, straightforward, environmentally benign, ultrasound-assisted, one-pot, three-component protocol was reported for the synthesis of a series of bioactive 2-aminobenzo[*b*]pyrans (**51**) with excellent yields from the reaction of various aromatic aldehydes (**1**), cyanoacetic esters (**2a**) and dimedone (**13**) using KF/basic Al_2O_3 as catalyst in water at room temperature (Scheme 17).[122]



Scheme 17. Ultrasound-promoted synthesis of 2-aminobenzo[*b*]pyrans in aqueous media.

2.2.5. Synthesis of 1,8-dioxooctahydroxanthenes

In 2006, Jin *et al*. (Scheme 18)[123] demonstrated a simple, facile, environmentally benign, aqueous-mediated, ultrasound-assisted protocol for the synthesis of bioactive 3,3,6,6-tetramethyl-9-aryl-1,8-dioxooctahydroxanthenes (**52**) *via* a pseudo three-component reaction between various aldehydes (**1**; 1 equiv.) and dimedone (**13**; 2 equiv.) using 10 mol % *p*-dodecylbenzenesulfonic acid (DBSA) as catalyst at ambient temperature. Later, in 2010, Rostamizadeh *et al*. (Scheme 18)[124] developed another efficient ultrasound-irradiated method for the same series of reactions using MCM-41-SO_3H as catalyst in water at room temperature.



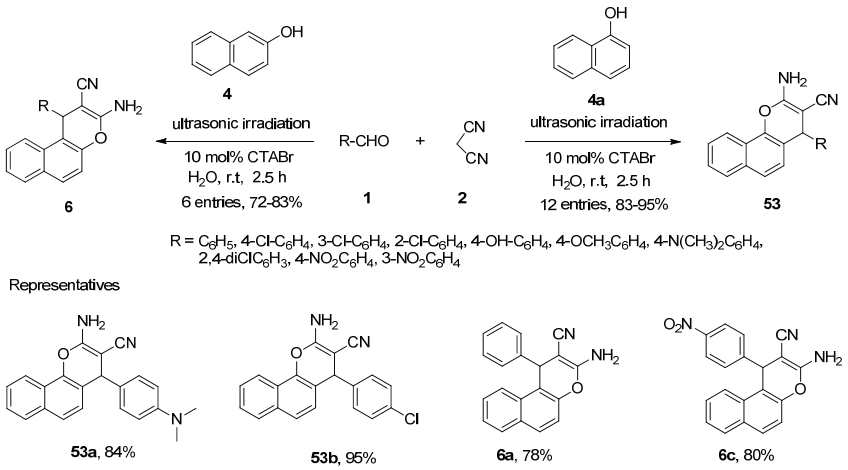
Scheme 18. Ultrasound-promoted synthesis of 1,8-dioxooctahydroxanthenes in aqueous media.

2.2.6. Synthesis of benzo[*f* or *h*]chromene

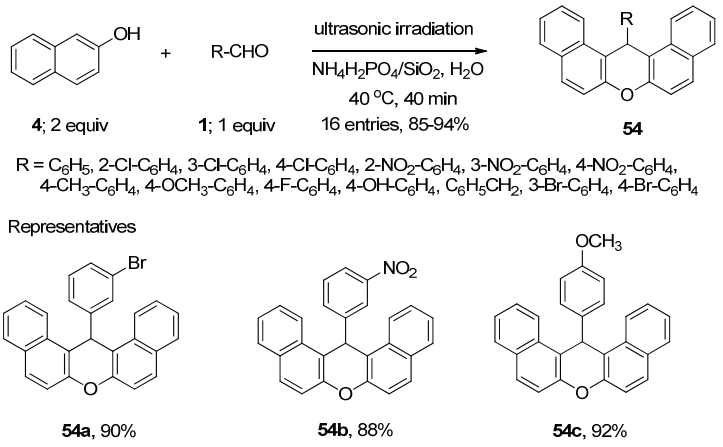
Jin *et al*. (Scheme 19)[125] described a simple, efficient, mild, water-mediated, ultrasound-assisted, practical procedure for the synthesis of 2-amino-3-cyanobenzo[*f*]chromenes (**6**) as well as benzo[*h*]chromenes (**53**) by one-pot, three-component reactions between aromatic aldehydes (**1**), malononitrile (**2**) and 2-napthol (**4**) or 1-naphthol (**4a**), respectively, using a catalytic amount of CTABr as catalyst at room temperature.

2.2.7. Synthesis of various aryl-14*H*-dibenzo[*a*,*j*]xanthenes

A simple, facile, environmentally benign, ultrasound-assisted protocol was reported for the synthesis of various biologically promising benzo[*a*,*j*]xanthenes (**54**) *via* pseudo three-component reactions between various aldehydes (**1**; 1 equiv.) and 2-naphthol (**4**; 2 equiv.) using silica-supported ammonium dihydrogen phosphate ($NH_4H_2PO_4/SiO_2$) as catalyst in water at 40 °C (Scheme 20).[126]



Scheme 19. Ultrasound-promoted synthesis of 2-aminobenzochromenes in water.



Scheme 20. Ultrasound-promoted synthesis of various benzo[*a*,*j*]xanthenes in water.

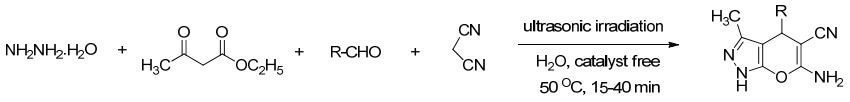
2.2.8. Synthesis of dihydropyrano[2,3-*c*]pyrazoles

A simple, mild, efficient, sustainable, catalyst-free, water-mediated protocol was developed by Zou *et al*. (Scheme 21)[127] for the synthesis of dihydropyrano[2,3-*c*]pyrazoles (**55**) *via* one-pot, four-component reactions of hydrazine hydrate (**10**), ethyl acetoacetate (**9**), various aldehydes (**1**), and malononitrile (**2**) under ultrasound irradiation at 50 °C. Ultrasonic irradiation enhanced the rate of the reaction as compared to the conventional stirring method.

2.2.9. Synthesis of 2-aminopyrano[3,2-*b*]pyrans

A simple, mild, ultrasound-assisted, catalyst-free, green and convenient approach to the synthesis of biologically important 2-amino-3-cyano-pyrano[3,2--*b*]pyrans (**57**) *via* one-pot, three-component reactions between various aldehydes

(**1**), malononitrile (**2**) and kojic acid (**56**) in water was described by Banitaba *et al*. (Scheme 22).[128] The ultrasonic-irradiated method was found to be advantageous as compared to the conventional stirring method.



10 9 1 2 55

Entry	R	With sonication		Conventional stirring	
		Time, min	Yield, %	Time, h	Yield, %
1	4-CH_3–C_6H_4	30	92	5	85
2	C_6H_5	30	88	3	72
3	4-Cl–C_6H_4	15	95	1	83
4	3-Cl–C_6H_4	20	93	2	80
5	2-Cl–C_6H_4	30	93	2	81
6	4-F–C_6H_4	15	89	2.5	85
7	4-Br–C_6H_4	15	90	2	83
8	4-OH–C_6H_4	30	80	3	75
9	4-NO_2–C_6H_4	30	85	1.5	78
10	3,4-OCH_2O–C_6H_4	40	82	4	72
11	3,4-$(Cl)_2$–C_6H_3	15	91	1	86
12	2-Thienyl	35	79	2	70
13	2-Pyridyl	40	87	2	72

Scheme 21. Ultrasound-promoted synthesis of dihydropyrano[2,3-*c*]pyrazoles in water.

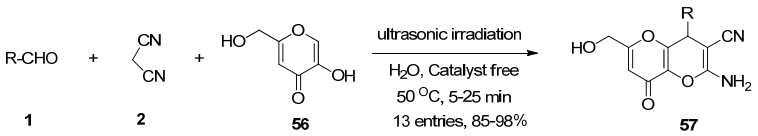
2.2.10. Synthesis of 2-amino-3-cyano-4*H*-pyran derivatives

A simple, mild, efficient protocol was developed for the synthesis of a series of novel 2-amino-3-cyano-4*H*-pyran derivatives (**59**) *via* one-pot, three-component condensations of aldehydes (**1**), malononitrile (**2**) and various active 1,3-dicarbonyl compounds (**58**) using molecular iodine as a catalyst in aqueous medium under ultrasonic irradiation (Scheme 23).[129] During the optimization for the synthesis of **59a**, it was found that the ultrasound (US)-irradiated pathway was more advantageous even than the microwave (MW)-irradiated pathway. Among the various solvents tested, water was found to be the most suitable for these reactions.

2.3. Ultrasound assisted synthesis of S-*heterocycles in aqueous media*

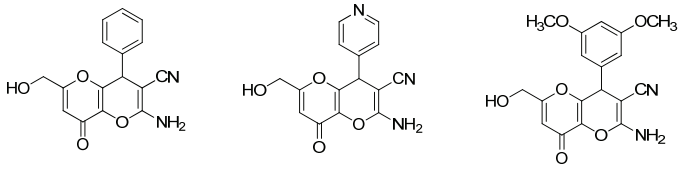
2.3.1. Synthesis of spiro[indole-3,4′-pyrazolo[3,4-*e*][1,4]thiazepines]

1,4-Thiazepine derivatives are important because of their significant therapeutic and biological activities.[130–132] Dandia *et al*. (Scheme 24)[133] developed a simple, straightforward, efficient, ultrasound-assisted, catalyst-free, one-pot, three-component protocol for the synthesis of biologically relevant spiro[indole-3,4′-pyrazolo[3,4-*e*][1,4]thiazepines] (**61**) by the domino reactions of isatins (**21**), 3-amino-5-methylpyrazoles (**18a**) and 2-mercaptoacetic acid/2-mercaptopropionic acid (**60**) in water at room temperature.



R = C_6H_5, 2-F-C_6H_4, 4-F-C_6H_4, 3-pyridyl, 4-pyridyl, 2-furyl, 2-thienyl, 3,5-$(OCH_3)_2$-C_6H_3, 2-Cl-C_6H_4, 2,4-diCl-C_6H_3, 3-F-C_6H_4, 3-CH_3-C_6H_4, 3-Br-C_6H_4

Representatives



57a, 95% **57b**,90% **57c**, 96%

Entry	R	With sonication		Without sonication	
		Time, min	Yield, %	Time, h	Yield, %
1	C_6H_5	10	95	1.2	79
2	2-F–C_6H_4	20	85	3.0	0
3	2-Cl–C_6H_4	25	88	3.0	0
4	2,4-$(Cl)_2$–C_6H_3	25	89	3.0	0
5	3-F–C_6H_4	15	93	2.5	65
6	3-CH_3–C_6H_4	5	97	1.0	68
7	3-Br–C_6H_4	15	95	1.7	67
8	3,5-$(OCH_3)_2$–C_6H_3	5	96	2.5	53
9	4-F–C_6H_4	6	98	1.5	60
10	3-pyridyl	5	98	1.2	56
11	4-pyridyl	15	90	2.0	55
12	2-furyl	5	98	1.5	60
13	2-thienyl	6	97	1.0	62

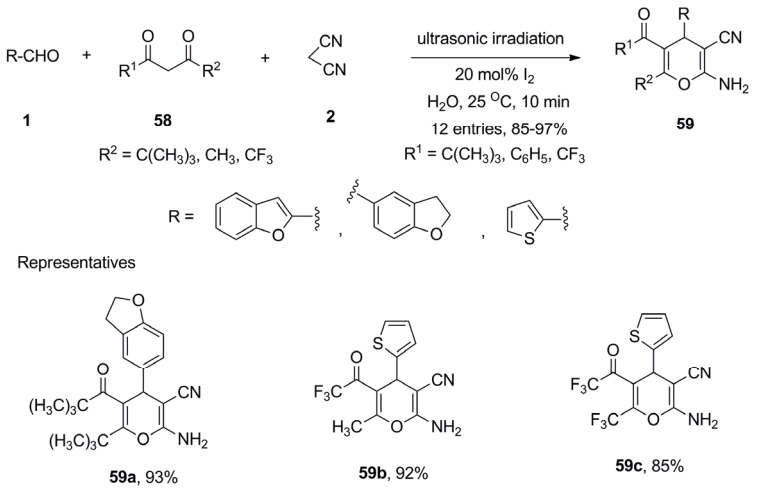
Scheme 22. Ultrasound-promoted synthesis of 2-aminopyrano[3,2-*b*]pyrans in water.

2.3.2. Synthesis of rhodanines

Rhodanines (**64**), in particular, have immense pharmacological efficacies that include the inhibition of targets such as HCV NS3 protease[134] and β-lactamase.[135] Rostamnia and Lamei (Scheme 25)[136] demonstrated a facile, straightforward, environmentally benign, catalyst-free, ultrasound-assisted, aqueous-mediated protocol for the synthesis of rhodanines (**64**) from the reactions of various primary amines (**14**), carbon disulfide (**62**) and dialkyl but-2-ynedioate (**63**) at room temperature.

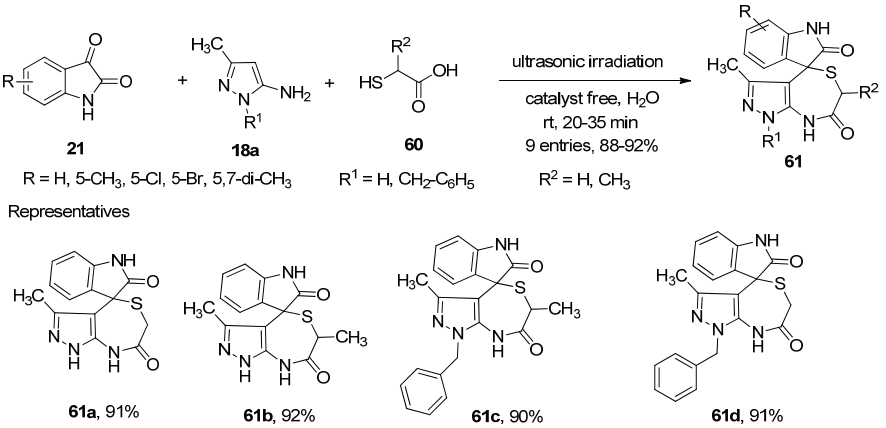
2.3.3. Synthesis of spiro[indole-thiazolidinones]

Dandia *et al*. (Scheme 26)[137] reported other simple, convenient, one-pot, three-component tandem reactions of isatins (**21**), various amines (**14**) and 2-mercaptoacetic acid/2-mercaptopropionic acid (**60**) to afford a series of pharmaceutically important spiro[indole-thiazolidinones] (**65**) using cetyltrimethylammonium bromide [CTAB] as a phase transfer catalyst in water under ultrasound-irradiation at room temperature.

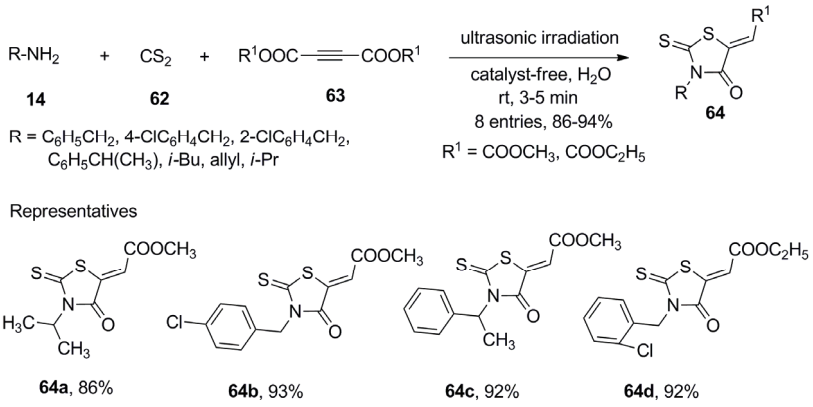


Entry	solvent	RT (25 °C)		Reflux (80 °C)		MW		US	
		Time min	Yield of **59a**, %	Time min	Yield of **59a**, %	Time min	Yield of **59a**, %	Time min	Yield of **59a**, %
1	Solvent-free	600	15	600	20	20	25	10	45
2	Diethyl ether	600	16	600	25	20	35	10	45
3	*n*-Hexane	600	15	600	25	20	40	10	50
4	CH_3CN	600	18	600	30	20	45	10	55
5	1,4-Dioxane	600	12	600	35	20	40	10	50
6	DMSO	600	12	600	25	20	30	10	58
7	THF	600	10	600	20	20	35	10	60
8	DMF	600	05	600	15	20	30	10	55
9	EtOH	600	10	600	20	20	30	10	80
10	H_2O	600	25	600	30	20	40	10	95

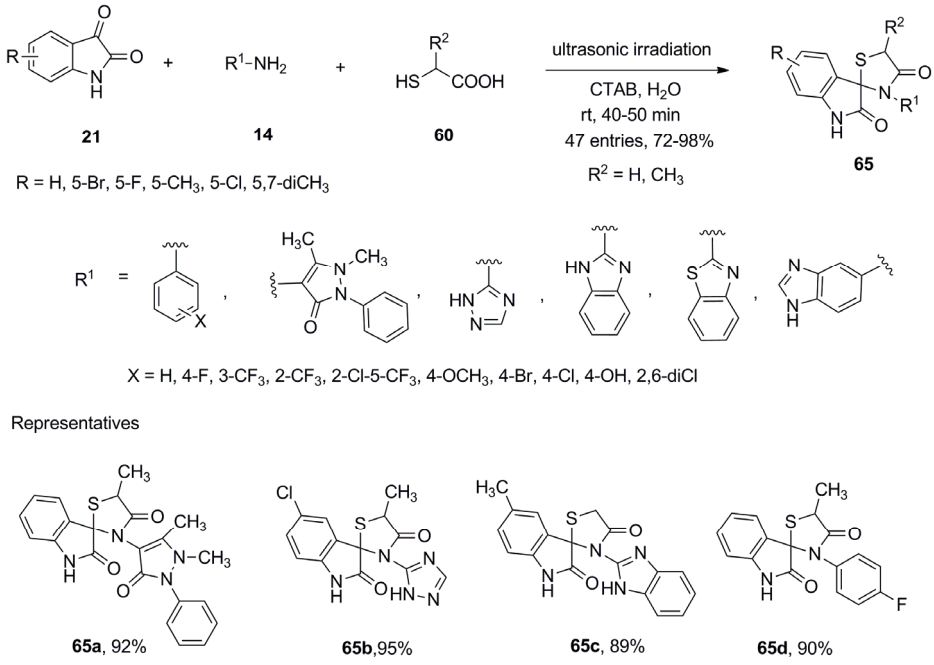
Scheme 23. Ultrasound-promoted synthesis of 2-amino-3-cyano-4*H*-pyran derivatives in water.



Scheme 24. Ultrasound-promoted synthesis of spiro[indole-3,4′-pyrazolo[3,4-*e*][1,4]thiazepines] in water.



Scheme 25. Ultrasound-mediated catalyst-free synthesis of rhodanines in aqueous media.

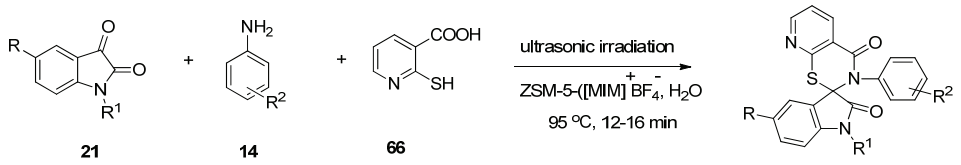


Scheme 26. Ultrasound-promoted synthesis of spiro[indole-thiazolidinones] in water.

2.3.4. Synthesis of spiro[indole-pyrido[3,2-*e*][1,3]thiazines]

Arya and his group (Scheme 27)[138] successfully employed their synthesized ZSM-5 zeolite-supported Brønsted-acid ionic liquid as a catalyst for the synthesis of novel biologically interesting spiro[indoline-3,2′-pyrido[3,2-*e*][1,3]thiazine]--2,4′(3′*H*)-diones (**67**) from one-pot, three-component reactions of isatins (**21**), various amines (**14**) and 2-mercaptonicotinic acid (**66**) in water under ultrasonic irradiation at room temperature. It was found that for the same catalytic system,

the ultrasound-assisted pathway was much more efficient than the conventional and MW irradiated pathway for this particular synthesis with respect to yield and time.



21 14 66 67

Entry	R	R^1	R^2	US		MW		Heating	
				Time min	Yield %	Time min	Yield %	Time min	Yield %
1	Br	$COCH_3$	H	12	92	10	80	48	20
2	Br	CH_3	H	15	90	13	78	50	Trace
3	Cl	$COCH_3$	H	15	90	13	80	42	32
4	Cl	CH_3	H	16	94	15	76	45	33
5	NO_2	$COCH_3$	H	15	89	15	78	48	Trace
6	NO_2	CH_3	H	15	88	13	78	45	35
7	H	$COCH_3$	4-OCH_3	13	89	11	75	45	Trace
8	H	CH_3	4-OCH_3	13	88	11	75	46	30
9	H	$COCH_3$	4-CH_3	15	80	12	68	48	20
10	H	CH_3	4-CH_3	15	85	12	70	48	Trace

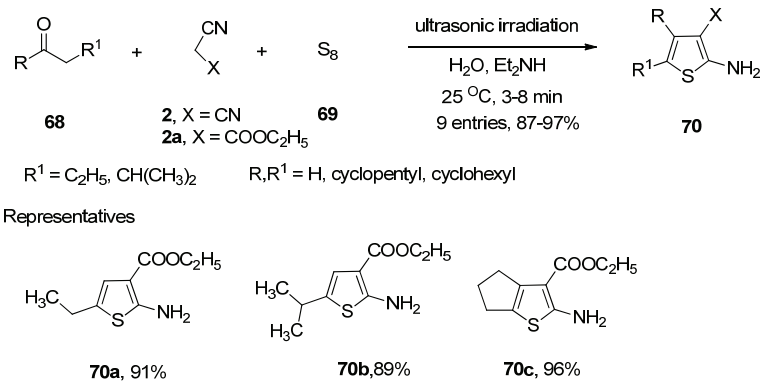
Scheme 27. Ultrasound-promoted synthesis of spiro[indoline-3,2′-pyrido[3,2-*e*][1,3]thiazine]--2,4′(3′*H*)-diones in aqueous media.

2.3.5. Synthesis of 2-aminothiophenes

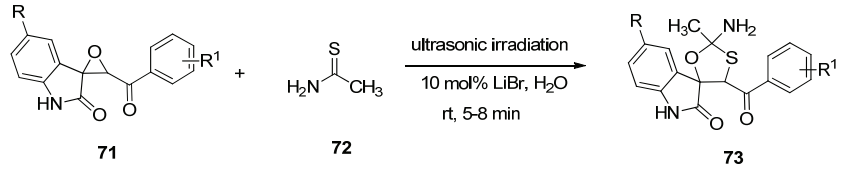
2-Aminothiophenes are very common in naturally occurring compounds[139] having various pharmaceutical[140–142] efficacies. In 1976, Gewald[143] first synthesized these scaffolds in a one-pot cyclocondensation of ketones or aldehydes, *β*-substituted acetonitriles and elemental sulfur. Recently, Mojtahedi *et al.* (Scheme 28)[144] synthesized a number of substituted 2-aminothiophenes (**70**) *via* one-pot, three-component condensations of carbonyl compounds (**68**), malononitrile (**2**) or ethyl cyanoacetate (**2a**) and elemental sulfur (**69**) in the presence of diethylamine as base in water under ultrasonic irradiation at room temperature.

2.3.6. Synthesis of spiro[indole-3,5′-[1,3]oxathiolanes]

Novel ultrasound-assisted syntheses of substituted spiro[indole-3,5′-[1,3]-oxathiolane]-2(1*H*)-ones (**73**) were achieved in the reactions of spiro[indole-3,2′--oxiranes] (**71**) with thioacetamide (**72**) using LiBr as catalyst in water at room temperature (Scheme 29).[145] The same reactions were also performed using both microwave and conventional pathways. Interestingly, it was observed that for these particular syntheses, the ultrasonic-irradiated process was superior in comparison to microwave irradiation or the conventional method in terms of reaction rates and yields.



Scheme 28. Ultrasound-promoted synthesis of 2-aminothiophenes in water.



Entry	R	R^1	US		MW		Stirring	
			Time min	Yield %	Time min	Yield %	Time min	Yield %
1	H	H	7	84	20	76	5	37
2	Cl	H	6	87	–	–	5	64
3	H	4-F	5	86	–	–	5	58
4	Br	H	6	87	–	–	5	61
5	F	4-Cl	7	82	–	–	5	59
6	CH_3	3-Cl	8	86	–	–	5	62

Scheme 29. Ultrasound–promoted synthesis of spiro[indole-3,5′-[1,3]oxathiolanes] in water.

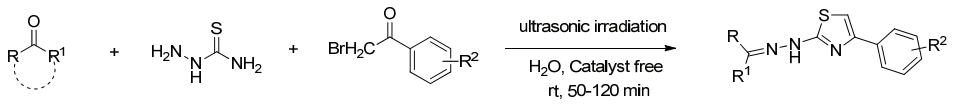
2.3.7. Synthesis of *N*-(4-arylthiazol-2-yl)hydrazones

Zhang *et al*. (Scheme 30)[146] demonstrated a simple, mild, catalyst-free, ultrasound-assisted, high-yielding, one-pot, three-component protocol for the synthesis of N-(4-arylthiazol-2-yl)hydrazones (**77**) in the reactions of various carbonyl compounds (**74**), hydrazinecarbothioamide (**75**) and 2-bromo-1-phenylethanones (**76**) in water at room temperature. The ultrasonic-assisted pathway was found to be advantageous as compared to the conventional stirring method in terms of reaction times and yields.

3. ULTRASOUND-ASSISTED SYNTHESIS OF OTHER VARIOUS SCAFFOLDS IN AQUEOUS MEDIUM

3.1. Synthesis of bis-coumarins

Bis-coumarins (**78**) are found to exhibit significant phytochemical efficacies, such as antimicrobial,[147] cytotoxic[148] and potent HIV-1 integrase inhibitor[149,150]



74 75 76 77

Entry	R	R^1	R^2	With sonication		Conventional stirring	
				Time, min	Yield, %	Time, h	Yield, %
1	C_6H_5	H	H	50	90	1.5	80
2	4-OCH_3–C_6H_4	H	H	60	95	2	76
3	2-OH–C_6H_4	H	H	100	90	4	82
4	2-Cl–C_6H_4	H	H	70	89	4	78
5	3-Cl–C_6H_4	H	H	120	89	4	89
6	4-Cl–C_6H_4	H	H	100	88	3	72
7	2-OH–, 4-OCH_3–C_6H_3	H	H	70	92	3	92
8	4-$N(CH_3)_2$–C_6H_4	H	H	100	86	3.5	65
9	$(CH_2)_4$	–	H	90	88	3	60
10	$(CH_2)_5$	–	H	80	89	3	80
11	4-CH_3–C_6H_4	CH_3	4-Cl	120	92	4	62
12	$(CH_2)_5$	–	4-CH_3	100	93	4	87

Scheme 30. Ultrasound-promoted synthesis of 2-(4-arylthiazol-2-yl)hydrazones in water.

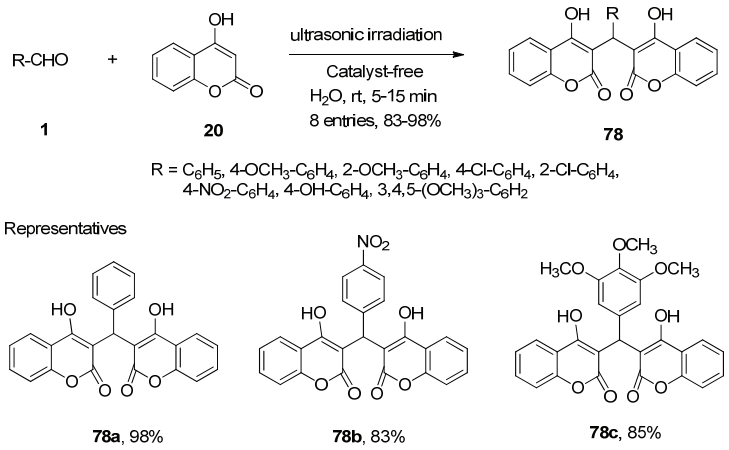
activities. The many diverse applications of bis-coumarins have drawn considerable interest of synthetic chemists during the last several years to develop useful synthetic routes to this scaffold of potential interest; as a result, many methods have already been reported.[151–160] Although these methods possess notable merits, they still suffer from drawbacks, such as the use of toxic organic solvents, harsh reaction conditions, expensive catalysts, longer reaction times, *etc*. Therefore, in 2012, Al-Kadasi and Nazeruddin (Scheme 31)[161] developed a simple, efficient, environmentally-friendly, ultrasound-assisted, catalyst-free, one pot, three-component protocol for the synthesis of bis-coumarins (**78**) by condensing 4-hydroxycoumarin (**20**) with various aromatic aldehydes (**1**) in water at ambient temperature.

3.2. Aza-Michael addition reaction

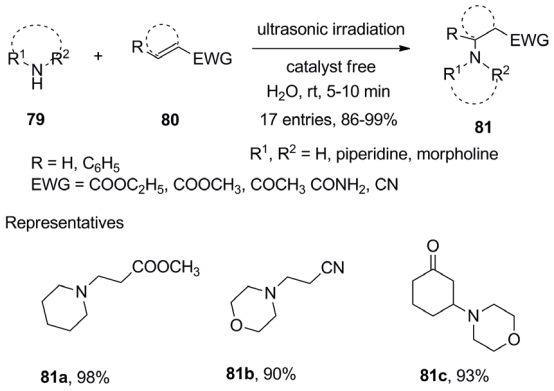
Banik and his group (Scheme 32)[162] described a rapid, simple, straightforward, ultrasound-assisted, catalyst-free reaction between several amines (**79**) and *α*-, and *β*-unsaturated carbonyls or nitriles (**80**) to afford the corresponding aza-Michael addition products (**81**) in water at room temperature. During optimization, the effect of various solvents was tested and water was found to be the superior under ultrasonic-irradiated condition.

3.3. Synthesis of dithiocarbamates

Organic dithiocarbamates are important synthetic intermediates of various biologically promising compounds.[163] Azizi *et al*. (Scheme 33)[164] reported a simple,



Scheme 31. Ultrasound-promoted, catalyst-free synthesis of bis-coumarins in an aqueous medium.



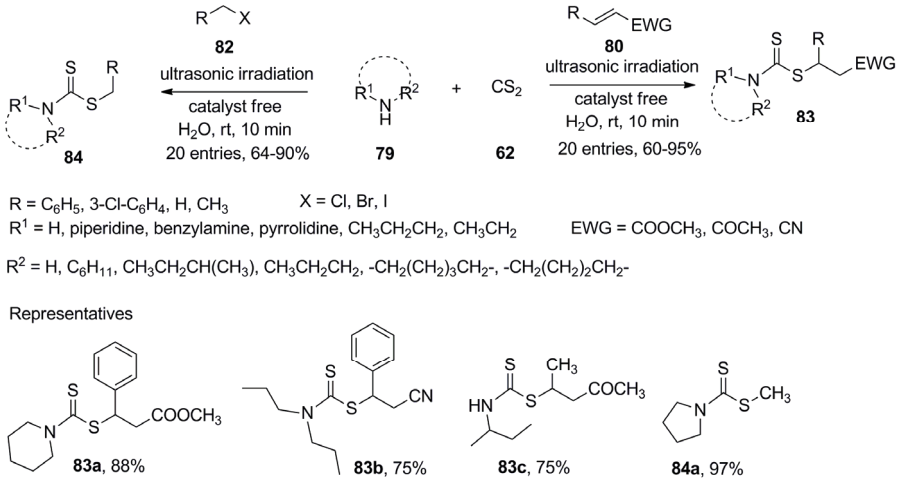
Entry	Solvent	Time, min	Yield of **81a**, %
1	Tetrahydrofuran	60	45
2	Ethanol	60	62
3	Methanol	60	57
4	Dichloromethane	60	55
5	Acetone	60	48
6	Acetonitrile	60	71
7	Water	5	98
8	Solvent-free	15	93

Scheme 32. Ultrasound-assisted synthesis of *β*-amino carbonyl and related compounds in water.

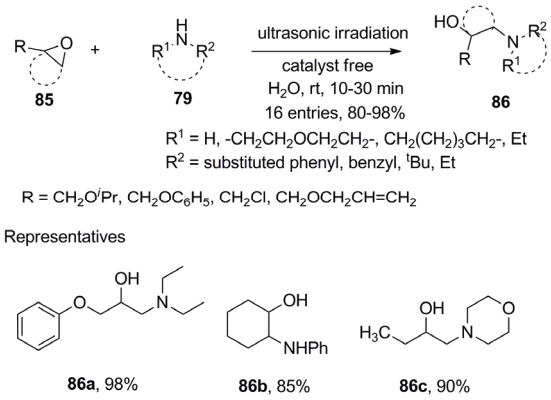
straightforward, ultrasound-assisted, catalyst-free, one-pot, three-component condensation of various amines (**79**), carbon disulfide (**62**) and *α,β*-unsaturated carbonyl compounds (**80**) or alkyl halides (**82**) for the rapid synthesis of dithiocarbamates (**83** and **84**) in water at room temperature.

3.4. Synthesis of β*-aminoalcohols*

β-Aminoalcohols (**86**) are useful building blocks of various biologically active natural products,[165] unnatural amino acids,[166] and chiral auxiliaries.[167] A simple, straightforward, environmentally benign, ultrasound-assisted, catalyst--free protocol was reported for the synthesis of *β*-aminoalcohols (**86**) from the reactions of epoxides (**85**) with various primary or secondary amines (**79**) in water at room temperature (Scheme 34).[168]



Scheme 33. Ultrasound-promoted, catalyst-free synthesis of dithiocarbamates in water.



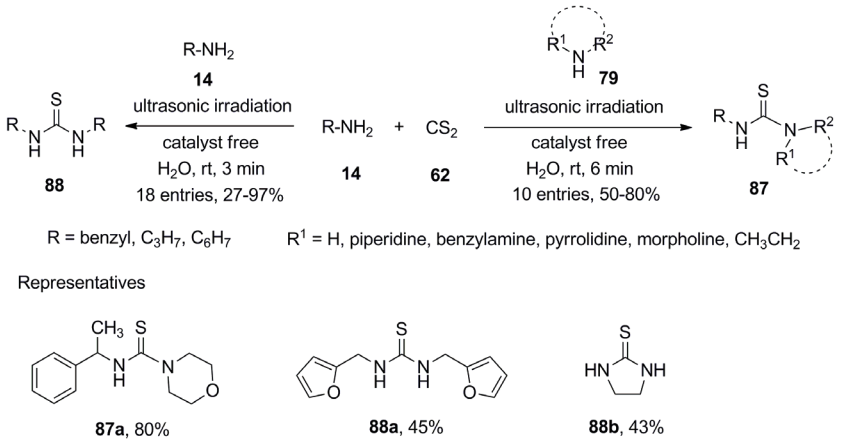
Scheme 34. Ultrasound-mediated, catalyst-free synthesis of β-aminoalcohols in water.

3.5. Synthesis of substituted thiourea

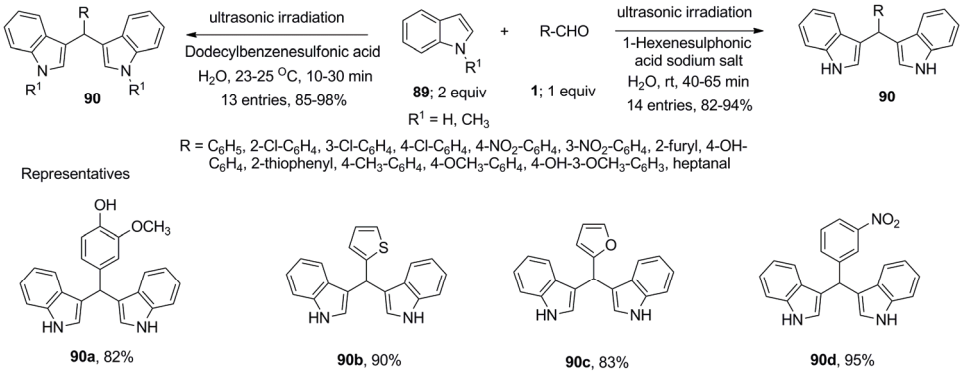
A simple, ultrasound-assisted, catalyst-free, one-pot pseudo three-component condensation of primary (**14**) or secondary amines (**79**) with carbon disulfide (**62**) was described by Azizi *et al*. (Scheme 35)[169] for the rapid synthesis of various thiourea derivatives (**87** and **88**) in water at room temperature. Under the same optimized conditions, the reactions also proceeded smoothly in poly(ethylene glycol) (PEG).

3.6. Synthesis of bis(indol-3-yl)methanes

Joshi *et al*. (Scheme 36)[170] reported an efficient, simple, rapid, ultrasound-promoted protocol for the synthesis of bis(indol-3-yl)methanes (**90**) in the reactions of indoles (**89**) with various aldehydes (**1**) in water using 1-hexenesulphonic acid sodium salt as a catalyst at room temperature.



Scheme 35. Ultrasound-mediated, catalyst-free synthesis of substituted thioureas in water.



Scheme 36. Ultrasound-promoted synthesis of bis(indol-3-yl)methanes in water.

The very next year, Li *et al*. (Scheme 36)[171] developed another ultrasound-assisted protocol for the synthesis of bis(indol-3-yl)methanes (**90**) from the same series of reactions using *p*-dodecylbenzenesulfonic acid as the catalyst in aqueous medium at room temperature.

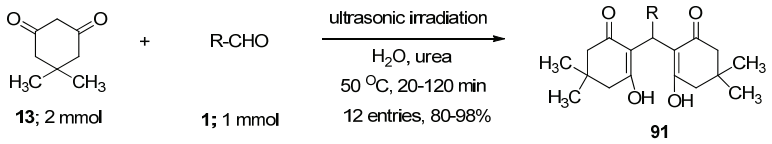
3.7. Synthesis of 2,2'-(arylmethylene)bis[3-hydroxy-5,5-dimethyl-2-cyclohexen--1-one] derivatives

A simple, straightforward, efficient, ultrasound-assisted protocol was developed for the synthesis of 2,2′-(arylmethylene)bis[3-hydroxy-5,5-dimethyl-2-cyclohexen-1-ones] (**91**) from the condensations of dimedone (**13**) with various aldehydes (**1**) using urea as the catalyst in water at 50 °C (Scheme 37).[172]

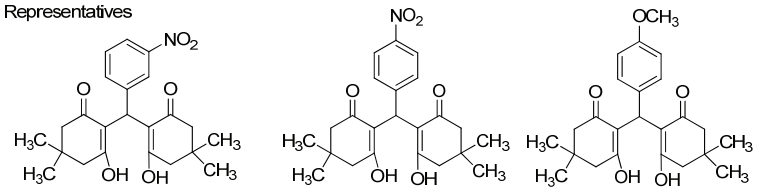
3.8. Synthesis of thioethers

Song *et al*. (Scheme 38)[173] synthesized 2-[aryl(arylthio)methyl]-3-hydroxy--5,5-dimethylcyclohex-2-enones (**93**) *via* simple, efficient, ultrasound-assisted,

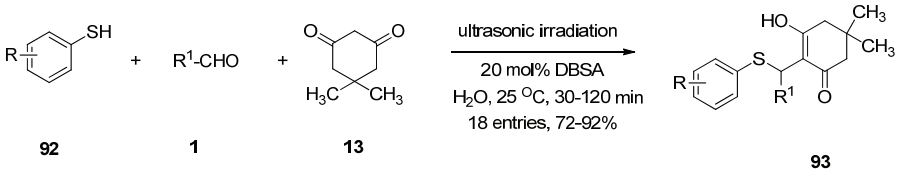
one-pot, three-component reactions between aromatic aldehydes (**1**), substituted thiophenol (**92**) and dimedone (**13**) using *p*-dodecylbenzenesulfonic acid (DBSA) as a catalyst in water at ambient temperature. It was observed that the use of ultrasonic irradiation greatly enhances the rate of the reaction.



R = C_6H_5, 2-$NO_2C_6H_4$, 3-$NO_2C_6H_4$, 4-$NO_2C_6H_4$, 2-ClC_6H_4, 3-ClC_6H_4, 4-ClC_6H_4, 2,4-diClC_6H_3, 4-$CH_3C_6H_4$, 4-$OCH_3C_6H_4$, 4-OH-3-$OCH_3C_6H_3$, 3,4-diClC_6H_3

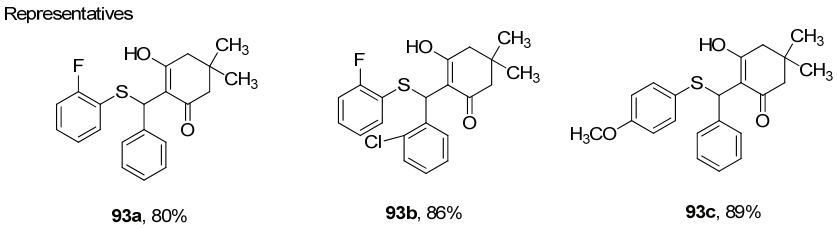


Scheme 37. Ultrasound-promoted synthesis of 2,2′-(arylmethylene)bis[3-hydroxy-5,5--dimethyl-2-cyclohexen-1-one] derivatives in water.



R = 4-F, 4-OCH_3, 2-F, 2-CH_3, 2-NH_2, 4-OH, 3-CH_3, 4-CH_3, 2,4-diCH_3, 2,4-diCl, 3-$CH(CH_3)_2$

R^1 = C_6H_5, 2-Cl-C_6H_4, 4-$OCH_3C_6H_4$, 4-Cl-C_6H_4, 4-Br-C_6H_4, 4-OH-C_6H_4, 4-$NO_2C_6H_4$

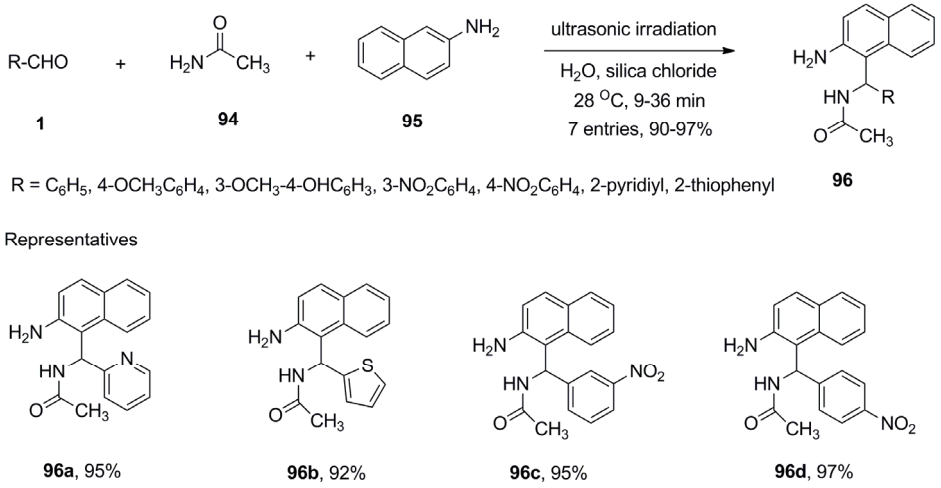


Scheme 38. Ultrasound-promoted synthesis of 2-[aryl(arylthio)methyl]-3-hydroxy-5,5--dimethylcyclohex-2-enone in water.

3.9. Synthesis of 1-(amidoalkyl)-2-naphthylamines

A straightforward, efficient, ultrasound-assisted protocol was developed for the synthesis of 1-(amidoalkyl)-2-naphthylamines (**96**) *via* one-pot, three-component

condensations of aldehydes (**1**), acetamide (**94**) and 2-naphthylamine (**95**) using silica chloride as a catalyst in aqueous medium at ambient temperature (Scheme 39).[174] Use of organic solvents, such as CH_3CN, $CHCl_3$, DCM, DCE and EtOH, gave inferior results in terms of both reaction times and yields. It was also observed that ultrasonic irradiation has a distinct effect on the rate of the reaction.



Scheme 39. Ultrasound-promoted synthesis of 1-(amidoalkyl)-2-naphthylamines in water.

3.10. Synthesis of 2-[3-aryl-1-(2-arylethyl)-2-propen-1-ylidene]hydrazinecarboximidamide hydrochloride

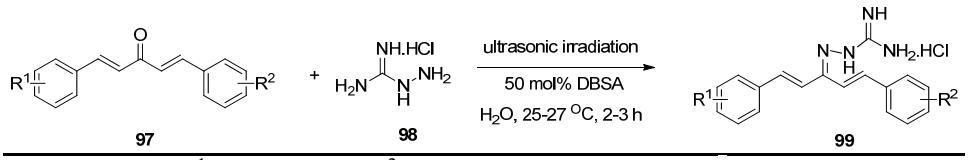
Li *et al*. (Scheme 40)[175] developed a simple, facile, ultrasound-assisted method for the synthesis of various biologically promising 2-[3-aryl-1-(2-arylethyl)-2-propen-1-ylidene]hydrazinecarboximidamide hydrochlorides (**99**) by the condensation between 1,5-aryl-1,4-pentadien-3-one (**97**) and aminoguanidine hydrochloride (**98**) using *p*-dodecylbenzenesulfonic acid (DBSA) as the catalyst in water at ambient temperature. It was well demonstrated that the application of ultrasonic irradiation enhances the reaction rate.

3.11. Synthesis of highly substituted propanamide derivatives

A mild, simple, rapid, ultrasound-assisted, one-pot, three-component, catalyst-free approach to the synthesis of highly substituted propanamide derivatives (**104**) from the reactions of 2-oxopropyl benzoate (**100**), isocyanide (**101**) and carboxylic acid (**102**) or cinnamic acid (**103**) derivatives in water at room temperature was developed by Ramazani *et al.* (Scheme 41).[176]

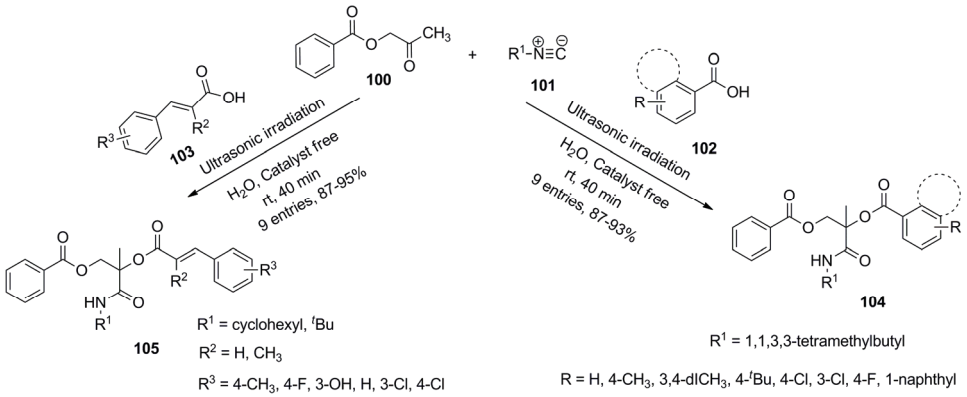
4. CONCLUSIONS

It is the cutting edge for today's methodologists to develop protocols in greener pathways by avoiding the extensive use of hazardous reagents and solvents,



Entry	R^1	R^2	With sonication		Without sonication	
			Time, h	Yield, %	Time, h	Yield, %
1	H	H	2	94	8	91
2	2-Cl	2-Cl	2	87	2	55
3	3-Cl	3-Cl	2	84	2	72
4	4-Cl	4-Cl	2	85	2	51
5	4-CH_3	4-CH_3	2	87	6	65
6	H	4-CH_3	2.5	95	8	72
7	H	4-OCH_3	2	90	2	45
8	4-OCH_3	4-OCH_3	2	91	5	80
9	3,4-(OCH_2O)	3,4-(OCH_2O)	2.5	92	2.5	63
10	2,4-diCl	2,4-diCl	3	88	3	75
11	3,4-diCl	3,4-diCl	3	89	3	78
12	H	4-Cl	2.5	94	2.5	67

Scheme 40. Ultrasound-promoted synthesis of 2-[3-aryl-1-(2-arylethyl)-2-propen-1-yli-dene]hydrazinecarboximidamide hydrochlorides in water.



Scheme 41. Ultrasound-promoted synthesis of highly substituted propanamide derivatives in water.

harsh reaction conditions, *etc*. For this reason, in recent time, aqueous-mediated syntheses are in high demand. Again, it is well established that the application of ultrasonic irradiation in organic synthesis is very advantageous in comparison to the conventional methods. Sometimes it is also more advantageous than a microwave irradiated pathway. Harsh reaction conditions could be avoided by application of ultrasonic irradiation. Therefore, it is not necessary to mention that ultrasound-assisted chemical synthesis in aqueous medium is one of the greenest approaches towards sustainability. As a result, in the recent past, there have been

an immense number of applications of ultrasound in organic reactions for the synthesis of diverse organic scaffolds in aqueous media. The present review summarizes the latest developments on ultrasound-assisted and water-mediated organic synthesis reported to date.

Acknowledgements. The author is thankful to the Indus International University, Una, Himachal Pradesh, India, for the support and the Kartha Education Society, Mumbai, India, for the financial help. Special thanks go to Dr. Sudipta Som, Samik Mukherjee, Pankaj Choudhari, Somananda Sanyal and Hridaynath Bhattachariya for their kind help during the literature search. The author is grateful to Prof. Dr. Goutam Brahmachari, Department of Chemistry, Visva-Bharati University, for his wholehearted guidance throughout.

ИЗВОД

НОВИЈИ ПРИМЕРИ УПОТРЕБЕ УЛТРАЗВУКА У ОРГАНСКОЈ СИНТЕЗИ У ВОДЕНИМ РАСТВОРИМА

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У протеклих неколико година публикован је значајан број радова о примени ултразвука у органским реакцијама за потребе синтезе врло различитих класа органских једињења. Истовремено, када је у питању „зелена" хемија, вода као растварач је најбоље решење. На основу тога може се рећи да су методе које обједињују примену ултразвука и употребу воде као растварача, главни ослонац у развоју протокола који су потпуно прихватљиви са становишта очувања животне средине. Ова ревија даје приказ најновијих резултата у примени ултразвука у воденим растворима у органској синтези.

(Примљено 17. фебруара, ревидирано 27. априла, прихваћено 3. маја 2017)

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