AN EFFICIENT SYNTHESIS OF 2-ARYLBENZOTHIAZOLES USING SILICA SULFURIC ACID, OXALIC ACID AND ALUMINUM CHLORIDE HYDRATE AS HETEROGENEOUS AND HOMOGENEOUS CATALYST SYSTEMS

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A convenient method for the synthesis of 2-arylbenzothiazoles from condensation of aromatic/heteroaromatic aldehydes with 2-aminothiophenol has been developed using inexpensive, green and reusable solid acids – silica sulfuric acid (SSA), oxalic acid or $AlCl_3 \cdot 6H_2O$ as efficient catalysts. The reactions occurred under mild conditions to afford the corresponding 2-arylbenzothiazoles in good to excellent yields.

Keywords: Acidity; Aldehydes; Antibiotics; Antibacterial activity; Antiaromaticity; Antifungal activity; 2-Arylbenzothiazoles; Silica sulfuric acid; Oxalic acid; $AlCl_3 \cdot 6H_2O$; 2-Aminothiophenol.

2-Arylbenzothiazoles are an important class of biologically active heterocyclic compounds which are becoming increasingly attractive due to their pharmacological and industrial importance[1]. Several functionalized 2-arylbenzothiazoles are widely used in medicinal chemistry and have been found to exhibit a wide spectrum of biological effects including antitumour, anticonvulsant, and antiviral activities[2–5]. In addition, several industrial applications have been reported for these compounds as antioxidants[6–7], fluorescents[8] and photochromic agents[9].

Thus, these advantages described to benzothiazoles make them to continue attracting the attention of researchers for the development of efficient approaches towards these compounds[10–13]. The methods previously reported for the synthesis of 2-arylbenzothiazoles include: (i) the condensation of 2-aminothiophenol with aldehydes[14–20], carboxylic acids[21–25], acid chlorides[26–27] and esters[28], and (ii) the cyclization of thiobenzanilides[29–31]. Other synthetic routes employ microwave-assisted reaction of 2-aminothiophenol with β-chlorocinnamaldehydes[32], palladium-catalyzed direct

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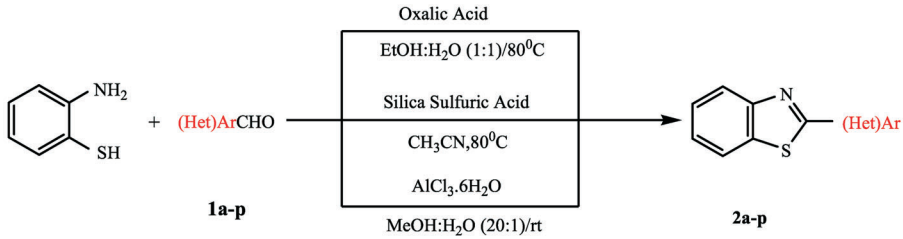
coupling of benzothiazoles with arylbromides[33], palladium-catalyzed condensation of arylhalides with 2-aminothiophenol followed by dehydrative cyclization[34], reaction of copper(I) thiobenzoate with 2-iodoanilines[35], and the aquatic reaction of the corresponding thioamidinium salts and 2-aminothiophenol[36]. Recently, some simple one-step methods of syntheses of 2-arylbenzothiazoles have been found to be effective using perchloric acid-doped polyaniline[37], ceric ammonium nitrate (CAN)[38], tungstophosphoric acid impregnated zirconium phosphate[39], zirconium(IV) oxide chloride and anhydrous copper(II) sulfate[40], Dowex 50W [41], activated carbon under oxygen as catalysts[42], *p*-toluenesulfonic acid[43], $H_2O_2/Fe(NO_3)_3$ [44], solid heteropolyacid supported on silica gel ($Cu_{3/2}PMo_{12}O_{40}/SiO_2$)[45], alum ($KAl(SO_4)_2{\cdot}12H_2O$)[46], Bakers' yeast[47], trichloroisocyanuric acid[48] and silica sulfuric acid under microwave irradiation[49]. However, many of these methods have one or more disadvantages such as high reaction temperature, expensive metallic compounds, requirement for excess of reagenets, harsh reaction conditions, air sensitive catalysts, multistep processes, prolonged reaction times, use of toxic and costly catalysts, requirement for hazardous and carcinogenic organic solvents. To avoid these drawbacks, development of simpler, safe, inexpensive, green and efficient protocols are highly demanded. In our previous work, we reported for the first time a simple, efficient and eco-friendly procedure for the synthesis of 2-arylbenzothiazoles utilizing glacial acetic acid as a non-toxic and inexpensive catalyst under microwave irradiation and solvent-free conditions[50].

RESULTS AND DISCUSSION

We describe a simple, efficient and practical method for a one-pot synthesis of 2-arylbenzothiazoles using catalytic amount of silica sulfuric acid (SSA), oxalic acid or $AlCl_3{\cdot}6H_2O$ as catalysts in acetonitrile, EtOH/H_2O (1:1) and MeOH/H_2O (20:1), respectively (Scheme 1).

Oxalic acid is the most acidic among the simple dicarboxylic acids having a first p*K*a value of 1.27. Application of oxalic acid as a catalyst was previ-



SCHEME 1

ously reported for one-pot synthesis of 2-aryl-1-arylmethyl-1*H*-benzimidazoles and 2,4,5-triaryl-1*H*-imidazoles[51], dealumination of zeolite[52], and synthesis of quinoxalines[53]. These special activities inherent to oxalic acid prompted us to explore this catalyst for the synthesis of 2-arylbenzothiazoles in a model reaction between 2-aminothiophenol (1 mmol) and 4-methoxybenzaldehyde (1 mmol) in different solvents and various amounts (mole %) of oxalic acid. The product 2-(4-methoxyphenyl)benzothiazole was isolated in 86% yield under the optimum reaction condition, that is the use of EtOH/H_2O (1:1) as solvent and 10 mole % of oxalic acid catalyst at 80 °C. Similarly, several other aromatic aldehydes (**1a–1p**) reacted well under the present reaction conditions to afford the corresponding 2-arylbenzothiazoles (**2a–2p**) in high yields (Table I).

TABLE I
Condensation of 2-aminothiophenol with aromatic/heteroaromatic aldehydes catalyzed by oxalic acid (10 mole %) in EtOH/H_2O (1:1)

(Het)Ar **1a–1p**	Product[a]	Time, min	Yield, %[b]	M.p., °C	
				Found	Reported
Ph	**2a**	30	80	111–112	112–114[41]
2-$MeOC_6H_4$	**2b**	40	74	103–105	101–103[41]
4-$MeOC_6H_4$	**2c**	25	86	120–121	120–121[41]
2-HOC_6H_4	**2d**	20	78	127–128	127–128[17]
4-MeC_6H_4	**2e**	40	74	82–84	85[17]
2-MeC_6H_4	**2f**	50	68	52–54	53–54[17]
4-BrC_6H_4	**2g**	25	86	132–133	132[41]
3-BrC_6H_4	**2h**	30	84	82–83	83–84[41]
4-$Me_2NC_6H_4$	**2i**	30	83	157–159	160–160[41]
4-ClC_6H_4	**2j**	25	80	116–118	115–117[41]
4-FC_6H_4	**2k**	30	84	100–102	101–103[48]
4- CNC_6H_4	**2l**	10	90	161–162	162–164[20]
3-$NO_2C_6H_4$	**2m**	30	86	179–180	181–182[41]
Pyridyl-4-yl	**2n**	30	80	132–133	134–135[17]
Pyridyl-2-yl	**2o**	20	72	136–137	137–138[48]
Thienyl-2-yl	**2p**	20	78	100–101	99[17]

[a] All the products were characterized from spectral (^1H, ^{13}C NMR, MS and IR) data which were in accord with those of the literature cited. [b] Isolated yields.

Silica sulfuric acid (SSA) is another eco-friendly, versatile solid acid which has recently found much interest as a catalyst[54–58] in various organic transformations. These salient catalytic activities allocated to silica sulfuric acid have encouraged us, in second part of the present work, to examine its catalytic efficiency in reaction of 2-aminothiophenol with aldehydes for the synthesis of 2-arylbenzothiazoles. Among various solvents like CH_3CN, MeOH, EtOH, EtOAc, CH_2Cl_2 and CCl_4 used for the condensation of 2-aminothiophenole (1 mmol) with 4-methoxybenzaldehyde (1 mmol) as model reaction, acetonitrile was the solvent of choice as best results in terms of the yield was obtained using 0.05 g of SSA per mmol of the aldehyde when the reaction was run at 80 °C for 20 min (Table II).

Similarly, to establish the scope and usefulness of this methodology, a number of reactions were conducted and the isolated products are summarized in Table III. Comparison of the experimental results summarized in Table I with those given in Table III indicates a general increase in reaction rates and also, in some cases, slight improvement of the yields when SSA is used as a catalyst.

Recently, Shingare and co-Workers reported one-step synthesis of 2-substituted benzothiazoles using silica sulfuric acid under microwave irradiation (domestic MW oven). It has been shown that application of microwave irradiation reduce the reaction time and increases the product yield[49]. The use of domestic microwave ovens involves disadvantages such as: without control of temperature, power and pressure inside the reaction vessel.

TABLE II
Optimization of the silica sulfuric acid-catalyzed condensation of 4-methoxybenzaldehyde (**1c**) with 2-aminothiophenol

Solvent	Amount of catalyst, g	Time, min	Yield, %[a]
CH_2Cl_2	0.05	50	40
CCl_4	0.05	50	30
EtOH	0.05	20	62
MeOH	0.05	20	54
EtOAc	0.05	20	60
MeCN	0.05	20	86
MeCN	0.03	30	80
MeCN	0.07	35	84

[a] Isolated yields.

Therefore, these studies carried out using domestic microwave ovens present incomplete experimental information, and the reaction conditions have poor reproducibility[59].

Finally, in search for other more robust and efficient acidic catalysts for the synthesis of 2-arylbenzothiazoles, we chose to explore the catalytic efficiencies of several solid halides like $AlCl_3 \cdot 6H_2O$, $ZnCl_2$ and $SbCl_3$. Among these, $AlCl_3 \cdot 6H_2O$ appeared as the most effective in catalyzing the condensation of benzaldehyde with 2-aminothiophenol as a model reaction. To optimize the reaction conditions, different solvents such as MeOH, EtOH, EtOAc, CH_2Cl_2, CCl_4 and CH_3CN were examined under various amounts of

TABLE III
Condensation of 2-aminothiophenol with aromatic/heteroaromatic aldehydes catalyzed by silica sulfuric acid (0.05 g) in CH_3CN

(Het)Ar **1a–1p**	Product[a]	Time, min	Yield, %[b]	M.p., °C	
				Found	Reported
Ph	**2a**	25	82	113–114	112–114[41]
2-$MeOC_6H_4$	**2b**	25	81	103–105	101–103[41]
4-$MeOC_6H_4$	**2c**	20	86	119–120	120–121[41]
2-HOC_6H_4	**2d**	20	78	127–128	127–128[17]
4-MeC_6H_4	**2e**	40	74	82–84	85[17]
2-MeC_6H_4	**2f**	50	70	53–55	53–54[17]
4-BrC_6H_4	**2g**	30	88	132–133	132[41]
3-BrC_6H_4	**2h**	20	82	82–83	83–84[41]
4-$Me_2NC_6H_4$	**2i**	30	87	157–159	160–161[41]
4-ClC_6H_4	**2j**	25	84	116–118	115–117[41]
4-FC_6H_4	**2k**	35	80	102–104	101–103[48]
4- CNC_6H_4	**2l**	10	96	161–162	162–164[20]
3-$NO_2C_6H_4$	**2m**	15	90	179–180	181–182[41]
Pyridyl-4-yl	**2n**	20	82	132–133	134–135[17]
Pyridyl-2-yl	**2o**	15	76	137–138	137–138[48]
Thienyl-2-yl	**2p**	15	80	99–100	99[17]

[a] All the products were characterized from spectral (^1H, ^{13}C NMR, MS and IR) data which were in accord with those of the literature cited. [b] Isolated yields.

the catalyst $AlCl_3 \cdot 6H_2O$. The best results in terms of the yields were obtained when 5 mole % of $AlCl_3 \cdot 6H_2O$ was used in MeOH/H_2O (20:1) as the solvent of choice and the reaction was run at room temperature for 30 min. These optimized reaction conditions were similarly extended to a series of other aldehydes and the corresponding products were isolated as given in Table IV. It is necessary to mention that, although the catalytic efficiency of $AlCl_3 \cdot 6H_2O$ as a nontoxic and inexpensive catalyst has already been explored in a variety of organic transformations, the use of $AlCl_3 \cdot 6H_2O$ in the present report to catalyze the synthesis of 2-arylbenzothiazoles is hitherto unreported.

TABLE IV
Condensation of 2-aminothiophenol with aromatic/heteroaromatic aldehydes catalyzed by $AlCl_3 \cdot 6H_2O$ (5 mole %) in MeOH/H_2O (20:1) at room temperature

(Het)Ar **1a–1p**	Product[a]	Time, min	Yield, %[b]	M.p., °C	
				Found	Reported
Ph	**2a**	30	90	111–112	112–114[41]
2-$MeOC_6H_4$	**2b**	30	76	103–105	101–103[41]
4-$MeOC_6H_4$	**2c**	20	92	119–120	120–121[41]
2-HOC_6H_4	**2d**	40	80	127–128	127–128[17]
4-MeC_6H_4	**2e**	20	84	84–85	85[17]
2-MeC_6H_4	**2f**	40	78	52–54	53–54[17]
4-BrC_6H_4	**2g**	30	92	132–133	132[41]
3-BrC_6H_4	**2h**	25	88	82–83	83–84[41]
4-$Me_2NC_6H_4$	**2i**	20	92	160–162	160–161[41]
4-ClC_6H_4	**2j**	30	86	116–118	115–117[41]
4-FC_6H_4	**2k**	40	83	102–104	101–103[48]
4- CNC_6H_4	**2l**	15	94	161–162	162–164[20]
3-$NO_2C_6H_4$	**2m**	20	89	179–180	181–182[41]
Pyridyl-4-yl	**2n**	50	74	134–136	134–135[17]
Pyridyl-2-yl	**2o**	60	68	136–138	137–138[48]
Thienyl-2-yl	**2p**	60	70	100–102	99[17]

[a] All the products were characterized from spectral (^1H, ^{13}C NMR, MS and IR) data which were in accord with those of the literature cited. [b] Isolated yield.

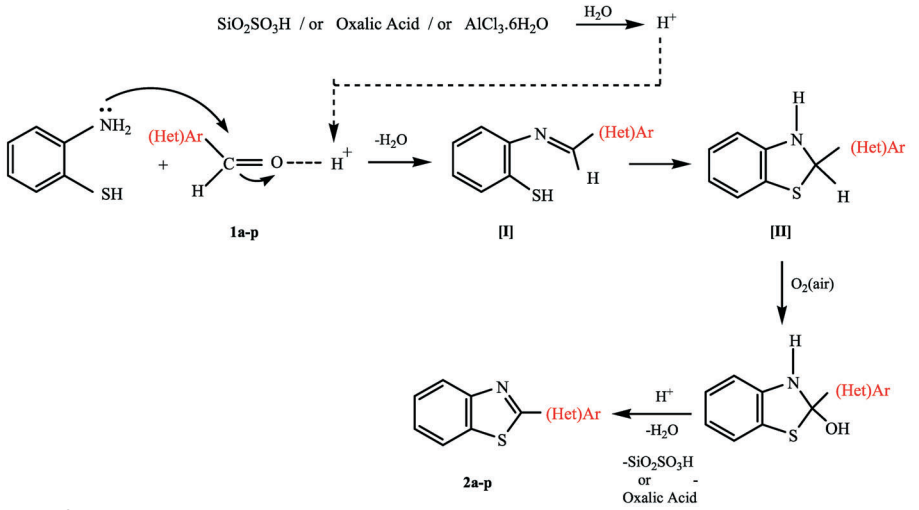
A comparison of the efficiency of this method with selected previous methods is collected in Table V. The results show that this method is superior to some previously reported methods in terms of yields and reaction times.

The presence of either silica sulfuric acid, oxalic acid or $AlCl_3 \cdot 6H_2O$ in the reaction as the catalyst can possibly bring about the generation of protons required to promote the reactions[51–58]. As previously suggested[37–50], the reaction may proceed through the formation of imine **I** from aldehydes and 2-aminothiophenol. Subsequent cyclization to 2,3-dihydrobenzothiazole **II** followed by oxidation in air would afford 2-arylbenzothiazoles (Scheme 2). It is important to mention that, when the same reaction with silica sulfuric acid, oxalic acid or $AlCl_3 \cdot 6H_2O$ was carried out under nitrogen atmosphere (in absence of oxygen), the reactions stopped at the benzothiazoline **II**

TABLE V
Comparison of methods for the synthesis of 2-arylbenzothiazoles **2**

Aldehyde	Conditions	Time	Yield, %
1c	$AlCl_3 \cdot 6H_2O$, MeOH/H_2O (20:1), r.t.	20 min	92
	Bakers' yeast, CH_2Cl_2, r.t.[47]	24 h	80
	CAN, MeOH, r.t.[38]	24 h	78
	[PmIm]Br, solvent-free, 80 °C [15]	6 h	90
	I_2, DMF, 100 °C [18]	30 min	71
	Dowex 50W, H_2O, 80 °C [41]	10 h	89
	H_2O_2, $Fe(NO_3)_3 \cdot 9H_2O$, solvent-free, 50 °C [44]	22 min	91
	MnO_2, SiO_2, solvent-free, MW oven, 140 °C [11]	4 min	91
1a	SSA, CH_3CN, 80 °C	25 min	82
	CAN, MeOH, r.t.[38]	24 h	75
	Activared carbon, O_2, Xylene, 50 °C [42]	3 h	79
	Bakers' yeast, CH_2Cl_2, r.t.[47]	24 h	75
	Dowex 50W, H_2O, 80 °C [41]	12 h	85
	[PmIm]Br, solvent-free, 80 °C [15]	6 h	80
1m	Oxalic acid, EtOH/H_2O (1:1), 80 °C	30 min	86
	P-TSA, H_2O, 80 °C [43]	300 min	84
	I_2, DMF, 100 °C [18]	35 min	78
	Bakers' yeast, CH_2Cl_2, r.t.[47]	24 h	71
	CAN, MeOH, r.t.[38]	24 h	88
	Dowex 50W, H_2O, 80 °C [41]	8 h	88

stage, which never proceeded to benzothiazoles **2a–2p**. This surely proves that aerial oxygen is not essential for benzothiazoline formation, though it is absolutely essential for the oxidation step leading to the formation of benzothiazoles.



SCHEME 2

CONCLUSION

In conclusion, an efficient one-pot procedure has been developed for the synthesis of 2-arylbenzothiazoles via condensation of 2-aminothiophenol with aldehydes catalyzed by either silica sulfuric acid (SSA), oxalic acid or $AlCl_3 \cdot 6H_2O$. Mild and simple experimental/product isolation procedure, green aspects avoiding hazardous solvents, use of eco-friendly and less toxic reagents, shorter reaction times and high yields of the products are the advantages of the present procedure.

EXPERIMENTAL

Solvents, reagents, and chemical materials were obtained from Aldrich (USA), Merck (Germany) and Fluka (Switzerland) chemical companies and purified prior to use. Melting points were determined in open capillary tubes in a BI Branstead Electrothermal Cat No. IA9200 apparatus and are uncorrected. Nuclear magnetic resonance spectra were recorded on JEOL FX 90Q using tetramethylsilane (TMS) as an internal standard. Infrared spectra were recorded on a PerkinElmer GX FT IR spectrometer (KBr pellets).

Synthesis of 2-Arylbenzothiazoles from Condensation of 2-Aminothiophenol with Aldehydes. Typical procedures

Synthesis with oxalic acid: 2-Aminothiophenol (1 mmol) and aromatic/heteroaromatic aldehydes **1a–1p** (1 mmol) were dissolved in EtOH/H_2O (1:1 v/v). Oxalic acid (10 mole %) was added to the above solution and the mixture was stirred at 80 °C until completion (TLC, ethyl acetate–hexane 2:8). After the complete conversion of the substrate, water (20 ml) was added and the mixture was allowed to stand at room temperature for 3–4 h. The formed crystals of the pure product were collected by filtration and dried. After isolation of the product, the filtrate was extracted with $CHCl_3$ (2 × 30 ml). The aqueous layer (including oxalic acid) was separated and the solvent was evaporated to obtain pure oxalic acid. The recycled catalyst was used for the next run under identical reaction conditions.

Synthesis with silica sulfuric acid: 2-Aminothiophenol (1 mmol) and aromatic/heteroaromatic aldehydes **1a–1p** (1 mmol) were dissolved in 10 ml of acetonitrile. Silica sulfuric acid (0.05 g) was added to the above solution at room tempetrature and the stirred mixture was refluxed. The progress of the reaction was monitored by TLC (hexane–ethyl acetate 8:2). After the complete conversion of the substrate as indicated by TLC analysis, the insoluble silica sulfuric acid was removed by filtration and washed with CH_3CN (2 × 5 ml). The recycled catalyst was used for the next run. The solvent was evaporated under reduced pressure to give the products **2a–2p**, which were recrystalized from EtOH (96%).

Synthesis with $AlCl_3{\cdot}6H_2O$: 2-Aminothiophenol (1 mmol) and aromatic/heteroaromatic aldehydes **1a–1p** (1 mmol) were dissolved in MeOH/H_2O (20:1 v/v). $AlCl_3{\cdot}6H_2O$ (5 mole %) was added to the above solution and the mixture was stirred at room temperature until completion (TLC, ethyl acetate–hexane 2:8). After completion, the solvent was evaporated and washed with water to give the crude products **2a–2p**. Then, the residue was recrystalized from (EtOH, 2 × 5 ml) to afford the pure product.

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