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Ultrasound mediated synthesis of aldimines from non-enolizable aldehydes at room temperature

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A user-friendly, energy efficient method of synthesizing *N*-sulfonyl aldimines at room temperature (25–28°C) from readily available and inexpensive reagents has been developed. The method employs ultrasonication from common laboratory ultrasonic cleaner to mediate the reaction. $FeCl_3$ as catalyst and molecular sieves as water scavenger have produced excellent yields of aldimines with short reaction time. The reaction set-up does not require inert atmosphere or anhydrous solvent thereby saving resources and time. The easily scalable method works well for the condensation of aromatic non-enolizable aldehydes and sulfonamides.

Keywords: Aldehyde, sulfonamide, *N*-sulfonyl aldimine, ultrasound, Lewis acid

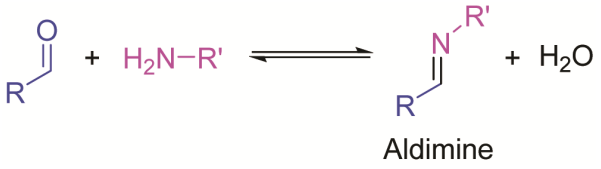
Imines formed from the condensation of an aldehyde with an amine are conventionally referred to as aldimines (Scheme I). In organic synthesis, aldimines finds its relevance as a synthetic intermediate that can generate a wide variety of nitrogen containing compounds upon addition, reduction or cyclization[1]. Aldimines are generally less reactive compared to the analogous aldehyde as a result an activating group is often tethered to the nitrogen atom to enhance its reactivity. In this regard, the use of sulfonyl moiety as an activating group has proven to be efficient. While there are several ways of synthesizing *N*-sulfonyl aldimines[1-8] but are commonlysynthesized via the condensation of an aldehyde with sulfonamide[9-17]. Owing to the reversible nature of the condensation reaction,continuous removal of water from the reaction mixture is necessary. As a result, harsh reaction conditions and long reaction times are often associated with the synthesis of *N*-sulfonyl aldimines. For instance, Love et al. developed a neat method of synthesizing aldimines which requires a high reaction temperature of above 150°C while employing $Si(OEt)_4$ as a water scavenger[18]. Varma et. al. reported a microwave mediated method to synthesize aldimines while employing a combination of several reagents viz. anhydrous $CaCO_3$,$CH(OCH_3)_3$ and K-10 clay[19]. A popular alternative of synthesizing aldimines is via the application of Dean-Stark assembly to continuously remove the water formed from the condensation reaction[2,20]. Long reaction time and high reaction temperature are among the limiting factors of this method employing Dean-Stark separation of water. Recently, an amino-catalytic process reported in 2014 offers an alternative method to synthesize aldimines in comparatively milder conditions at a reaction temperature of 60°C[21]. We envisaged that a user-friendly, energy efficient method of synthesizing aldimines at room temperature from readily available and inexpensive reagents can be a meaningful addition to the existing literature.

In this regard, we report a method of synthesizing *N*-sulfonyl aldimines from aldehydes and sulfonamides employing ultrasound at room temperature. Ultrasound mediated reactions are energy efficient and provides a complimentary route to synthesize valuable organic molecules[22,23]. The developed method exploits the heat generated from the numerous micro cavity implosions taking place when ultrasound is propagated through the reaction medium. It may be noted that, these micro hotspots have shown temperatures higher than 1000 K but heat dissipation throughout the bulk keeps the overall temperature largely unchanged[24,25]. The reaction in essence is carried out at room temperature under the presence of $FeCl_3$ as the catalyst. The method also employs activated molecular sieves to absorb the water released from the condensation reaction. The ultrasound mediated

reaction produces excellent yields of *N*-sulfonyl aldimines in short reaction time. The reaction works well with aromatic non-enolizable aldehydes and sulfonamides.

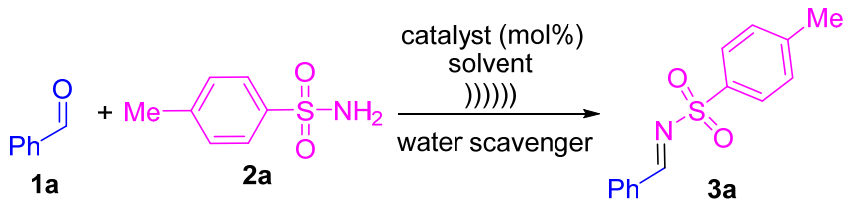
Results and Discussion

We considered benzaldehyde **1a** and *p*-tolunesulfonamide **2a** as our model reactants for optimizing the reaction conditions. It may be noted here that all the reactions are carried out using an ultrasonic cleaning bath in 1.0 mmol scale in a 5 mL round bottom flask sealed with a septum. The reaction set-up employs a meagre 0.5 mL of the solvent as well as powdered and activated molecular sieves (MS 4Å) to aid in homogeneity and efficiency, respectively. The bath temperature was maintained between 25°C to 28°C by periodically changing the water of ultrasonic cleaning bath.Using this reaction set-up, we tested several commercially available acid catalysts for the activation of the aldehyde. We observed that the use of anhydrous $FeCl_3$ as the catalyst for the condensation reaction yields 94% yield of **3a** in 3 h, while $SnCl_2.H_2O$, $BiCl_3$ and$ZnCl_2$ prove to be comparably inefficient (Table I, entries 1–4). We also noted that Brønsted acid *p*-



Scheme I — Synthesis of aldimine via the condensation of aldehyde and amine

Table I — Optimisation of reaction conditions[a]



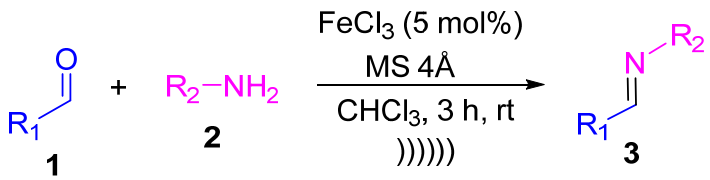
Entry	Catalyst	Solvent	Water Scavenger	Time (h)	Yield (%)[b]
1	$FeCl_3$	$CHCl_3$	MS 4Å	3	94
2	$SnCl_2.H_2O$	$CHCl_3$	MS 4Å	3	64
3	$BiCl_3$	$CHCl_3$	MS 4Å	3	68
4	$ZnCl_2$	$CHCl_3$	MS 4Å	3	43
5	PTSA	$CHCl_3$	MS 4Å	3	90
6	TfOH	$CHCl_3$	MS 4Å	3	82
7[c]	$FeCl_3$	$CHCl_3$	MS 4Å	3	95
8	$FeCl_3$	DCM	MS 4Å	3	84
9	$FeCl_3$	DCE	MS 4Å	3	79
10	$FeCl_3$	THF	MS 4Å	3	56
11	$FeCl_3$	MeCN	MS 4Å	3	68
12	$FeCl_3$	Toluene	MS 4Å	3	NR
13	$FeCl_3$	EtOH	MS 4Å	3	NR
14	$FeCl_3$	^{i}PrOH	MS 4Å	3	NR
15[d]	$FeCl_3$	$CHCl_3$	MS 4Å	3	81
16[e]	$FeCl_3$	$CHCl_3$	MS 4Å	3	95
17	$FeCl_3$	$CHCl_3$	$CaCl_2$(2 mmol)	3	64
18	$FeCl_3$	$CHCl_3$	P_2O_5(2 mmol)	0.5	97
19[e]	$FeCl_3$	$CHCl_3$	MS 4Å	0.5	60
20	$FeCl_3$	$CHCl_3$	MS 4Å	2	82
21[f]	$FeCl_3$	$CHCl_3$	MS 4Å	3	62
22[g]	$FeCl_3$	$CHCl_3$	MS 4Å	3	94
23[h]	$FeCl_3$	$CHCl_3$	MS 4Å	3	92

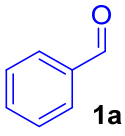
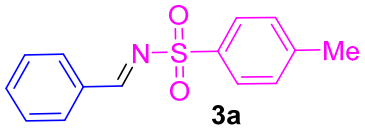
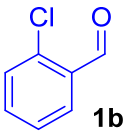
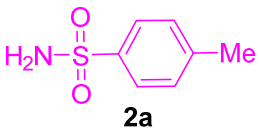
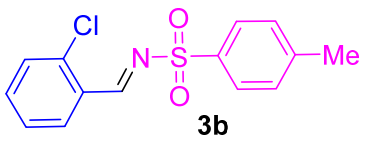
[a]Reaction conditions: **1a** (1.1 mmol), **2a** (1 mmol), solvent 0.5 mL, catalyst (5 mol%), MS 4Å (200 mg), sealed 5mL round bottom flask; ultrasonic cleaner (US-1) having frequency of 40 kHz and power of 50 W, temperature25–28°C.[b]Isolated yields; [c]catalyst (10 mol%); [d]MS 4Å (100 mg); [e]MS 4Å (400 mg); [f]open vessel reaction; [g]reaction under inert atmosphere using dry $CHCl_3$; [h]ultrasonic cleaner (US-2) having a frequency of 33 kHz and power of 100 W. NR = no reaction.

toluenesulfonic acid (PTSA) as the catalyst also produces very good yield of **3a** (Table I, entry 5). A catalyst loading of 5 mol% $FeCl_3$was found to be optimum (Table I, entries 1, 7). With $FeCl_3$ proving to be an efficient catalyst, we evaluated the effect of solvents on the condensation reaction. The use of DCM and DCE as the reaction media produced 84% and 79% yields, respectively (Table I, entries 8–9). The higher polarity of $CHCl_3$ in comparison to DCM and DCE contributes to better solubility of *p*-tolunesulfonamide **2a** which supports the progress of the reaction. Other solvents that were screened did not prove to be competent (Table I, entries 10–14). It was surprising to us when we did not obtain any desired aldimine formation in EtOH (Table I, entry 13). Although, in a recent report, Darcel et al. reported aldimine formation in good yield using $FeCl_3$ catalyst at room temperature without sonication.[11]The reason behind this observation is still unknown to us. We studied the role of water scavenger in this reversible condensation reaction and recognized its pivotal role. When we decreased the amount of MS 4Åadded in the reaction from 200 mg/mmol to 100 mg/mmol, we observed that the reaction produces lower yield in identical reaction conditions (Table I, entries 1, 15). However, increasing the amount of MS 4Å to 400 mg/mmol had no noticeable effect on the yield of the reaction after 3 h (Table I, entry 16). The use of anhydrous $CaCl_2$ as the water scavenger did not offer satisfactory result (Table I, entry 17). However, when we used P_2O_5 as the water scavenger, the reaction completed in 30 min producing an excellent 97% yield of the aldimines (Table I, entry 18). Considering the factors such as ease of handling, reusability, environmental impact, and purification, we selected 200 mg/mmol MS 4Å over P_2O_5 as the water scavenger for the synthesis of aldimines. The reaction produced only 60% yield of the aldimine**3a** in 30 min and increased to 82% yield after 2 h after higher loading of MS 4Å was employed in a bid to reduce the reaction time (Table I, entries 19–20). To study the role of the reaction set-up, we carried the reaction in open vessel allowing for the exchange of atmosphere. We observed a sharp decline in the efficiency of the reaction as only 62% yield of the aldimine **3a** was isolated after 3 h (Table I, entry 21). On the contrary, the use of inert atmosphere and anhydrous solvent for the reaction had no discernible impact on the yield (Table I, entry 22). This can be understood as the reactions are carried out in small and sealed round bottom flask where the residual moisture (from air and solvent) is trapped by the activated molecular sieves. The change of frequency and intensity of ultrasound did not significantly impact the progress of the reaction (Table I, entry 23). It may be pointed out that the powdered molecular sieves (MS 4Å) used in the condensation reaction is recovered via filtration, washed, and reused after reactivation.

After optimising the reaction conditions, we tested the scope of the reaction with several aldehydes and sulfonamides. Aromatic aldehydes consisting of electron-withdrawing substituents produced higher yield of the corresponding aldimine (Table II, entry \

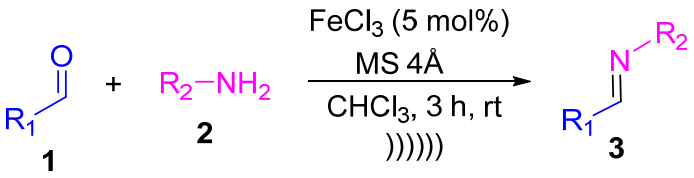
Table II — Substrate scope of the ultrasound-mediated method[a]

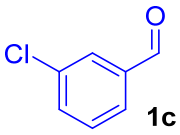
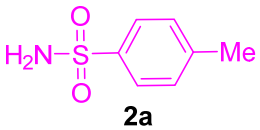
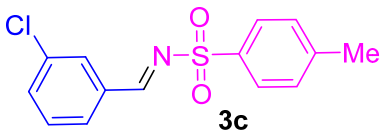
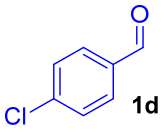
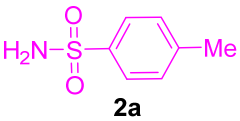
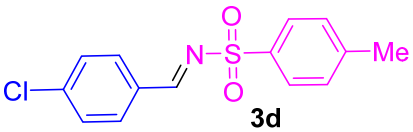
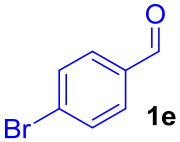
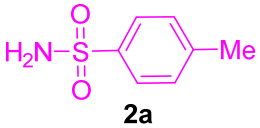
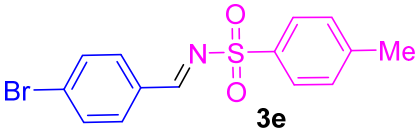
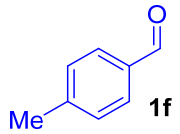
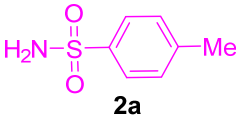
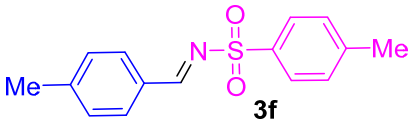
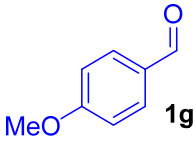
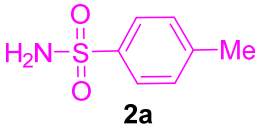
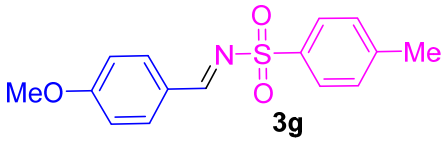
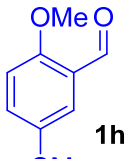
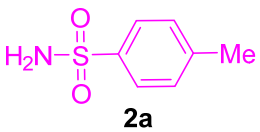
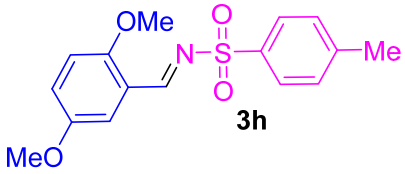
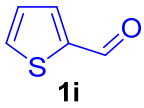
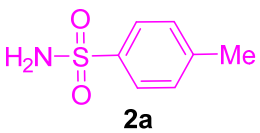
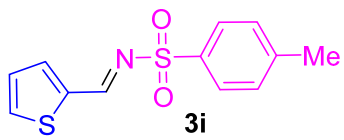
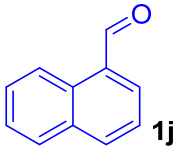
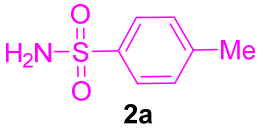
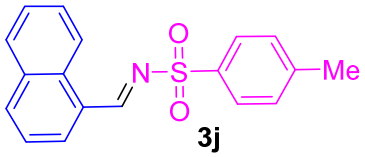
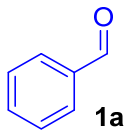
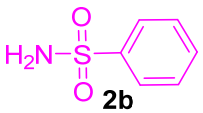
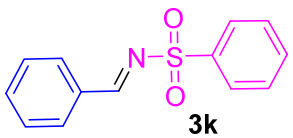


Entry	Aldehyde **1**	Substrate **2**	Aldimine **3**	Yield (%)[b]
1	 **1a**	 **2a**	 **3a**	94
2	 **1b**	 **2a**	 **3b**	95

(*Contd.*)

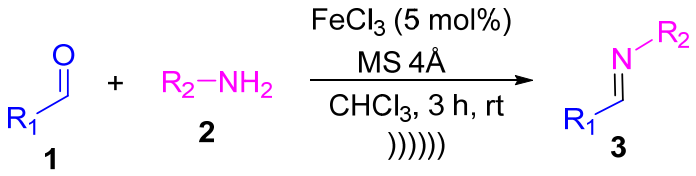
Table II — Substrate scope of the ultrasound-mediated method[a]

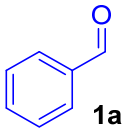
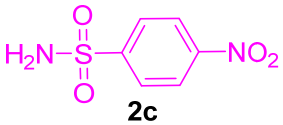
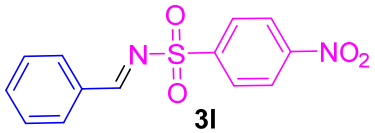
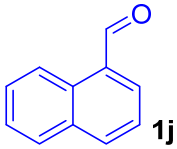
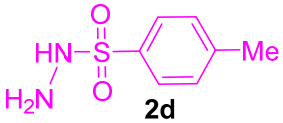
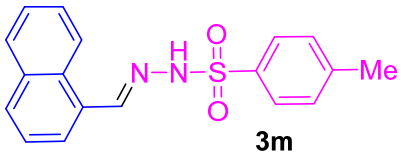
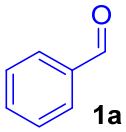
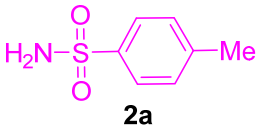
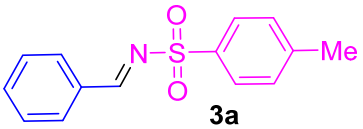
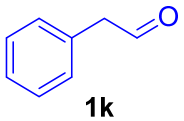
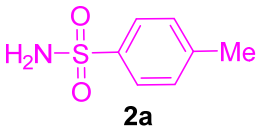
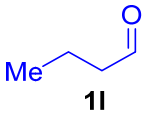
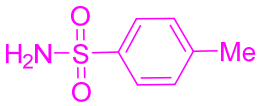
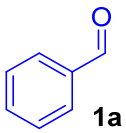
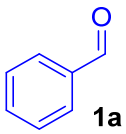
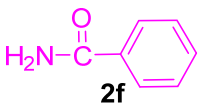


Entry	Aldehyde **1**	Substrate **2**	Aldimine **3**	Yield (%)[b]
3	 **1c**	 **2a**	 **3c**	96
4	 **1d**	 **2a**	 **3d**	95
5	 **1e**	 **2a**	 **3e**	93
6	 **1f**	 **2a**	 **3f**	89
7	 **1g**	 **2a**	 **3g**	86
8	 **1h**	 **2a**	 **3h**	82
9	 **1i**	 **2a**	 **3i**	88
10	 **1j**	 **2a**	 **3j**	96
11	 **1a**	 **2b**	 **3k**	96

(*Contd.*)

Table II — Substrate scope of the ultrasound-mediated method[a]



Entry	Aldehyde **1**	Substrate **2**	Aldimine **3**	Yield (%)[b]
12	 **1a**	 **2c**	 **3l**	92
13	 **1j**	 **2d**	 **3m**	94
14[c]	 **1a**	 **2a**	 **3a**	94
15	 **1k**	 **2a**	No reaction	NR
16	 **1l**	 **2a**	No reaction	NR
17	 **1a**	 **2e**	No reaction	NR
18	 **1a**	 **2f**	No reaction	NR

[a]Reaction conditions: **1a** (1.1 mmol), **2a** (1 mmol), $CHCl_3$(0.5 mL), $FeCl_3$ (5 mol%), MS 4Å (200 mg), sealed 5 mL RB flask, 3 h, temperature25–28°C. ultrasonic cleaner (US-1) having frequency of 40 kHz and power of 50 W;[b]Isolated yields; [c]Reaction conditions: **1a** (22 mmol), **2a** (20 mmol), $CHCl_3$(10 mL), $FeCl_3$ (5 mol%), MS 4Å (4 g), sealed 25 mL RB flask, 3h, temperature25–28°C. ultrasonic cleaner (US-1) having frequency of 40 kHz and power of 50 W. NR = no reaction.

2–5). The presence of -Cl substituent on *ortho*-, *meta*- or *para*-position did not impede the efficiency of the reaction (Table II, entries 2–4).Aromatic aldehydes consisting of electron-donating substituents caused a slight decline in the product yield (Table II, entries 6–8). Aldehyde with a heteroaromatic substituent viz. 2-thienyl and with polycyclic aromatic substituent viz. 1-naphthyl also produced excellent yield of the corresponding aldimine (Table II, entries 9–10). The changes in the substituents of sulfonamide had little effect on the efficiency of the reaction (Table II, entries 11–12). The method can also be applied for the condensation of aromatic aldehyde **1j** with *p*-toluenesulfonyl hydrazide **2d** for the synthesis of 4-methyl-*N*'-(naphthalen-1-ylmethylene) benzene-sulfonohydrazide**3m** in 94% yield (Table II, entry 13).

A method allowing the synthesis of aldimines in large scale is desirable as aldimines often serve as starting materials in multi-step organic synthesis of nitrogen-containing moieties. To show the scalability of the developed method, the reaction was carried out in a larger scale (20 mmol) which produced 94% yield of the aldimine (Table II, entry 14). The developed method fails when aliphatic aldehydes or enolizable aldehydes are used for the condensation reaction (Table II, entries 15–16). It needs to be pointed out that the use of 4-nitroaniline **2e** or benzylamide **2f** did not give the condensation reaction (Table II, entries 17–18).

It may be mentioned that at large scale, the amount of water formed as a by-product of the condensation reaction is significant enough to cause in-situ hydrolysis of the aldimine. This phenomenon may cause severe decline in the efficiency of the reaction at large scale unless the water by-product is continuously removed from the reaction mixture. Our method employs powdered MS 4Å to trap the water formed from condensation reaction thereby maintaining the efficiency of the method at large scale. In recent times, several methods have been reported for the synthesis of aldimines from the condensation of aldehydes with sulphonamides (Table III) but their efficiency at large scales were not tested.

Experimental Section

All reagents and solvents are of AR grade and were purchased from SRL, Alfa Aesar,Spectrochem, Merck Pvt. Ltd. Aldehydes were purified before use. All the reactions were done in oven-dried glass apparatus in an air atmosphere. Ultrasound irradiation was supplied using a Citizon Ultrasonic cleaner (US-1) having a frequency of 40 kHz and power of 50 W or a BIOCRAFT ultrasonic cleaner (US-2) having a frequency of 33 kHz and power of 100 W. Molecular sieves (4Å) was activated by heating the powdered molecular sieves at about 120°C under high vacuum for overnight followed by cooling it under argon atmosphere to eliminate the moisture. The activated MS 4Å was stored under argon atmosphere.Reaction vessel was dipped in the sonicator bath such that the surface of the reaction mixture is lower than the level of the water in the bath. The bath temperatures were maintained between 25°C to 28°C by periodically changing the water. Reactions were monitored by thin-layer chromatography on silica gel 60 F254 using UV light and Iodine absorption. Melting points reported are uncorrected. ^{1}H and ^{13}C NMR spectra were measured on a BrukerAvance II (^{1}H NMR: 400 MHz) spectrometer. Chemical shifts are reported in δ (ppm) from tetramethylsilane (TMS), with the solvent resonance as the internal standard (unless otherwise mentioned, chloroform: δ 7.26 ppm). Data are reported as follows: chemical shifts, multiplicity (s=singlet, d=doublet, t=triplet, q=quartet, br=broad, dd=double doublet, m=multiplet), coupling constant (in Hz), integration. The spectra obtained for aldimines match well with the reported data.

General reaction procedure for reaction optimization

A 5 mL round-bottom flask equipped with a rubber septum (rubber septum not used for entry 21, Table I) was charged with **1a** (1.1 mmol), **2a** (1 mmol), solvent (0.5 mL), water scavenger (amount as specified in Table I) and catalyst (amount as specified in Table I) in an air atmosphere (N_2 atmosphere for entry 22, Table I). The flask was irradiated in an ultrasonic cleaner (US-1 for entries 1-22; US-2 for entry 23, Table I) at 25–28°C and the progress of the reaction was monitored by TLC. After the time as specified in Table I, the reaction mixture was passed through a short bed of silica under vacuum. The silica bed was washed with EtOAc (5 mL×3) and the filtrate

Table III — A comparison of the present method with the literature* for the one-step condensation of **1a** and **2a**

Entry	Reagents and solvent	Temp (°C)	Time	Isolated yield (%) (Scale of reaction)	Ref. (Year)
1.	$ZrO_2/S_2O_8^{2-}$solid superacid, solvent-free	MW, 650 W	6 min	92 (2 mmol)	16 (2004)
2.	Montmorillonite K-10, toluene	114	50 min	93 (1 mmol)	27 (2006)
3.	$AlCl_3$, solvent-free	RT	15 min	97 (1 mmol)	12 (2007)
4.	P_2O_5/SiO_2, solvent-free	110	2 h	91 (1 mmol)	17 (2008)
5.	Zeolite HY9, CH_2Cl_2, Dean-Stark, MS 4Å	40	18 h	87.4 (3.5 mmol)	10 (2008)
6.	Cyanuric chloride, neat	110	2 h	92 (1 mmol)	15 (2010)
7.	WCl_6, solvent-free	100	45 min	88 (1 mmol)	28 (2013)
8.	Pyrrolidine, CH_2Cl_2, MS 4Å	60	24 h	99 (1.2 mmol)	21 (2014)
9.	[Dsim]$AlCl_4$, solvent-free	100	4 min	92 (1 mmol)	29 (2014)
10.	$FeCl_3$, $CHCl_3$, MS 4Å	MW, 50 W, RT	3 h	94 (20 mmol)	Present method

*This is a representative list of the methods present in the literature and does not mention all known reports.

was concentrated to obtain the crude product which is purified via crystallization (from 2% EtOAc/Hex).

General procedure for the synthesis of product 3

A 5 mL round-bottom flask equipped with a rubber septum was charged with **1** (1.1 mmol), **2** (1 mmol), $CHCl_3$ (0.5 mL), MS 4Å (200 mg) and $FeCl_3$ (5mol%) in an air atmosphere. The flask was irradiated in an ultrasonic cleaner at 25–28°C and the progress of the reaction was monitored by TLC. After 3 h, the reaction mixture was passed through a short bed of silica under vacuum. The silica bed was washed with EtOAc (5 mL× 3) and the filtrate was concentrated to obtain the crude product which is purified via crystallization.

Analytical Data

N-Benzylidene-4-methylbenzenesulfonamide, 3a:[11,12] White solid. m.p.112°C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.04 (s, 1H), 7.91 (dd, *J* = 15.3 and 8.3 Hz, 3H), 7.82 (d, *J* = 8.0 Hz, 2H), 7.62 (t, *J* = 7.5 Hz, 1H), 7.32 (d, *J* = 8.0 Hz, 2H), 2.43 (s, 3H).

N-(2-Chlorobenzylidene)-4-methylbenzenesulfonamide, 3b[12,26]: White solid. m.p.128°C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.50 (s, 1H), 8.15 (dd, *J* = 7.9 and 1.4 Hz, 1H), 7.90 (d, *J* = 8.2 Hz, 2H), 7.55–7.50 (m, 1H), 7.46 (d, *J* = 7.2 Hz, 1H), 7.38–7.31 (m, 3H), 2.45 (s, 3H).

N-(3-Chlorobenzylidene)-4-methylbenzenesulfonamide, 3c[12,27]: White solid. m.p.94°C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.98 (s, 1H), 7.95–7.93 (m, 1H), 7.88 (d, *J* = 8.3 Hz, 2H), 7.78 (d, *J* = 7.6 Hz, 1H), 7.60–7.55 (m, 1H), 7.44 (t, *J* = 7.9 Hz, 1H), 7.36 (d, *J* = 8.2 Hz, 2H), 2.45 (s, 3H).

N-(4-Chlorobenzylidene)-4-methylbenzenesulfonamide, 3d:[11,12] White solid. m.p.172°C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.99 (s, 1H), 7.88 (t, *J* = 8.1 Hz, 4H), 7.47 (d, *J* = 8.4 Hz, 2H), 7.36 (d, *J* = 8.1 Hz, 2H), 2.45 (s, 3H).

N-(4-Bromobenzylidene)-4-methylbenzenesulfonamide, 3e:[11,12] Light brown solid. m.p.182°C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.97 (s, 1H), 7.88 (d, *J* = 8.2 Hz, 2H), 7.78 (d, *J* = 8.5 Hz, 2H), 7.62 (d, *J* = 8.4 Hz, 2H), 7.35 (d, *J* = 8.1 Hz, 2H), 2.43 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$): δ 168.83, 144.84, 132.60, 132.42, 131.20, 130.28, 129.89, 128.16, 77.38, 77.06, 76.74, 21.70.

4-Methyl-N-(4-methylbenzylidene) benzenesulfonamide, 3f[12,27]: White solid. m.p.111°C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.99 (s, 1H), 7.89 (d, *J* = 8.2 Hz, 2H), 7.82 (d, *J* = 8.1 Hz, 2H), 7.35 (d, *J* = 8.3 Hz, 2H), 7.29 (d, *J* = 8.1 Hz, 2H), 2.44 (s, 6H).

N-(4-Methoxybenzylidene)-4-methylbenzenesulfonamide, 3g[10,11]: White solid. m.p.127°C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.09 (s, 1H), 8.03 (dd, *J* = 8.6 and 2.6 Hz, 4H), 7.48 (d, *J* = 8.1 Hz, 2H), 7.11 (d, *J* = 8.8 Hz, 2H), 4.03 (s, 3H), 2.57 (s, 3H).

N-(2,5-Dimethoxybenzylidene)-4-methylbenzenesulfonamide, 3h[12,28]: White solid. m.p.124°C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.50 (s, 1H), 7.88 (d, *J* = 8.2 Hz, 2H), 7.50 (d, *J* = 3.2 Hz, 1H), 7.33 (d, *J* = 8.1 Hz, 2H), 7.14 (dd, *J* = 9.1 and 3.2 Hz, 1H), 6.90 (d, *J* = 9.1 Hz, 1H), 3.87 (s, 3H), 3.76 (s, 3H), 2.43 (s, 3H).

4-Methyl-N-(thiophen-2-ylmethylene)benzenesulfonamide, 3i[11,12,29]: Light brown solid. m.p.104°C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.11 (s, 1H), 7.87 (d, *J* = 8.2 Hz, 2H), 7.78 (d, *J* = 4.2 Hz, 2H), 7.34 (d, *J* = 8.2 Hz, 2H), 7.21 (t, *J* = 4.3 Hz, 1H), 2.44 (s, 3H).

4-Methyl-N-(naphthalen-1-ylmethylene) benzenesulfonamide, 3j[10,11]: White solid. m.p.196°C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.60 (s, 1H), 8.99 (d, *J* = 8.5 Hz, 1H), 8.14 (d, *J* = 7.2 Hz, 1H), 8.09 (d, *J* = 8.2 Hz, 1H), 7.96 (d, *J* = 8.2 Hz, 2H), 7.91 (d, *J* = 8.1 Hz, 1H), 7.67 (dd, *J* = 11.3, and 4.1 Hz, 1H), 7.57 (dd, *J* = 16.2 and 8.5 Hz, 2H), 2.43 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$): δ 169.84, 144.57, 136.18, 135.23, 133.78, 131.81, 129.87, 129.09, 128.95, 128.06, 126.98, 125.13, 124.26, 21.68.

N-Benzylidene-4-nitrobenzenesulfonamide, 3k[30]: Light yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.14 (s, 1H), 8.40 (d, *J* = 8.8 Hz, 2H), 8.22 (d, *J* = 8.7 Hz, 2H), 7.96 (d, *J* = 7.5 Hz, 2H), 7.68 (t, *J* = 7.4 Hz, 1H), 7.54 (d, *J* = 7.7 Hz, 2H).

N-Benzylidenebenzenesulfonamide, 3l:[12,26] White solid. m.p.78°C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 9.07 (s, 1H), 8.02 (d, *J* = 7.8 Hz, 2H), 7.94 (d, *J* = 7.8 Hz, 4H), 7.55–7.49 (m, 4H).

4-Methyl-N'-(naphthalen-1-ylmethylene) benzenesulfonohydrazide, 3m[31]: White solid. ^{1}H NMR (400 MHz, $CDCl_3$,DMSO-d_6): δ 9.47 (s, 1H), 9.01 (d, *J* = 7.9 Hz, 1H), 8.80 (s, 1H), 8.15 (d, *J* = 7.1 Hz, 1H), 8.06–7.93 (m, 2H), 7.75–7.71 (m, 3H), 7.63 (d, *J* = 6.5 Hz, 1H), 7.44 (s, 1H), 7.28 (s, 2H), 2.43 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$, DMSO-d_6): δ 161.48, 143.16, 131.32, 128.78, 128.70, 128.24, 127.78, 126.86, 125.74, 124.80, 124.00, 21.00.

Conclusion

In conclusion we have developed a user-friendly, energy efficient and rapid method for the synthesis of

aldimines at room temperature(25–28°C). The method uses the local hotspots as the source of energy created in the reaction medium by the propagation of ultrasound. The thoughtfully designed reaction set-up does not require the use of inert atmosphere or pre-drying of reaction solvent saving time and resources. The method does not lose its efficiency at large scale. We believe that the ultrasound mediated, room temperature synthesis of aldimines will be an important addition to the existing literature. Further studies in a bid to broaden the scope of the reaction is planned to undergo in our laboratory.

Supporting Information

^{1}H and ^{13}C NMR spectra of all products are available in the electronic supporting information.

Conflict of interest

The authors declare no conflict of interest.

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References

1 Ruano J L G, Alemán J, Cid M B & Parra A, *Org Lett*, 7 (2005) 179.
2 Jin T-S, Feng G-L, Yang M-N & Li T-S, *J Chem Res*, (2003) 591.
3 Huang D, Wang X, Wang X, Chen W, Wang X & Hu Y, *Org Lett*, 18 (2016) 604.
4 Li Z, Ren X, Wei P, Wan H, Shi Y & Ouyang P, *Green Chem*, 8 (2006) 433.
5 Lee K Y, Lee C G & Kim J N, *Tetrahedron Lett*, 44 (2003) 1231.
6 Chemla F, Hebbe V & Normant J-F, *Synthesis*, 75 (2000).
7 Trost B M & Marrs C, *J Org Chem*, 56 (1991) 6468.
8 Boufas W, Belhani B, Cheloufi H, K'tir H, Aouf N-E & Berredjem M, *J Chem Pharm Res*, 6 (2014) 876.
9 Wynne J H, Price S E, Rorer J R & Stalick W M, *Synth Commun*, 33 (2003) 341.
10 Wang K, Xing Z, Ma Y & Wang Q, *Catal Lett*, 123 (2008) 129.
11 Wu X-F, Vovard-Le Bray C, Bechki L & Darcel C, *Tetrahedron*, 65 (2009) 7380.
12 Sharghi H, Hosseini-Sarvari M & Ebrahimpourmoghaddam S, *Arkivoc*, 15 (2007) 255.
13 Deng G S, Zou J Y & Sun T F, *Chin Chem Lett*, 22 (2011) 511.
14 Wu L, Cui T, Ma W & Yan F, *Asian J Chem*, 22 (2010) 8209.
15 Wu L, Yang X, Wang X & Yan F, *J Sulfur Chem*, 31 (2010) 509.
16 Jin T, Feng G, Yang M & Li T, *Synth Commun*, 34 (2004) 1277.
17 Hasaninejad A, Zare A, Sharghi H & Shekouhy M, *Arkivoc*, 2008 (2008) 64.
18 Love B E, Raje P S & Williams T, *Synlett*, 493 (1994).
19 Vass A, Dudás J & Varma R S, *Tetrahedron Lett*, 40 (1999) 4951.
20 Lu K, Kwon O, Brummond K M & DavisM M, *Org Synth*, 86 (2009) 212.
21 Morales S, Guijarro F G, Ruano J L G & Cid M B, *J Am Chem Soc*,136 (2014) 1082.
22 Grib I, Bouzina A, Aouf N-E & Berredjem M, *Phosphorus Sulfur Silicon Relat Elem*, 191 (2016) 1086.
23 Paul D, Borah A, Khatua S & Chatterjee P N, *Asian J Org Chem*, 8 (2019) 1870.
24 Canselier J, Delmas H, Wilhelm A & Abismail B, *J Dispersion Sci Technol*, 23 (2002) 333.
25 Okouchi S, Nojima O & Arai T, *Water Sci Technol*, 26 (1992) 2053.
26 Fan R, Pu D, Wen F, Ye Y & Wang X, *J Org Chem*, 73 (2008) 3623.
27 Jin T S, Yu M J, Liu L B, Zhao Y & Li T S, *Synth Commun*, 36 (2006) 2339.
28 Aliá Zolfigol M & Rezaá Moosavi-Zare A, *RSC Adv*, 3 (2013) 7692.
29 Moosavi-Zare A R, Zolfigol M A, Noroozizadeh E, Khakyzadeh V, Zare A & Tavasoli M, *Phosphorus Sulfur Silicon Relat Elem*, 189 (2014) 149.
30 Jia Y-X, Xie J-H, Duan H-F, Wang L-X & Zhou Q-L, *Org Lett*, 8 (2006) 1621.
31 Creary X, Tam W, Albizati K & Stevens R, *Org Synth*, 64 (1986) 207.