Research Article

A Facile Synthesis of *N*-H- and *N*-Substituted Acridine-1,8-diones under Sonic Condition

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Synthesis of an assembly of structurally important *N*-H- and *N*-substituted acridine-1,8-diones by CAN (ceric ammonium nitrate) catalysed one-pot four-component reaction of electron-deficient and electron-rich aromatic aldehydes and aromatic amines or ammonium acetate and dimedone or cyclohexyl-1,3-diones at 26°C under sonic condition is reported. The method is clean and energy efficient as it uses a greener method and an eco-friendly catalyst.

1. Introduction

Acridine and its derivatives are important structural motifs possessing antimalarial, antiviral, and antiallergic properties [1–3]; acridines act as potent drugs for antitumor activity both in vitro and in vivo against a range of murine and human tumors [4]. They are also found to act as fluorescent molecular probes for monitoring polymerization processes [5] and are used as *n*-type semiconductors and in the electroluminescent devices. Recently fluorinated acridones are reported to possess anticancer activity [6–9]. There are a few reports in the literature on the three-component Hantzsch-type condensation of aromatic aldehydes, anilines, and dimedone *via* traditional heating in organic solvents [10, 11], under microwave irradiation [12], and in ionic liquids [13]. The main drawbacks of these methods are the inability to synthesize profuse quantity of acridines using substituted anilines containing electron withdrawing groups [14]. Further, the reactions are carried out in refluxing organic solvents, which require higher temperature and longer hours for completion [10, 15] and unusual breaking of C–N bond takes place under certain reaction conditions as noticed in a few cases [16]. Hence, the exploration of a simple, efficient, and green method for the synthesis of acridines using electron-deficient amines and electron-deficient aldehydes is of current interest. In continuation with our work on one-pot multicomponent reactions under sonic condition [17–19], we, herein, report the synthesis of a series of acridines by a one-pot four-component reaction as shown in Schemes 1 and 2. To the best of our knowledge, the synthesis of acridines from fluorinated aromatic amines and heterocyclic amines using an inexpensive catalyst under sonic condition is not reported yet.

Sonic reactions are viewed as green and contemporary methods in synthetic organic chemistry. Sonochemical reactions are classified into three types based on their chemical effects induced by cavitation; they are homogeneous sonochemistry of liquids, heterogeneous sonochemistry of liquid-liquid or solid-liquid, and sonocatalysis (which overlaps the first two) and other mechanical effects. Study of chemical reactions under the mechanical effects sonic conditions is also called "false sonochemistry" which is also an important component of regular sonochemistry. In addition, the use of an inexpensive and versatile catalyst, CAN, in conjunction with ultrasound is considered to be economical and beneficiary in organic synthesis [20].

2. Methods

2.1. Materials and Instruments. All starting materials were commercial products and were used without further purification except liquid aldehydes and amines which were distilled before use. Melting points were measured on a Raaga make melting point apparatus. Nuclear magnetic resonance spectra were obtained on 400 MHz and 100 MHz Bruker AMX
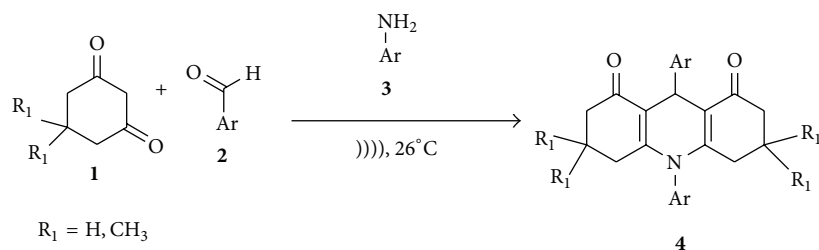
Scheme 1: Synthesis of acridines under sonic condition.
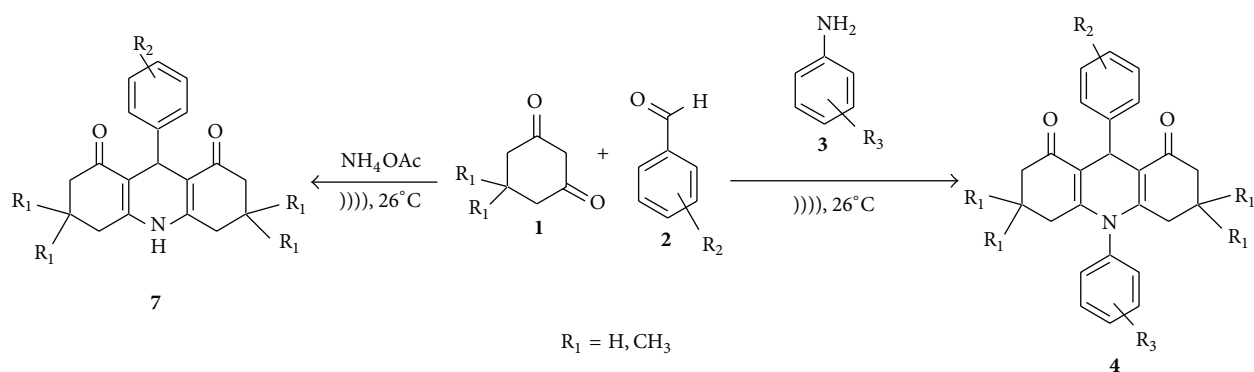
Scheme 2: Synthesis of *N*-H-acridines (**7**) and *N*-substituted acridines (**4**).

instruments in CDCl$_3$ using TMS as a standard. ESI-Mass spectra were recorded using ESI-Q TOF instrument. All the reactions were carried out using SIDILU make sonic bath working at 35 kHz (constant frequency: 120 W) maintained at 26°C by circulating water without mechanical stirring. Yields refer to yield of the isolated products.

2.2. Typical Procedure for the Synthesis of Acridines. Dimedone/cyclohexa-1,3-dione (2 mmol), aromatic aldehyde (1 mmol), and substituted aniline (1 mmol) were taken in acetonitrile (0.5 mL) and mixed well, and to this CAN (5 mol%) was added. The reaction was subjected to ultrasonic irradiation in a bath working at 26°C (35 kHz). The course of the reaction was monitored on TLC (3 : 7: EtOAc : hexane). After completion of the reaction, water (10 mL) was added and the separated solid was filtered. The crude product was then subjected to silica gel column chromatography (3 : 7: EtOAc : hexane) to get the pure products. The same procedure was followed for the reactions carried out with ammonium acetate or 2-aminopyridine.

2.3. Spectral Data

   4a: ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.50–7.45 (m, 1H, ArH); 7.41–7.38 (m, 1H, ArH); 5.15 (s, 1H); 3.74 (s, 3H, OCH$_3$); 2.44 (s, 2H); 2.25–2.15 (m, 4H); 2.14–1.96 (m, 1H); 0.98 (s, 3H, CH$_3$); 0.93 (s, 3H, CH$_3$); 0.84 (s, 3H, CH$_3$).

   ^{13}C NMR (100 MHz, CDCl$_3$): δ 194.9 (C=O), 163.2 (Ph–C–F), 135–116 (Ph), 59.2 (OCH$_3$), 49.8 (CH$_2$), 40.1 (CH$_2$), 32.6 (CH), 25.9 [C(CH$_3$)$_2$], ESI-MS: 507.2 (507.2). M.p: 265–267°C.

4b: ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.39–7.37 (m, 1H, ArH); 7.28–7.20 (m, 4H, ArH); 7.19–1.17 (m, 1H, ArH); 7.05–7.02 (m, 2H, ArH); 6.99–6.83 (m, 1H, ArH); 5.6 (s, 1H); 2.25–2.17 (m, 4H); 2.06–2.03 (m, 2H); 1.83–1.62 (m, 4H); 0.99–0.88 (m, 12H, 4CH$_3$).

^{13}C NMR (100 MHz, CDCl$_3$): δ 197.0 (C=O), 163.4 (Ph–C–F), 161.2 (Ph–C–F), 135–116 (Ph), 49.7 (CH$_2$–CO), 40.2 (CH$_2$–C), 32.5 (C, methine), 25.8 [C(CH$_3$)$_2$]. ESI-MS: 534.8 (495 + ^{39}K). M.p: 240–242°C.

4c: ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.39–7.38 (m, 1H, ArH); 7.37–7.28 (m, 1H, ArH); 7.26–7.20 (m, 1H, ArH); 7.18–7.05 (m, 2H, ArH); 7.02–6.83 (m, 1H, ArH); 5.7 (s, 1H); 2.25–2.17 (m, 2H); 2.06–1.99 (m, 2H); 1.66–1.58 (m, 6H); 0.95–0.88 (m, 12H, 4CH$_3$).

^{13}C NMR (100 MHz, CDCl$_3$): δ 195.4, 163.4 (Ph–C–F), 149–125 (Ph and thiophene), 50.2 (CH$_2$–CO), 40.2 (CH$_2$–C), 32.8 (C, methine) 22.6 [C(CH$_3$)$_2$]. ESI-MS: 506 (483 + ^{23}Na). M.p: 245–247°C.

4d: ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.42–7.33 (m, 2H, ArH); 7.38–7.36 (m, 1H, ArH); 7.27–7.22 (m, 1H, ArH); 7.08–6.94 (m, 2H, ArH); 6.81–6.71 (m, 1H, ArH); 5.2 (s, 1H); 2.28–2.21 (m, 4H); 2.08–2.04 (d, J = 16, 2H); 1.90–1.86 (d, J = 16, 2H); 1.00 (s, 6H, 2CH$_3$); 0.88 (s, 6H, 2CH$_3$).

^{13}C NMR (100 MHz, CDCl$_3$): δ 195.6 (C=O), 153.4 (Ph–C–F), 135–115 (Ph), 49.9 (CH$_2$–CO), 41.7 (CH$_2$–C), 32.1 (C, methine), 27.4 [C(CH$_3$)$_2$]. ESI-MS: 512.1 (511.1 + ^{1}H). M.p: 250–252°C.
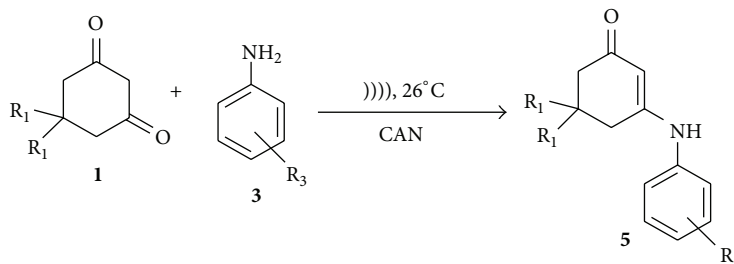
Scheme 3: Formation of β-enaminones.

Table 1: Comparison between CAN catalysed silent and sonic reactions.

Entry	Reactions	Silent condition[a]		Sonic condition[b]	
		Time	Yield[c]	Time	Yield[c]
1	Dimedone (1 mmol) and 2-chloro-4-fluoroaniline (1 mmol) to form **5**	45 min	75%	20 min	95%
2	Reaction between intermediates **5** and **6** to give product **4**	4 h	50%	1 h	90%
3	Mixing all the reactants (**1**, **2**, and **3**)	5 h	50%	1 h	90%

[a]In acetonitrile (3–5 mL) at 70°C. [b]In a sonic bath at 26°C (35 kHz). [c]Isolated yields.

4e: ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.56–7.41 (m, 2H, ArH); 7.41–7.39 (m, 1H, ArH); 7.31–7.20 (m, 5H, ArH); 7.17–7.06 (m, 1H, ArH); 5.20 (s, 1H); 2.19–2.13 (m, 4H); 1.62–1.57 (m, 4H); 0.93 (s, 6H, 2CH$_3$); 0.83 (s, 6H, 2CH$_3$).

^{13}C NMR (100 MHz, CDCl$_3$): δ 197.0 ($\underline{C}$=O), 161.2 (Ph–$\underline{C}$–F), 135–115 (Ph), 49.0 ($\underline{C}$H$_2$–CO), 40.2 ($\underline{C}$H$_2$–C(CH$_3$)$_2$), 32.5 (C, methine), 25.8 [$\underline{C}$(CH$_3$)$_2$]. ESI-MS: 500.1 (477.1 + [23]Na). M.p: 260–262°C.

4f: ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.4–6.7 (m, 9H, ArH); 5.2 (s, 1H); 2.3–2.26 (m, 4H); 2.21–2.17 (d, J = 16, 2H); 2.08–2.04 (d, J = 16, 2H); 1.6 (s, 3H); 1.07 (s, 6H, 2CH$_3$); 0.87 (s, 6H, 2CH$_3$).

^{13}C NMR (100 MHz, CDCl$_3$): δ = 197.0 ($\underline{C}$=O), 161.2 (Ph–$\underline{C}$–F), 150.6 (Ph–$\underline{C}$–OH), 135–115 (Ph), 50.0 ($\underline{C}$H$_2$–CO), 40.2 ($\underline{C}$H$_2$–C), 32.5 (C, methine), 25.8 [$\underline{C}$(CH$_3$)$_2$]. ESI-MS: 516.1 (493.1 + [23]Na). M.p: 253–255°C.

8a: ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.56–7.41 (m, 1H, ArH); 7.41–7.39 (m, 2H, ArH); 7.31–7.21 (m, 4H, ArH); 7.20–7.08 (m, 2H, ArH); 5.2 (s, 1H); 2.19–2.13 (m, 4H); 1.62–1.57 (m, 4H); 0.93 (s, 6H, 2CH$_3$); 0.83 (s, 6H, 2CH$_3$). ESI-MS: 426.2 (449.3 + [23]Na). M.p: 258–263°C.

8b: ^{1}H NMR (400 MHz, CDCl$_3$): δ 7.13–6.63 (m, 8H, ArH); 5.2 (s, 1H); 3.50–3.27 (m, 2H); 2.72–2.54 (m, 2H); 2.26–2.08 (m, 2H); 1.06 (s, 6H, 2CH$_3$). ESI-MS: 398.2 (399.2 + [1]H). M.p: 263–267°C.

3. Results and Discussion

To begin with, we planned to work with highly electron deficient 2-chloro-4-fluoroaniline (1 mmol), dimedone (2 mmol), and an electron deficient 4-fluorobenzaldehyde (1 mmol) in 3–5 mL acetonitrile as a solvent. We studied the reaction using various Lewis acid catalysts such as ZnCl$_2$, ZnBr$_2$, SnCl$_4$, AlCl$_3$, CuCl, and CAN under sonic condition (26°C, 35 kHz) and found that CAN (5 mole%) catalysed the reaction effectively and gave very high yield (90%, 1 h) of the product under sonic condition, and with other catalysts the yield was below 40% after 2 h.

To understand the effect of ultrasound on the present reaction, we carried out a comparative study on the CAN catalysed reaction under sonic and silent condition. Under silent condition, the reaction was carried out using dimedone (2 mmol), 2-chloro-4-fluoroaniline (1 mmol), and 4-fluorobenzaldehyde (1 mmol) in acetonitrile (3–5 mL) as a solvent at 70°C for 4 h, and we observed the formation of acridine-1,8-dione in 50% yield (Table 1, entry 3). This is because formation of β-enaminone (Scheme 2) under silent condition from electron-deficient aniline and an aldehyde is generally difficult; on sonication (26°C, 35 kHz) the yield was 90% after 1 h (entry 3) (Scheme 3).

In order to understand the role of ultrasound and the catalyst we decided to study the mechanism of formation of acridines in detail. From the literature, it is clear that formation of acridines involves the condensation of β-enaminone (**5**), with the Knoevenagel adduct **6** derived from the dimedone or cyclohexan-1,3-dione and an aromatic aldehyde as shown in Scheme 4, followed by the cyclization of intermediates **A** and **B** followed by the removal of a molecule of water to give the product as shown in Scheme 5. We feel that the catalyst may activate the intermediates **6**, **A**, and **B** to give product **4**. Although the formation of intermediates **5** and **6** is a must, isolation of these intermediates is not a necessary step under sonic condition, mixing all the starting materials and CAN, and subjecting the mixture to sonication in acetonitrile 26°C will give the products in very high yields as shown in Table 1 (entry 3).

After optimizing the reaction conditions, we applied the optimized condition to a range of other aromatic aldehydes and aromatic amines, ammonium acetate, and a heterocyclic

TABLE 2: A small library of acridines synthesized under sonic condition[a].

Entry	(1)	(2)	Amines (3)	Product	Yield[b] %	Time (min)
1	$R_1 = CH_3$	4-Methoxybenzaldehyde	4-Chloro, 2-fluoroaniline (3a)	4a[†]	90	40
2	$R_1 = CH_3$	2-Fluorobenzaldehyde	(3a)	4b[†]	95	45
3	$R_1 = CH_3$	Thiophene-2-aldehdye	(3a)	4c[†]	95	45
4	$R_1 = CH_3$	4-Chlorobenzaldehyde	(3a)	4d[†]	93	45
5	$R_1 = CH_3$	Benzaldehyde	(3a)	4e[†]	95	45
6	$R_1 = CH_3$	4-Hydroxybenzaldehyde	(3a)	4f[†]	90	40
7	$R_1 = CH_3$	Benzaldehyde	Aniline (3b)	4g[¥]	95	40
8	$R_1 = CH_3$	4-Chlorobenzaldehyde	(3b)	4h[¥]	93	40
9	$R_1 = H$	4-Chlorobenzaldehyde	(3b)	4j[¥]	90	40
10	$R_1 = H$	Benzaldehyde	NH_4Ac	7a[¥]	90	20
11	Dimedone	4-Chlorobenzaldehyde	NH_4Ac	7b[¥]	95	20
12	$R_1 = CH_3$	Benzaldehyde	2-Aminopyridine	8a[†]	90	50
13	Dimedone and cyclohexa-1,3-dione	Benzaldehyde	2-Aminopyridine	8b[†]	85	50

[a]Reaction condition as mentioned in general procedure. [b]Isolated yields. [†]Novel compounds. [¥]Known compounds were characterized by comparison of their physical data and on TLC with the authentic samples.
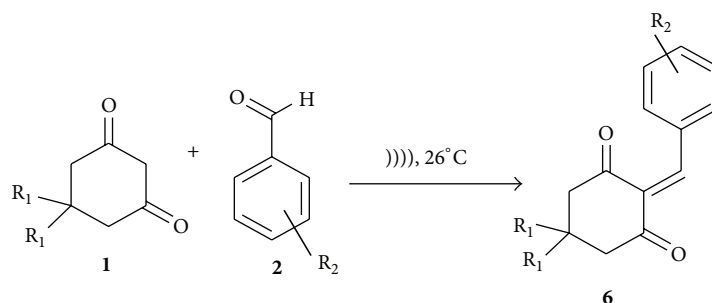

SCHEME 4: Formation of Knoevenagel adduct.
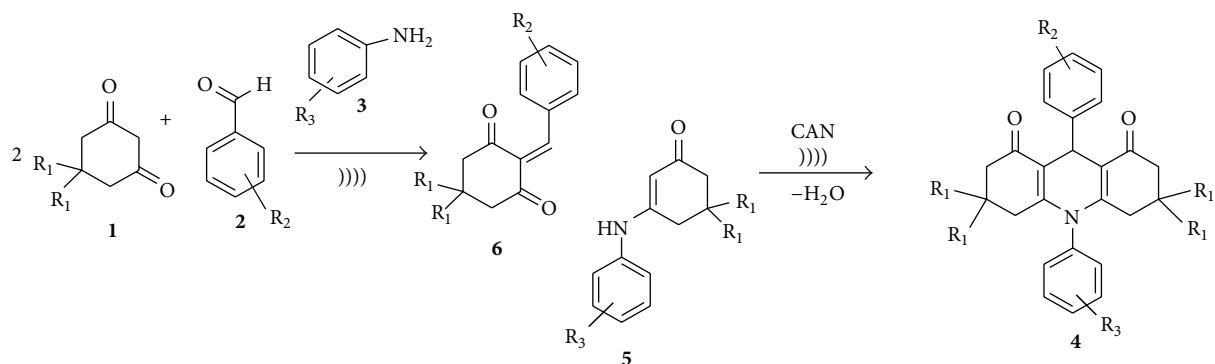
SCHEME 5: Formation of N-substituted acridines.

amine; the results of this study are presented in Table 2. The reaction using ammonium acetate was found to be faster than the reaction with aromatic amines and aniline under sonic condition. Electron releasing and electron withdrawing groups on aldehydes and amines did not show much effect on the rates and yield of the reaction under sonic condition. To study the efficacy of the developed method, we extended the work to heterocyclic aldehyde: thiophene-2-aldehdye (Table 2, entry 3) and heterocyclic amine: 2-aminopyridine (Table 2, entries 11 and 12), and found to give the respective products in very high yield (Scheme 6).

3.1. Effect of Sonication. The noteworthy qualities of the sonication are improved reaction rates and formation of pure products in high yields. Additionally, sonication allows benefits like easier manipulation, improved energy conservation, and low waste generation as compared to the traditional methods; in recent years, the sonochemical energy delivery has been used as an excellent alternative to thermal energy in promoting organic reactions. The origin of the energy lies in the "cavitation" phenomenon which involves formation, growth, and collapse of several millions of tiny vapour bubbles in the liquid medium [21, 22]. This effect is mainly
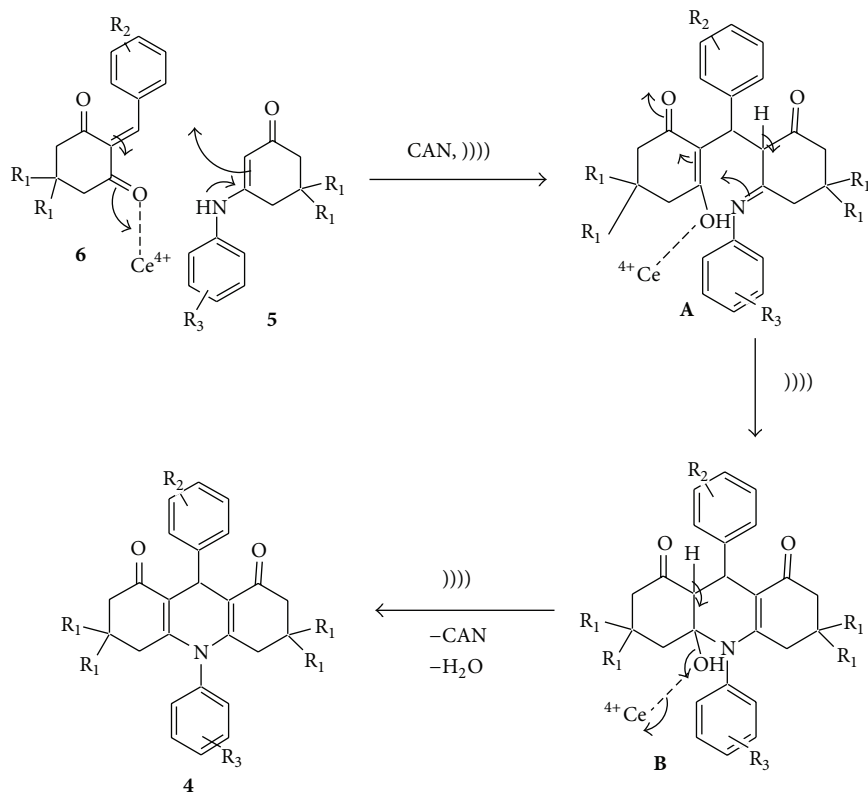
SCHEME 6: A plausible mechanism for the formation of acridines from **5** and **6**.

responsible for generating strong convection in the liquid media through several mechanisms such as microstreaming, microturbulence, shock waves, and microjets. The fast imploding bubbles generate extremely high temperatures of the order of 5000°C and very high pressures of about 1000 atm within the cavity. In such microreactors sound energy is transformed into the chemical energy [23]. The possible nuclei for the "cavitation" are also the gas pockets trapped in the walls and crevices of the solid reagents, reactants, and vessel walls, or they could be small bubbles already present in the reaction medium. Thus the collapsing bubbles generate localized "hot spots" and the reactant molecules enter into the cavities where dissolution of reactants can take place; molecular fragmentation can also take place and can produce highly reactive intermediates and species. Cavitation is responsible for the chemical effects including the acceleration of the rates of the reactions through the heat and the mass transformations in homogeneous solutions [24].

We have applied the effective combination of suitable solvents and ultrasound in the synthesis of heterocyclic and other important compounds through one-pot multicomponent reaction strategies under sonic condition earlier [18, 19]. Therein, we have explained the effectiveness of these combinations in improving the reaction parameters and conserving the energy. We have now applied this strategy in the synthesis of N-H- and N-substituted acridines by a one-pot four-component reaction under sonic condition as shown in Scheme 4.

4. Conclusions

To conclude, we have developed a general, practical, and high yielding procedure to construct different N-H- and N-substituted acridines from electron-deficient as well as electron-rich aromatic aldehydes and aromatic amines or ammonium acetate and dimedone or cyclohexyl-1,3-diones at 26°C under sonic condition (35 kHz). High yields, shorter reaction durations, and mild reaction conditions are the added advantages of our energy efficient method.

Conflict of Interests

The authors declare that there is no conflict of interests; that is, the authors of the paper do not have a direct financial relation that might lead to a conflict of interests for any of the authors.

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