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Green, convenient, $CuSO_4 \cdot 5H_2O$ assisted one-pot four-component synthesis of Aryl-hexahydro-acridine-1,8-diones in aqueous EtOH

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ABSTRACT

A simple, convenient and economical method for the synthesis of 9-aryl-tetrahydro-acridine-1,8-diones by a one-pot four-component condensation of aromatic/hetero-aromatic aldehydes, dimedone and ammonium acetate with catalytic $CuSO_4 \cdot 5H_2O$ in a 1:1 H_2O-EtOH mixture at 80 °C is reported. The use of the readily available catalyst pronounces the methodology to be a greener approach in obtaining very high yields of the desired products in shorter reaction duration.

Key words: 9-Aryl-hexahydro-acridine-1,8-diones, aldehydes, dimedone, ammonium acetate, $CuSO_4 \cdot 5H_2O$, H_2O-ethanol

1. Introduction

One-pot multicomponent reactions (MCRs) have provided a promising step in the synthesis of complex organic molecules and medicinally important compounds in recent years. Multicomponent reactions proceed through the reaction between three or more reactants in a single step and produce products incorporating substantial functionalities of all the reactants in one-step [1]. Multicomponent processes are now a much fact-finding approaches and are rapidly evolving. Working on one-pot multicomponent processes is a challenging task as one needs to understand the reactivity of the starting materials and also the reactivity of the in situ generated intermediates. MCRs not only provide an ease in selection of the synthetic pattern but also require an intellectual fashioning of the organic molecules. With much advances, MCRs have now developed as an integral part of a one-step, target-oriented and diversity-oriented syntheses of several highly complex organic molecules [2].

Acridine derivatives are found to be widely distributed in many natural products and exhibit a variety of biological activities such as: anti-malarial, anti-microbial, anti-inflammatory, anti–cancer and acetylcholinesterase inhibition activities. The acridine motif is also found in chemosensors, dyes, fluorescent probes, as ligands in metal-promoted catalysis or very recently in hole transport or chiroptical materials and in the two photon absorption devices [3]. There are a few reports in the literature on the one-pot three-component Hantzsch-type condensation of aromatic aldehydes, anilines and dimedone via the traditional heating in organic solvents; under microwave irradiation and in ionic liquids leading to acridinediones [4]. Recently, we reported the synthesis of acridinediones using SiO_2–I as a new heterogeneous catalyst [5]. A report on the application of CuO nanoparticles in the synthesis of acridinediones is also available [6]. While the reported methods have their own advantages and disadvantages, the last two methods involve tedious preparation of the catalysts.

$CuSO_4$ has been used earlier in various organic transformations [7]. We thought that, use of this readily available Lewis acid catalyst can be adopted in a one-pot four-component reaction which could pronounce a methodical make over and would be a useful attempt to the synthesis of 9-aryl-

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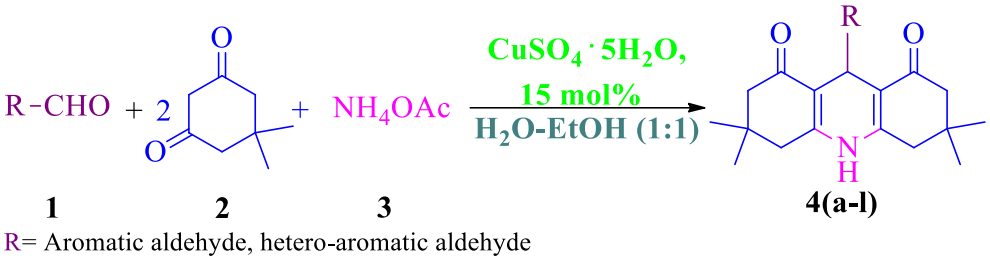
hexahydroacridine-1,8-diones in very high yields. The reaction between various aromatic/hetero-aromatic aldehydes, dimedone and ammonium acetate took place readily in the presence of a catalytic amount of CuSO4 in 1:1 H_2O-EtOH at 80 °C as shown in the **Scheme 1**.



Scheme 1: Synthesis of 9-aryl-hexahydroacridine-1,8-diones

2. RESULTS AND DISCUSSION

A series of acridinediones were synthesized by using different aromatic/hetero-aromatic aldehydes, dimedone and ammonium acetate in the presence of $CuSO_4{\cdot}5H_2O$ as a catalyst. In order to understand the effect of solvent on the rate of the present reaction, ether, DCM, *n*-hexane, toluene, THF, water, ethanol and a binary mixture of THF-EtOH (1:1) and H_2O-EtOH (1:1) were selected and the reaction was carried out at 80 °C. The reaction of dimedone (2 mmol), 3,4,5-trimethoxy benzaldehyde (1 mmol) and ammonium acetate (1 mmol) afforded the corresponding product with varying yields (Table 1). In water alone or ethanol alone, the product was obtained in moderate yields (**Table 1**, entries 6 and 7). 1:1::H_2O:EtOH mixture (4 mL) afforded the corresponding product in relatively good yield in shorter duration (entry 9). As observed on TLC, under the solvent-free condition, the reaction did not proceed (entry 10).

Table 1: Effect of solvent on the synthesis of 3,4,6,7-tetrahydro-9-(3′,4′,5′-trimethoxyphenyl)-3,3,6,6-tetramethylacridine-1,8-dione (**4a**)

Entry	Solvent	Temperature (°C)	Time (h)	Yield[c] (%)
1	Et_2O[a]	RT	8	20
2	DCM[a]	30	5	30
3	n-Hexane [a]	60	5	ND
4	Toluene [a]	90	5	40
5	THF[a]	60	5	45
6	H_2O[a]	60	5	50
27	EtOH[a]	65	5	50
8	THF-EtOH[b]	70	3	47
9	**H_2O-EtOH**[b]	**80**	**3**	**55**
10	Solvent-free	70	3	ND[d]

Reaction conditions: [a]2 mL; [b](1:1, 4 mL); [c]Isolated yield; [d]ND-Not detected.

After finding a suitable solvent, it was important to study the role of the catalyst on the rate of the reaction and the yield of the product. In order to find a suitable catalyst, about 15 different Lewis acidic and basic catalysts were selected for the synthesis of 4a; among all the catalysts used,

$CuSO_4{\cdot}5H_2O$ in H_2O-EtOH solvent system superseded the observed catalytic role of the other catalysts including the activity of other copper salts in terms of rate of the reaction and yield of the product as shown (**Table 2**, entry 15).

Table 2: Influence of various catalysts and copper salts in the synthesis of **4a**

Entry	Catalyst[a]	Temperature (°C)	Time (h)	Yield[d] (%)
1	K_2CO_3	70 – 80	3	35
2	$Ba(OH)_2$	70 – 80	3	55
3	$Na_2S_2O_6$[b]	70 – 80	3	40
4	NiO[b]	70 – 80	3	47
5	CuO[b]	70 – 80	3	65
6	ZnO[b]	70 – 80	3	45
7	$Cu(OAc)_2$[b]	70 – 80	2.5	74
8	CuI	70 – 80	2.5	87
9	$Cu(NO_3)_2$	70 – 80	2.5	77
10	Cu_2O/EtOH/H_2SO_4[b,c]	70 – 80	2.5	80
11	CuO/EtOH/H_2SO_4[b,c]	70 – 80	2.5	89
12	$LaCl_3{\cdot}5H_2O$	70 – 80	3	87
13	$BiNO_3{\cdot}5H_2O$	70 – 80	3	70
14	$SnCl_2{\cdot}H_2O$	70 – 80	3	85
15	**$CuSO_4{\cdot}5H_2O$**	**70 – 80**	**1.5**	**94**
16	No catalyst	70 – 80	8	ND[e]

Reaction conditions: [a]15 mol%; [b]EtOH (2mL); [c]2M H_2SO_4 (1 mL); [d] Isolated yield; [e]Not detected.

Further studies were carried out to optimize the amount of catalyst required for the reaction. Different quantities of $CuSO_4{\cdot}5H_2O$ (30, 25, 20, 15, 10, 5, 2 and 1 mol%) was used for establishing the catalytic load of $CuSO_4{\cdot}5H_2O$ in the above reaction. The result of this study are presented in the Table 3. The data presented in the **Table 3** portrayed that, 15 mol% of $CuSO_4{\cdot}5H_2O$ in H_2O-EtOH (1:1, 4 mL) afforded the product in 94% isolated yield.

Table 3: Optimization of the amount of the catalyst for the synthesis of **4a** in H_2O-EtOH

Entry	$CuSO_4{\cdot}5H_2O$ (mol%)	H_2O/EtOH (mL)	Yield (%)[a]
1	30	2/2	87
2	25	2/2	88
3	20	2/2	91
4	**15**	**2/2**	**94**
5	10	2/2	86
6	5	2/2	66
7	2	2/2	45
8	1	2/2	45

[a]isolated yield.

Considering the above observations, in order to extend the method for the synthesis of different substituted acridinediones, we next carried out a series of reactions involving various aromatic and hetero-aromatic aldehydes, dimedone and ammonium acetate in the presence of $CuSO_4{\cdot}5H_2O$ (15 mol%) in H_2O-EtOH at 80 °C as shown (**Table 4**).

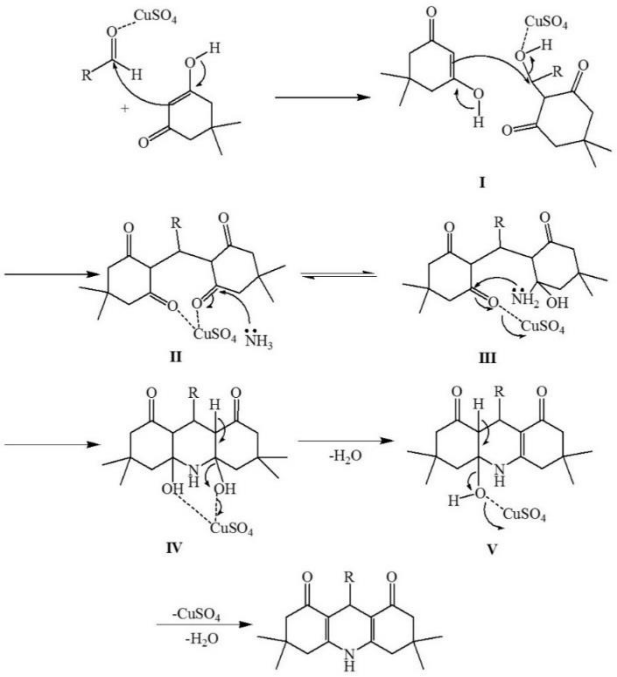
Table 4: Synthesis of acridinediones (**4a–l**) using $CuSO_4{\cdot}5H_2O$ in H_2O-EtOH at 80 °C

Entry	Aromatic/ hetero-aromatic aldehydes	Product[a]	Time (h)	Yield[b] (%)	Mp (°C)	
					Found	Reported
1	3,4,5-$(CH_3O)_3C_6H_2CHO$	**4a**	1.5	94	308–310	-
2	4-HOC_6H_4CHO	**4b**	1.5	90	361–363	> 300 [8]
3	2-HOC_6H_4CHO	**4c**	1.5	89	310–312	310–312 [9]
4	2-ClC_6H_4CHO	**4d**	1.5	84	221	220–222 [10]
5	4-$CF_3C_6H_4CHO$	**4e**	1.5	86	241–243	241–243 [11]
6	Thiophene-2-CHO	**4f**	1.5	89	306	306–308 [12]
7	4-Cl,3-FC_6H_3CHO	**4g**	1.5	84	255–257†	-
8	2,5-Dimethyl-1-phenyl-1*H*-pyrrole-3-CHO	**4h**	1.5	85	302–304†	-
9	4-FC_6H_4CHO	**4i**	1.5	84	273	275–276 [13]
10	5-(Methylthio)thiophene-2-CHO	**4j**	1.5	89	298–300†	-
11	6-Hydroxynaphthalene-2-CHO	**4k**	1.5	90	238–240†	-
12	4-$(CF_3O)C_6H_4CHO$	**4l**	1.5	90	139–142†	-

[a]All the synthesized products were characterized from their spectroscopic & analytical data; [b]Isolated yield; †Novel compound.

2.1. Mechanism

A plausible mechanism for the formation of acridinediones involves the $CuSO_4{\cdot}5H_2O$ mediated activation of the carbonyl group of the aldehyde followed by the attack of an enol form of the dimedone to give the intermediate **I**. The intermediate I may react with another molecule of dimedone to give **II**; N insertion is then facilitated by the generation of ammonia from ammonium acetate to give **III**. The intermediate **III** may then cyclize to give the acridine derivative **IV** which after the elimination of a molecule of water may give the intermediate **V**. The intermediate **V**, may lose another molecule of water to give acridinediones as shown in the **Scheme 2**. All the products exhibited a singlet in ^{1}HNMR spectra between δ = 5.00–5.79 ppm for -CH-9 and peaks between δ = 8.80–12.72 ppm for -NH group respectively.



Scheme 2: A plausible mechanism for the synthesis of acridine-1,8-diones.

3. Experimental

3.1. Materials and methods

All chemicals were commercially available and were used without further purification. All yields refer to the yield of the isolated products after purification. All the products were characterized by the IR, ^{1}HNMR, ^{13}CNMR, Mass spectral and elemental analyses. Melting points were measured on a Raaga, Indian make melting point apparatus. NMR spectra were obtained on a 400 MHz and 100 MHz Bruker AMX instruments in $CDCl_3$ using TMS as an internal standard. ESI-MS analysis was carried out using ESI-Q TOF instrument. CHN analysis was performed using Elementar vario MICRO cube analyzer instrument.

3.2 General procedure for the synthesis of substituted 3,3,6,6-tetramethyl-9-aryl-3,4,6,7,9,10-hexahydroacridine-1,8-diones

In a 50 mL of round-bottomed flask, a mixture of aryl/heteroaryl aldehyde (1 mmol), dimedone (2 mmol), ammonium acetate (1 mmol), $CuSO_4{\cdot}5H_2O$ (15 mol%), H_2O:EtOH mixture (1:1, 4 mL) were refluxed at 80 °C with continuous stirring for 1.5 h. The progress of reaction was monitored by TLC. After the completion of the reaction, the reaction mixture was cooled to room temperature. The solid product was repeatedly washed with water, the left over solid was dissolved in ethyl acetate (10 mL). The concentrated organic layer was dried over anhydrous Na_2SO_4, filtered and evaporated under vacuum. The obtained solid was collected and purified by recrystallization using absolute ethanol.

3.3 Spectral data of the synthesized aryl-hexahydroacridine-1,8-diones:

3,4,6,7-tetrahydro-9-(3',4',5'-trimethoxyphenyl)-3,3,6,6-tetramethylacridine-1,8 (2H,5H,9H,10H)-dione (**4a**):
^{1}HNMR (400 MHz, $CDCl_3$): δ 1.12 (6H, s, $2CH_3$), 1.24 (6H, s, $2CH_3$), 2.42–2.35 (8H, m, $4CH_2$), 3.75 (6H, s, $2OCH_3$), 3.81 (3H, s, OCH_3), 5.49 (1H, s, CH), 6.34 (2H, s, Ar-H), 12.00 (1H, s, NH) ppm;
^{13}CNMR (100 MHz, $CDCl_3$): δ 27.2, 30.1, 39.7, 42.0, 51.1, 56.1, 56.9, 106.1, 111.0, 131.0, 132.0, 149.3, 150.0, 198.0 ppm;
Mass (m/z): $[M]^+$: 439.2;
Anal. Calcd for: $C_{26}H_{33}NO_5$: C, 71.05; H, 7.57; N, 3.19 %. Found: C, 70.91; H, 7.38; N, 2.91 %.

3,4,6,7-tetrahydro-9-(4'-hydroxyphenyl)-3,3,6,6-tetramethylacridine-1,8 (2H,5H,9H,10H)-dione (**4b**):
^{1}HNMR (400 MHz, $CDCl_3$): δ 1.12 (6H, s, $2CH_3$), 1.24 (6H, s, $2CH_3$), 2. 24 – 2.15 (8H, m, $4CH_2$), 5.09 (1H, s, OH), 5.41 (1H, s, CH), 6.53 (2H, d, J = 1.6 Hz, Ar-H), 7.17 (2H, d, J = 8.4 Hz, Ar-H), 12.7 (1H, s, NH) ppm;
^{13}CNMR (100 MHz, $CDCl_3$): δ 27.9, 31.9, 43.1, 47.4, 51.3, 112.8, 115.1, 129.4, 130.1, 149.4, 155.5, 198.4 ppm;
Mass (m/z): $[M+1]^+$: 366.2;
Anal. Calcd for: $C_{23}H_{27}NO_3$: C, 18 75.59; H, 7.45; N, 3.83 %. Found: C, 74.91; H, 7.20; N, 2.34 %.

3,4,6,7-tetrahydro-9-(2'-hydroxyphenyl)-3,3,6,6-tetramethylacridine-1,8 (2H,5H,9H,10H)-dione (**4c**):
^{1}HNMR (400 MHz, $CDCl_3$): δ 1.02 (9H, s, $3CH_3$), 1.12 (3H, s, CH_3), 2.62 –1.90 (8H, m, $4CH_2$), 4.66 (1H, s, OH), 5.30 (1H, s, CH), 7.15–6.99 (4H, m, Ar-H), 10.4 (1H, s, NH) ppm;
^{13}CNMR (100 MHz, $CDCl_3$): δ 26.7, 30.6, 33.2, 42.2, 51.2, 110.0, 115.7, 122.1, 122.9, 125.0, 131.1, 149.4, 156.4, 198.9 ppm;
Mass (m/z): $[M+1]^+$. 366.2;
Anal. Calcd for: $C_{23}H_{27}NO_3$: C, 75.59; H, 7.45; N, 3.83%. Found: C, 75.01; H, 7.28; N, 2.80 %.

9-(4′-chlorophenyl)-3,4,6,7-tetrahydro-3,3,6,6-tetramethylacridine-1,8 (2H,5H,9H,10H)-dione (**4d**):
^{1}HNMR (400 MHz, $CDCl_3$): δ 0.98 (6H, s, $2CH_3$), 1.06 (6H, S, $2CH_3$), 2.40–2.17 (8H, m, $4CH_2$), 5.62 (1H, s, CH), 7.38–7.18 (4H, m, Ar-H), 11.9 (1H, s, NH) ppm;
^{13}CNMR (100 MHz, $CDCl_3$): δ 26.7, 30.0, 32.3, 42.2, 51.3, 111.9, 115.7, 126.9, 127.1, 128.4, 131.4, 134.3, 143.1, 149.4, 198.9 ppm;
Mass (m/z): $[M+1]^+$: 383.1;
Anal. Calcd for: $C_{23}H_{26}ClNO_2$: C, 71.96; H, 6.83; N, 3.65 %. Found: C, 70.89; H, 6.30; N, 3.20 %.

9-(4′-trifluoromethyl phenyl)-3,4,6,7-tetrahydro-3,3,6,6-tetramethylacridine-1,8 (2H,5H,9H,10H)-dione† (**4e**):
^{1}HNMR (400 MHz, $CDCl_3$): δ 0.97 (6H, s, $2CH_3$), 1.04 (6H, s, $2CH_3$), 2.53–2.15 (8H, m, $4CH_2$), 5.12 (1H, s, CH), 7.76–7.74 (2H, d, *J* = 8 Hz, Ar-H), 8.23–8.21 (2H, d, *J* = 8.4 Hz, Ar-H), 10.0 (1H, s, NH) ppm;
^{13}CNMR (100 MHz, $CDCl_3$): δ 27.2, 30.5, 34.5, 42.2, 51.2, 111.1, 123.1, 123.8, 127.9, 128.8, 145.0, 149.4, 198.8 ppm;
Mass (m/z): $[M+1]^+$: 417.1;
Anal. Calcd for: $C_{24}H_{26}F_3NO_2$: C, 69.05; H, 6.28; N, 3.36 %. Found: C, 68.09; H, 6.21; N, 3.12 %.

3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-9-(thiophen-2′-yl)acridine-1,8 (2H,5H,9H,10H)-dione (**4f**):
^{1}HNMR (400 MHz, $CDCl_3$): δ 1.04 (6H, s, $2CH_3$), 1.09 (6H, s, $2CH_3$), 2.40–2.00 (8H, m, $4CH_2$), 5.47 (1H, s, CH), 6.98–6.80 (3H, m, Ar-H), 10.1 (1H, s, NH) ppm;
^{13}CNMR (100 MHz, $CDCl_3$): δ 28.1, 30.9, 31.1, 43.3, 52.0, 111.9, 123.2, 128.0, 128.3, 139.0, 149.6, 198.8 ppm;
Mass (m/z): $[M]^+$: 355.1;
Anal. Calcd for: $C_{21}H_{25}NO_2S$: C, 70.95; H, 7.09; N, 3.94 %. Found: C, 70.28; H, 7.01; 18 N, 2.99 %.

9-(4′-chloro-3′-fluorophenyl)-3,4,6,7-tetrahydro-3,3,6,6-tetramethylacridine-1,8 (2H,5H,9H,10H)-dione (**4g**)†:
^{1}HNMR (400 MHz, $CDCl_3$): δ 1.36 (6H, s, $2CH_3$), 1.60 (6H, s, $2CH_3$), 2.28–2.19 (8H, m, $4CH_2$), 4 5.28 (1H, s, CH), 7.19 (1H, s, Ar-H), 7.36–7.33 (1H, d, *J* = 2 Hz, Ar-H), 7.50 (1H, d, *J* = 2 Hz, Ar-5 H), 10.1 (1H, s, NH) ppm;
^{13}CNMR (100 MHz, $CDCl_3$): δ 27.6, 29.5, 31.4, 23 41.9, 50.3, 111.4, 116.20, 116.23, 124.6, 128.0, 140.1, 151.4, 169.4, 196.9 ppm;
Mass (m/z): $[M]^+$: 1 401.1;
Anal. Calcd for: $C_{23}H_{25}ClFNO_2$: C, 68.73; H, 6.27; N, 3.49 %. Found: C, 67.28; H, 6.12; N, 2 3.09 %.

3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-9-(2′,5′-dimethylphenyl-1H-pyrrol-3′′-yl)acridine-1,8 (2H, 5H,9H,10H)-dione (**4h**)†:
^{1}HNMR (400 MHz, $CDCl_3$): δ 1.080 (6H, s, $2CH_3$), 1.086 (6H, s, $2CH_3$), 2.34–2.12 (8H, m, $4CH_2$), 5.30 (1H, s, CH), 5.66 (1H, s, pyrrole-H), 7.31 (5H, s, Ar-H), 8.81 (1H, s, NH) ppm;
^{13}CNMR (100 MHz, $CDCl_3$): δ 5.21, 12.02, 27.2, 30.5, 34.5, 42.2, 51.2, 110.9, 112.2, 120.9, 122.9, 125.0, 125.9, 131.4, 132.0, 140.4, 150.8, 198.1 ppm;
Mass (m/z): $[M]^{+\cdot}$ 442.2;
Anal. Calcd for: $C_{29}H_{34}N_2O_2$: C, 78.70; H, 7.74; N, 6.33 %. Found: C, 77.28; H, 7.21; N, 6.09 %.

9-(4′-fluorophenyl)-3,4,6,7-tetrahydro-3,3,6,6-tetramethylacridine-1,8 (2H,5H,9H,10H)-dione (**4i**):
^{1}HNMR (400 MHz, $CDCl_3$): δ 1.07 (6H, s, $2CH_3$), 1.22 (6H, s, $2CH_3$), 2.08–1.67 (8H, m, $4CH_2$), 5.06 (1H, s, CH), 7.16–7.14 (2H, d, *J* = 8.8 Hz, Ar-H), 7.25–7.23 (2H, d, *J* = 11.2 Hz, Ar-H), 11.8 (1H, s, NH) ppm;
^{13}CNMR (100 MHz, $CDCl_3$): δ 24.2, 31.0, 40.2, 43.5, 52.2, 111.9, 115.7, 129.4, 132.0, 149.4, 157.2, 198.3 ppm;
Mass (m/z): $[M]^{+\cdot}$ 367.1;
Anal. Calcd for: $C_{23}H_{26}FNO_2$: C, 75.18; H, 7.13; N, 3.81 %. Found: C, 74.04; H, 7.20; N, 3.79 %.

3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-9-[5′-(methylmercapto)thiophen-2′-yl]acridine-1,8 (2H,5H,9H,10H)-dione (**4j**)†:
1H NMR (400 MHz, $CDCl_3$): δ 1.05 (6H, s, $2CH_3$), 1.07 (6H, s, $2CH_3$), 2.26– 2.11 (4H, m, $2CH_2$), 2.22 (3H, s, CH_3), 2.62–2.29 (4H, m, $2CH_2$), 5.39 (1H, s, CH), 6.76–6.75 (1H, d, *J* = 3.6Hz), 6.81–6.80 (1H, d, *J* = 3.6 Hz), 10.8 (1H, s, NH) ppm;
13C NMR (100 MHz, $CDCl_3$): δ 15.8, 27.1, 30.1, 31.1, 43.0, 51.3, 111.9, 127.9, 131.0, 133.1, 142.6, 149.4, 198.9 ppm;
Mass (m/z): $[M]^+$: 401.1;
Anal. Calcd for: $C_{22}H_{27}NO_2S_2$: C, 65.80; H, 6.78; N, 3.49 %. Found: C, 64.04; H, 6.30; N, 3.20 %.

3,4,6,7-tetrahydro-9-(2-hydroxynaphthalen-6-yl)-3,3,6,6-tetramethylacridine-1,8(2H,5H,9H,10H)-dione (**4k**) †:
^{1}HNMR (400 MHz, $CDCl_3$): δ 1.10 (6H, s, $2CH_3$), 1.12 (6H, s, $2CH_3$), 2.50–2.22 (8H, m, $4CH_2$), 5.64 (1H, s, CH), 7.62–6.90 (6H, m, Ar-H), 10.2 (1H, s, OH), 11.9 (1H, s, NH) ppm;
^{13}CNMR (100 MHz, $CDCl_3$): δ 27.2, 30.1, 36.0, 39.7, 51.3, 107.0, 115.5, 116.0, 123.1, 123.8, 127.9, 128.8, 131.0, 133.1, 137.6, 149.5, 155.5, 198.8 ppm;
Mass (m/z): $[M]^+$: 415.2;
Anal. Calcd for: $C_{27}H_{29}NO_3$: C, 78.04; H, 7.03; N, 3.37 %. Found: C, 73.64; H, 6.90; N, 3.20 %.

3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-9-(4′-trifluoromethoxyphenyl)acridine-1,8 (2H,5H,9H,10H) -dione (**4l**)†:
^{1}HNMR (400 MHz, $CDCl_3$): δ 1.12 (6H, s, 2CH3), 1.25 (6H, s, 2CH3), 2.48–2.29 (8H, m, $4CH_2$), 5.54 (1H, s, CH), 7.14 (2H, d, *J* = 1.6 Hz, Ar-H), 7.17–7.16 (2H, d, *J* = 2.4 Hz, Ar-H), 10.2 (1H, s, NH) ppm;
^{13}CNMR (100 MHz, $CDCl_3$): δ 27.2, 31.7, 40.2, 43.1, 51.9, 111.9, 114.2, 121.8, 130.0, 134.5, 149.5, 159.1, 198.8 ppm;
Mass (m/z): $[M]^+$: 433.1;
Anal. Calcd for: $C_{24}H_{26}F_3NO_3$: C, 66.50; H, 6.05; N, 3.23 %. Found: C, 64.44; H, 5.46; N, 3.20 %.

4. CONCLUSIONS

A convenient, simple, efficient, eco-friendly, economical and greener approach has been developed for the synthesis of a few known and novel acridinediones using readily available $CuSO_4{\cdot}5H_2O$ as a catalyst at 80 °C. The method has been proved to be an improvisation which provides high yields of the products in short reaction duration. The present method of preparation of 9-aryl-tetrahydro-acridine-1,8-diones is an asset to the chemists and biologists as it involves application of an inexpensive, readily available and extremely simple and cheap catalyst in the synthesis of 9-aryl-tetrahydro-acridine-1,8-diones.

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