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Research Article

Poly(4-Vinylpyridinium)Hydrogen Sulfate Catalyzed an Efficient and Ecofriendly Protocol for the One-Pot Multicomponent Synthesis of 1,8-Acridinediones in Aqueous Medium

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Highly efficient, ecofriendly, and improved protocol for the synthesis of 1,8-acridinediones has been developed *via* one-pot multicomponent condensation of 1,3-cyclohexanedione/dimedone, aromatic aldehydes, and ammonium acetate utilizing poly(4-vinylpyridinium)hydrogen sulfate as catalyst in aqueous medium. Excellent yields in shorter reaction time, simple work-up procedure, easy recovery, and reusability of the catalyst are attractive features of this green protocol.

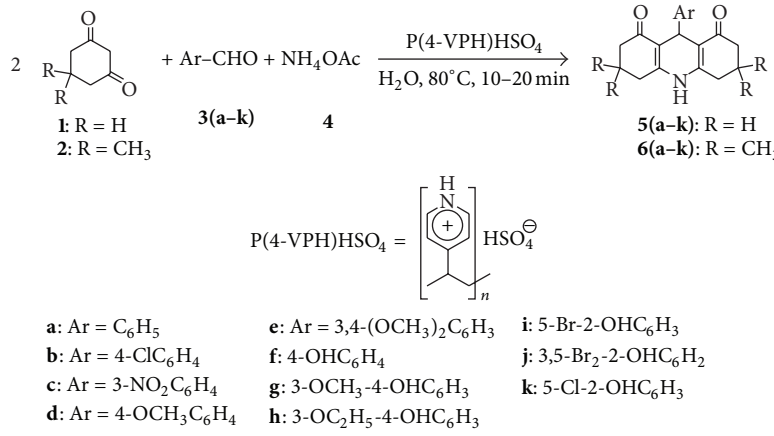
1. Introduction

Multicomponent reactions (MCRs) have emerged as an efficient and powerful tool in modern organic chemistry towards the generation of highly diverse and complex product from readily available substrates in a single operation without isolation of intermediates in minimal time with maximum selectivity, high atom economy, and high purity with excellent yields [1]. In addition, reactions under aqueous medium have received considerable attention in organic synthesis, because of both economic and environmental safety reasons [2]. Moreover, heterogeneous catalysts have advantages over homogeneous catalysts in terms of efficiency, operational simplicity, recovery from the reaction mixture, and reusability [3].

1,8-Acridinediones having 1,4-dihydropyridine (1,4-DHP) parent nucleus, which are versatile intermediates in the synthesis of numerous pharmaceuticals including those for the treatment of cardiovascular diseases and hypertension as effective calcium channel blockers [4, 5], Alzheimer's disease, and as chemosensitizer in tumor therapy [6, 7], are also used as laser dyes [8, 9] and photo initiators for radical polymerization reactions [10–12].

In view of the above applications, many methods have been developed including microwave irradiation [13, 14], using various catalytic systems such as *p*-toluenesulfonic acid [15], ceric ammonium nitrate [16], $InCl_3$ [17], $CeCl_3{\cdot}7H_2O$ [18], L-proline, $Zn(OAc)_2{\cdot}7H_2O$ [19], $In(OTf)_3$ [20], MCM-41-SO_3H [21], Amberlite IR-120H [22], methanesulfonic acid [23], silica bonded s-sulfonic acid [24], silica-supported preyssler nanoparticles [25], *p*-dodecylbenzenesulfonic acid [26], carbon based solid acid [27], and ionic liquids [28, 29]. However, most of these methods suffer from drawbacks such as low yield of the product, longer reaction time, multistep process, formation of side products, critical isolation procedure, use of hazardous organic solvents and expensive catalysts, difficult in recovery and reusability of the catalyst. Thus, there is a need for the development of new and efficient methods for the preparation of such compounds in high yields and under mild reaction conditions.

In continuation of our studies towards the development of novel methodologies for the synthesis of biologically



Scheme 1: $P(4\text{-VPH})HSO_4$ catalyzed synthesis of 1,8-acridinediones.

active heterocyclic compounds [30, 31], herein we report an ecofriendly method for the synthesis of 1,8-acridinediones using poly(4-vinylpyridinium)hydrogen sulfate [$P(4\text{-VPH})HSO_4$] as an efficient, heterogeneous, and recyclable catalyst in aqueous medium.

2. Results and Discussions

The schematic representation for the synthesis of title compounds (**5a–k** and **6a–k**) *via* one-pot multicomponent condensation of 1,3-cyclohexanedione (**1**)/dimedone (**2**) aromatic aldehydes (**3a–k**), and ammonium acetate (**4**) utilizing $P(4\text{-VPH})HSO_4$ as catalyst in aqueous medium was shown in Scheme 1. The catalyst $P(4\text{-VPH})HSO_4$ was prepared according to the literature procedure [32].

In order to find out the optimal conditions, a model reaction was carried out by the condensation of dimedone (**2**) with *p*-chlorobenzaldehyde (**3b**) and ammonium acetate (**4**) in aqueous medium at different temperatures by varying the amount of catalyst, and the results were summarized in Table 1. From Table 1, we found that only 0.02 g of $P(4\text{-VPH})HSO_4$ at 80°C is sufficient for completion of the reaction with maximum yield (Table 1, Entry 7). We also observed that as the amount of catalyst increases >0.02 g and the temperature >80°C, the yield of the product has decreased due to the unidentified impurities.

At these optimal conditions (0.02 g of $P(4\text{-VPH})HSO_4$, 80°C, aqueous medium), we have synthesized various 1,8-acridinediones (**5a–k** and **6a–k**) using different aromatic aldehydes with excellent yields in shorter reaction times (Table 2). All the synthesized compounds were characterized by their spectral studies and compared with the literature values where both were in good agreements. After completion of the reaction, the catalyst was recovered, washed with dichloromethane, dried, and reused for subsequent reactions for additional five times, and a slight decrease in its activity in terms of product yield was observed (Table 2, Entry 13).

The catalytic efficiency of $P(4\text{-VPH})HSO_4$ was compared with some other reported catalysts for the synthesis of 3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-9-phenylacridine-1,8-(2*H*,5*H*,9*H*,10*H*)-dione (**6b**). The results proved that the $P(4\text{-VPH})HSO_4$ is an efficient catalyst in terms of product yield and reaction times (Table 3).

Table 1: Optimizing the reaction conditions[a].

Entry	$P(4\text{-VPH})HSO_4$ (g)	Temperature (°C)	Time (min)	Yield[b] (%)
1	—	RT	120	—
2	—	80	120	28
3	—	Reflux	120	36
4	0.01	RT	60	Trace
5	0.01	80	30	74
6	0.01	Reflux	30	74
7	0.02	80	10	95
8	0.02	Reflux	10	95
9	0.03	80	10	94
10	0.03	Reflux	10	92

[a] Dimedone (2 mmol), *p*-chlorobenzaldehyde (1 mmol), ammonium acetate (3 mmol), and $P(4\text{-VPH})HSO_4$ in aqueous medium.
[b] Isolated yields.

A plausible mechanism for the formation of 1,8-acridinediones catalysed by $P(4\text{-VPH})HSO_4$ is shown in Scheme 2. In the presence of catalyst, the electrophilicity of aldehydic carbonyl carbon increases and readily reacts with the enolic form of 1,3-dicarbonyl compounds which resulted the formation of chalcone derivative [**C**]. Compound [**C**] reacts with aminated 1,3-dicarbonyl compound [**B**] which is formed *in situ* by the reaction of 1,3-dicarbonyl compound with ammonium acetate that furnished the corresponding 1,8-acridinediones through the cyclisation followed by the dehydration.

Table 2: P(4-VPH)HSO_4 catalyzed one-pot synthesis of 1,8-acridinediones (**5a–k** and **6a–k**).

Entry[a]	Aldehyde	Product	Time (min)	Yield[b] (%)	Melting point (°C)	
					Found	Lit. [Ref.]
1	Benzaldehyde	**5a**	15	92	278–280	279–281 [16]
2	4-Chlorobenzaldehyde	**5b**	10	94	297–299	298-299 [16]
3	3-Nitrobenzaldehyde	**5c**	15	89	278–281	282–284 [16]
4	4-Methoxybenzaldehyde	**5d**	10	91	306–308	304–306 [16]
5	3,4-Dimethoxybenzaldehyde	**5e**	10	93	355-356	—
6	4-Hydroxybenzaldehyde	**5f**	15	90	305–307	307–309 [16]
7	3-Methoxy-4-hydroxybenzaldehyde	**5g**	15	90	281–283	—
8	3-Ethoxy-4-hydroxybenzaldehyde	**5h**	20	88	299–302	—
9	5-Bromo-2-hydroxybenzaldehyde	**5i**	20	91	272–274	—
10	3,5-Dibromo-2-hydroxybenzaldehyde	**5j**	20	90	250-251	—
11	5-Chloro-2-hydroxybenzaldehyde	**5k**	20	92	295–297	—
12	Benzaldehyde	**6a**	10	93	258–260	258–260 [18]
13	4-Chlorobenzaldehyde	**6b**	10	95 (93, 90, 89, 86, 84)[c]	299–301	298–300 [18]
14	3-Nitrobenzaldehyde	**6c**	15	88	290–293	294–296 [18]
15	4-Methoxybenzaldehyde	**6d**	10	90	273-274	272-273 [23]
16	3,4-Dimethoxybenzaldehyde	**6e**	10	94	259–261	258–260 [13]
17	4-Hydroxybenzaldehyde	**6f**	15	92	361–363	>300 [23]
18	3-Methoxy-4-hydroxybenzaldehyde	**6g**	15	92	294–296	294-295 [18]
19	3-Ethoxy-4-hydroxybenzaldehyde	**6h**	15	89	280–282	—
20	5-Bromo-2-hydroxybenzaldehyde	**6i**	20	90	254-255	—
21	3,5-Dibromo-2-hydroxybenzaldehyde	**6j**	20	90	294-295	—
22	5-Chloro-2-hydroxybenzaldehyde	**6k**	20	92	234–236	—

[a]Reaction conditions: 1,3-cyclohexanedione/dimedone (2 mmol), arylaldehyde (1 mmol), ammonium acetate (3 mmol), and P(4-VPH)HSO_4 (0.02 g), heat at 80°C for 10–20 min in 5 mL of H_2O.
[b]Isolated yields.
[c]Yields refer to the reusability of the catalyst over additional five times.

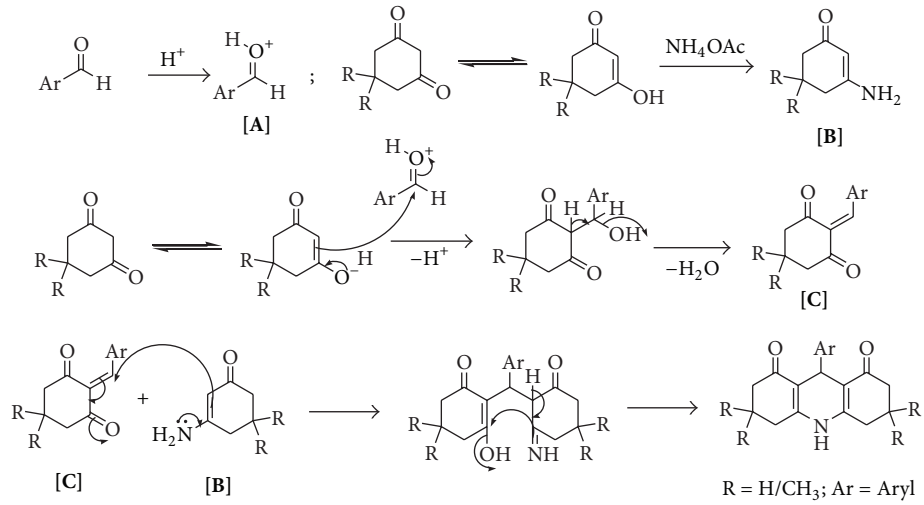
Table 3: Comparing the catalytic efficiency of P(4-VPH)HSO_4 for the synthesis of **6b** with some reported catalysts.

Entry[a]	Catalyst (g)	Reaction conditions	Time (min)	Yield (%)[Ref.]
1	L-Proline (0.041 g)	H_2O, Reflux	120	88 [19]
2	$Zn(OAc)\cdot 2H_2O$ (0.0172 g)	H_2O, Reflux	120	91 [19]
3	CAN (0.0274 g)	PEG 400, Rt	25	92 [16]
4	$InCl_3$ (0.0022 g)	EtOH, Rt	27	93 [17]
5	$CeCl_3\cdot 7H_2O$ (0.0182 g)	Ionic liquid, 100°C	180	89 [18]
6	MSA[a] (0.0096 g)	Solvent-free, 120°C	60	92 [23]
7	CBSA[b] (0.0056 g)	Solvent-free, 100°C	15	90 [27]
8	P(4-VPH)HSO_4 (0.02 g)	H_2O, 80°C	10	95 [present work]

[a]MSA: methanesulfonic acid; [b]CBSA: carbon-based solid acid
For comparison, mole percentages were converted into grams.

3. Experimental

All the chemicals were procured from Aldrich/Merck, and solvents were used without further purification. Melting points were recorded on Stuart SMP30 apparatus and are uncorrected. Thin layer chromatography was performed with F254 silica-gel precoated sheets using hexane/ethyl acetate (8 : 2) as eluent, visualized by UV light and iodine vapor. Products were characterized by comparing with the authentic samples and by spectral data (IR, ^{1}H NMR, and Mass). IR spectra were recorded on Perkin-Elmer 100S spectrophotometer using KBr disk. ^{1}H NMR spectra were recorded on Bruker 400 MHz spectrometer using DMSO-d_6 as solvent and TMS as internal standard. Elemental analysis was performed on a Carlo Erba model EA1108. Mass spectra were recorded on a Jeol JMSD-300 spectrometer.



Scheme 2: Proposed mechanism for the formation of 1,8-acridinediones.

3.1. General Procedure for the Synthesis of 1,8-Acridinediones. To a mixture of 1,3-cyclohexanedione/dimedone (2 mmol), arylaldehyde (1 mmol), and ammonium acetate (3 mmol) in 5 mL of water, 0.02 g of poly(4-vinylpyridinium)hydrogen sulfate was added and stirred at 80°C for an appropriate time as shown in Table 2. After completion of the reaction (TLC monitoring), the solid separated out was filtered, washed with water, dried, and recrytallized from ethanol/acetic acid to afford the pure 1,8-acridinediones in excellent yields. Aqueous layer containing catalyst was recovered under reduced pressure, dried, and reused for additional five times for subsequent reactions.

3.2. Spectral Data of New Compounds

*3.2.1. 9-(3,4-Dimethoxyphenyl)-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (**5e**).* Pale yellow solid; IR (KBr) υ_{max} (cm^{-1}): 3417 (NH), 1635 (C=O), 1597 (C=C), 1026 (C–O–C); ^{1}H NMR (400 MHz, DMSO-d_6): δ 1.75–1.79 (m, 2H), 1.83–1.90 (m, 2H), 2.16–2.19 (m, 4H), 2.45–2.48 (m, 4H), 3.47 (s, 3H), 3.49 (s, 3H), 4.83 (s, 1H), 6.47–6.49 (m, 1H), 6.62 (d, J = 8.0 Hz, 1H), 6.74 (s, 1H), 9.42 (s, 1H); MS (ESI) *m/z*: 354 $(M+H)^+$; anal. calcd. for $C_{21}H_{23}NO_4$: C, 71.37; H, 6.56; N, 3.96; found: C, 71.09; H, 6.71; N, 3.76.

*3.2.2. 9-(4-Hydroxy-3-methoxyphenyl)-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (**5g**).* Pale yellow solid; IR (KBr) υ_{max} (cm^{-1}): 3411 (NH), 3302 (OH), 1637 (C=O), 1596 (C=C), 1132 (C–O–C); ^{1}H NMR (400 MHz, DMSO-d_6): δ 1.76–1.80 (m, 2H), 1.86–1.91 (m, 2H), 2.17–2.20 (m, 4H), 2.45–2.49 (m, 4H), 3.47 (s, 3H), 4.80 (s, 1H), 6.46–6.47 (m, 1H), 6.55 (d, J = 8.0 Hz, 1H), 6.73 (s, 1H), 8.49 (s, 1H), 9.41 (s, 1H); MS (ESI) *m/z*: 340 $(M + H)^+$; anal. calcd. for $C_{20}H_{21}NO_4$: C, 70.78; H, 6.24; N, 4.13; found: C, 70.54; H, 6.43; N, 4.22.

*3.2.3. 9-(3-Ethoxy-4-hydroxyphenyl)-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (**5h**).* Red solid; IR (KBr) υ_{max} (cm^{-1}): 3414 (NH), 3329 (OH), 1644 (C=O), 1591 (C=C), 1119 (C–O–C); ^{1}H NMR (400 MHz, DMSO-d_6): δ 1.29 (t, J = 7.2 Hz, 3H), 1.77–1.81 (m, 2H), 1.88–1.93 (m, 2H), 2.19–2.22 (m, 4H), 2.47–2.49 (m, 4H), 3.88–3.93 (m, 2H), 4.81 (s, 1H), 6.45–6.47 (m, 1H), 6.55 (d, J = 8.0 Hz, 1H), 6.71 (s, 1H), 8.48 (s, 1H), 9.36 (s, 1H); MS (ESI) *m/z*: 354 $(M + H)^+$; anal. calcd. for $C_{21}H_{23}NO_4$: C, 71.37; H, 6.56; N, 3.96; found: C, 71.18; H, 6.69; N, 3.80.

*3.2.4. 9-(5-Bromo-2-hydroxyphenyl)-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (**5i**).* Pale yellow solid; IR (KBr) υ_{max} (cm^{-1}): 3414 (NH), 3279 (OH), 1625 (C=O), 1593 (C=C), 615 (C–Br); ^{1}H NMR (400 MHz, DMSO-d_6): δ 1.78–1.83 (m, 2H), 1.92–1.96 (m, 2H), 2.26–2.30 (m, 4H), 2.55–2.61 (m, 4H), 4.84 (s, 1H), 6.65 (d, J = 8.4 Hz, 1H), 6.87 (d, J = 8.4 Hz, 1H), 7.09–7.12 (m, 1H), 9.82 (s, 1H), 9.91 (s, 1H); MS (ESI) *m/z*: 411 $(M + Na)^+$; anal. calcd. for $C_{19}H_{18}BrNO_3$: C, 58.78; H, 4.67; N, 3.61; found: C, 58.62; H, 4.83; N, 3.79.

*3.2.5. 9-(3,5-Dibromo-2-hydroxyphenyl)-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (**5j**).* Yellow solid; IR (KBr) υ_{max}(cm^{-1}): 3415 (NH), 3285 (OH), 1620 (C=O), 1580 (C=C), 616 (C–Br); ^{1}H NMR (400 MHz, DMSO-d_6): δ 1.83–1.91 (m, 2H), 1.93–1.97 (m, 2H), 2.30–2.33 (m, 4H), 2.57–2.64 (m, 4H), 4.85 (s, 1H), 6.83 (s, 1H), 7.49 (s, 1H), 9.91 (s, 1H), 10.72 (s, 1H); MS (ESI) *m/z*: 468 $(M + H)^+$; anal. calcd. for $C_{19}H_{17}Br_2NO_3$: C, 48.85; H, 3.67; N, 3.00; found: C, 48.64; H, 3.86; N, 3.12.

*3.2.6. 9-(5-Chloro-2-hydroxyphenyl)-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (**5k**).* Pale yellow solid; IR (KBr) υ_{max} (cm^{-1}): 3414 (NH), 3280 (OH), 1622 (C=O), 1593 (C=C), 617 (C–Cl); ^{1}H NMR (400 MHz, DMSO-d_6): δ 1.79–1.82 (m, 2H), 1.92–1.96 (m, 2H), 2.26–2.29 (m, 4H), 2.54–2.60 (m, 4H), 4.84 (s, 1H), 6.70 (d, J = 8.4 Hz, 1H), 6.74 (d, J = 8.4 Hz, 1H), 6.97–7.00 (m, 1H), 9.84 (s, 1H), 9.89 (s, 1H); MS (ESI) *m/z*: 344 $(M + H)^+$; anal. calcd. for $C_{19}H_{18}ClNO_3$: C, 66.38; H, 5.28; N, 4.07; found: C, 66.17; H, 5.40; N, 4.26.

*3.2.7. 9-(3-Ethoxy-4-hydroxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (**6h**).* Pale yellow solid; IR (KBr) υ_{max} (cm^{-1}): 3416 (NH), 3277 (OH), 1620 (C=O), 1598 (C=C), 1122 (C–O–C); ^{1}H NMR (400 MHz, DMSO-d_6): δ 0.88 (s, 6H), 1.01 (s, 6H), 1.28 (t, J = 7.2 Hz, 3H), 1.99 (d, J = 16.0 Hz, 2H), 2.17 (d, J = 16.0 Hz, 2H), 2.30 (d, J = 16.8 Hz, 2H), 2.43 (d, J = 16.8 Hz, 2H), 3.85–3.91 (m, 2H), 4.70 (s, 1H), 6.49–6.56 (m, 2H), 6.68 (s, 1H), 8.48 (s, 1H), 9.20 (s, 1H); MS (ESI) m/z: 432 $(M + Na)^+$; anal. calcd. for $C_{25}H_{31}NO_4$: C, 73.32; H, 7.63; N, 3.42; found: C, 73.18; H, 7.77; N, 3.56.

*3.2.8. 9-(5-Bromo-2-hydroxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (**6i**).* White solid; IR (KBr) υ_{max} (cm^{-1}): 3415 (NH), 3279 (OH), 1622 (C=O), 1570 (C=C), 656 (C–Br); ^{1}H NMR (400 MHz, DMSO-d_6): δ 0.89 (s, 6H), 0.96 (s, 3H), 1.04 (s, 3H), 2.03 (d, J = 16.0 Hz, 2H), 2.22–2.57 (m, 6H), 5.03 (s, 1H), 6.95 (d, J = 8.8 Hz, 1H), 7.03 (s, 1H), 7.26–7.29 (m, 1H), 9.86 (s, 1H), 10.60 (s, 1H); MS (ESI) m/z: 445 $(M + H)^+$; anal. calcd. for $C_{23}H_{26}BrNO_3$: C, 62.17; H, 5.90; N, 3.15; found: C, 62.04; H, 5.99; N, 3.31.

*3.2.9. 9-(3,5-Dibromo-2-hydroxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (**6j**).* Pale yellow solid; IR (KBr) υ_{max} (cm^{-1}): 3416 (NH), 3284 (OH), 1635 (C=O), 1613 (C=C), 665 (C–Br); ^{1}H NMR (400 MHz, DMSO-d_6): δ 0.93 (s, 6H), 1.04 (s, 6H), 2.13 (d, J = 16.4 Hz, 2H), 2.30 (d, J = 16.4 Hz, 2H), 2.46 (d, J = 17.6 Hz, 4H), 4.82 (s, 1H), 6.88 (s, 1H), 7.49 (s, 1H), 9.87 (s, 1H), 10.63 (s, 1H); MS (ESI) m/z: 524 $(M + H)^+$; anal. calcd. for $C_{23}H_{25}Br_2NO_3$: C, 52.79; H, 4.82; N, 2.68; found: C, 52.65; H, 4.97; N, 2.80.

*3.2.10. 9-(5-Chloro-2-hydroxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (**6k**).* White solid; IR (KBr) υ_{max} (cm^{-1}): 3415 (NH), 3285 (OH), 1629 (C=O), 1579 (C=C), 656 (C–Cl); ^{1}H NMR (400 MHz, DMSO-d_6): δ 0.89 (s, 6H), 0.97 (s, 3H), 1.04 (s, 3H), 2.03 (d, J = 16.0 Hz, 2H), 2.24 (d, J = 16.0 Hz, 2H), 2.33 (d, J = 17.2 Hz, 2H), 2.54 (d, J = 17.2 Hz, 2H), 5.03 (s, 1H), 6.90 (s, 1H), 7.00 (d, J = 8.8 Hz, 1H), 7.14–7.17 (m, 1H), 9.82 (s, 1H), 10.58 (s, 1H); MS (ESI) m/z: 400 $(M + H)^+$; anal. calcd. for $C_{23}H_{26}ClNO_3$: C, 69.08; H, 6.55; N, 3.50; found: C, 68.92; H, 6.73; N, 3.68.

4. Conclusion

In conclusion, we have developed a simple, mild, and efficient protocol for the synthesis of 1,8-acridinediones utilizing poly(4-vinylpyridinium)hydrogen sulphate as catalyst in aqueous medium. Excellent yields in shorter reaction times, easy work-up procedure, environmentally begin nature, recovery, and reusability of the catalyst are the obvious advantages of this methodology.

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