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ACTIVATED FLY-ASH PROMOTED COST EFFECTIVE AND GREEN SYNTHESIS OF HEXAHYDROACRIDINE-1,8(*2H,5H*)-DIONES IN AQUEOUS MEDIUM

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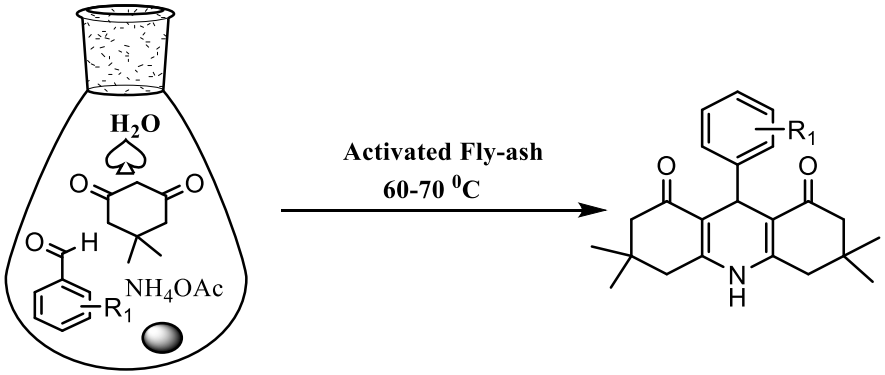
Abstract: A simple approach to the synthesis of hexahydroacridine-1,8(2*H*,5*H*)-dione via one-pot three component condensation of aromatic aldehydes, dimedone, and ammonium acetate in water using activated fly ash, (an industrial waste pollutant) as an efficient catalyst is described. Excellent yields, catalyst recovery and reusability, easy work-up, environmentally benign clean and green processes are attractive features of this protocol. All the synthesized hexahydroacridine-1,8(2*H*,5*H*)-dione were characterized on the basis of their melting-points, elemental analysis and spectral data.

Keywords: Activated Fly-ash, Green synthesis, multicomponent reactions, hexahydroacridine-1,8(2*H*,5*H*)-dione, 1, 4-dihydropyridine.

Introduction

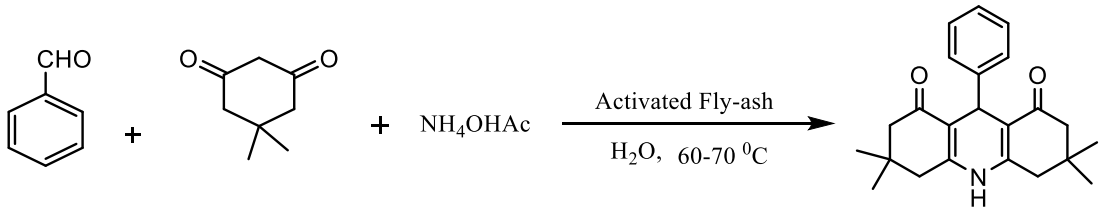
The development of an environmentally benign and efficient protocol for the synthesis of complex molecules and heterocycles privileged moieties has been a challenging task for synthetic chemists.[i] Furthermore, multi-component reactions (MCRs) are one of the most versatile method in advanced synthetic organic chemistry, as they have all the features that lead to an ideal synthesis, high atomic performance, quick and easy implementation, time and energy savings, eco-friendly, target oriented synthesis for variety of molecules.[ii] hexahydroacridine-1,8(2*H*,5*H*)-dione are a nitrogen-containing heterocyclic containing a 1, 4-dihydropyridine nucleus is used as laser dyes with very high efficiencies of photo initiators[iii] and a wide range of biologically active properties.[iv] The latest literature reviews reveals that these skeleton shows many biological activities such as anti-HIV,[v] multidrug resistance modulators,[vi] anti-tumor,[vii] anti-cancer,[viii] anti-glaucoma[ix] and anti-tubercular[x] etc. Their application in photochemical and electrochemical properties have been examined in details by various researchers.[xi] The Typical synthetic route for the synthesis of hexahydroacridine-1,8(2*H*,5*H*)-dione requires multicomponent condensation of two molecules of dimedone (5, 5-dimethyl-1,3-cyclohexadione) and specific aromatic aldehydes in the presence of ammonium acetate as a nitrogen source. A variety of metals and compounds were used as catalysts, in

different organic synthesis of hexahydroacridine-1,8(2*H*,5*H*)-dione such as palladium,[xii] nickel,[xiii] copper,[xiv] titanium,[xv] [Hmim]TFA,[xvi] CBSA,[xvii] (CAN),[xviii] and K_2CO_3[xix] However, almost all of these catalyst systems have limitations, such as the use of hazardous solvents, prolonged reaction time, expensive reagents, and catalyst, tough to recycle the catalyst used in procedure. Thus, there is a need for the development of new, efficient, and eco-friendly methods for the preparation of such compounds in high yields and under mild reaction conditions by using low costly catalyst. The great catalytic efficiency, operational simplicity, and the recyclability of activated Fly-ash were utilized for the synthesis of numerous drugs, pharmaceuticals and other biologically active moieties.[xx] The objective of present investigation is to activate the as-received Fly ash by a physical method followed by a thermal treatment and to examine the influence of catalyst. Herein we reported activated fly ash (an industrial waste pollutant) catalysed reaction for the synthesis of various hexahydroacridine-1,8(2*H*,5*H*)-dione via one-pot three component condensation of aromatic aldehydes, 5,5-dimethyl-1,3 cyclohexanedione (dimedone), and ammonium acetate in water as green solvent.



Result and Discussions

The hexahydroacridine-1,8(2*H*,5*H*)-dione compounds were synthesised in water from dimedone, ammonium acetate and substituted aromatic aldehydes (**4a**–**4o**) in a single process through multicomponent reactions (Knoevenagel condensation, Michael addition, dehydration and cyclization). These compounds were prepared by a one-pot reaction in a high-yield (upto 95%) process with simple workup procedure (**Scheme 1**).



Scheme 1. Synthesis of Hexahydroacridine-1,8(*2H,5H*)-dione derivatives in water.

Table 1. Comparison of the efficiency of catalysts (activated fly ash) in water solvents employed in the synthesis of Hexahydroacridine-1,8(*2H,5H*)-dione derivatives in water.

(**Scheme 1**)

Entry	Catalyst (gm)	Solvent Water (ml)	Tem (°C)	Time (h)	Yield (%)
1	No Catalyst	20	Rt	2.5	Trace
2	No Catalyst	10	Reflux	2.5	Trace
3	0.2	10	Rt	2.0	70

4	0.2	10	60-70	2.0	80
5	0.5	15	Rt	1.5	85
6	0.5	20	60-70	1.5	95

We investigated the effects of the catalyst in the model reaction catalysed by activated fly ash in various amounts in water solvent (**Table 1**) and the results are shown. It is obvious that 0.5 gm of catalyst is the best choice as it can be seen in (**Table 1**) using water as solvent. The model reaction without catalyst at refluxing and room temperature there was no yields in more than two hours whereas using 0.2 gm of catalyst in water at 70℃ temperatures provides 90-95% yields. The model reaction given highest yields 95% when we use 0.5 gm of catalyst in 1.5 hours. There are various aromatic aldehyde containing cyano, nitro, halogen (-F, -Cl, -Br), methyl, methoxy, hydroxyl, and some other moieties are used (**Table 2**). The reaction of dimedone, benzaldehyde, and ammonium acetate in the presence of activated fly ash, catalyst in water was selected as the model reaction. Other solvent we tested are outlined in **Table 3**. This reaction did not form products in the presence of other selected solvents

Table 2. General reaction for synthesis of hexahydroacridine-1,8(2*H*,5*H*)-dione[a]

Entry	R-	Compound	Time (min)	Yields (%)[b]	mp (°C)[c]
1.	H-	4a	60	94	289–290
2.	4-Chloro	4b	52	90	298–300
3.	2-Nitro	4c	60	93	296–298
4.	3-OH	4d	52	88	307–309
5.	4-Methyl	4e	58	93	278–280
6.	3-Methyl	4f	55	90	304-305
7.	3-Chloro	4g	58	89	289-290
8.	4-Floro	4h	64	93	247-248
9.	3,4-Dihydroxy	4i	66	90	308–309
10.	4-Cyno	4j	72	94	270–271
11.	4-Nitro	4k	70	93	298–300
12.	4-Hydroxy	4l	58	95	306–307
13.	3-Nitro	4m	67	92	286-288
14.	3-Floro	4n	70	94	274-275
15.	4-Methoxy	4o	78	95	272-273

a Reaction condition: aromatic aldehydes **1** (1 mmol), dimedone **2** (2 mmol), ammonium acetate **3** (1mmol), and activated fly-ash (0.5 gm) in water (15 mL)

b Isolated pure products

c All synthesized compounds were characterized with spectral analysis and melting points were consistent with literature value (xxi-xxv)

Table 3. Comparison of various solvents employed in the synthesis of Hexahydroacridine-1,8(*2H,5H*)-dione derivatives in water. in catalyst activated fly ash.

Entry	Solvent	Time (h)	Yield (%)
1	Water	1.5	95
2	Ethanol	2.5	75
3	Methanol	2.5	68
4	THF	3.0	55
5	Ethyl acetate	3.0	42
6	Neat	5.0	Trace

Table 4: Comparison of the catalytic efficiency of Activated fly ash catalyst and other catalyst for synthesis of Hexahydroacridine-1,8(*2H,5H*)-dione derivatives in water.

Entry	Catalyst	Time (min)	Solvent	Temperature °C	Yields (%)
1	Ferric chloride	120	Ethanol	Reflux	60
2	L-Proline	240	Ethanol	Reflux	80
3	Urea	360	H_2O:EtOH	RT	85
4	Red sea sand	360	Ethanol	Reflux	84
5	NaBr	20	No Solvent	70 MWI	90
6	[Ch][OH]	60	Water	80	60
7	Nano-Zinc	90	Water	60-90	90
8	Activated fly-ash	90	Water	60-70	95

In most of the cases, many disclosed methods endure the suffering from many drawbacks such as using hazardous catalyst and environmental degradation, non-recyclable catalysts, laboured work-up procedure, long reaction times, and low separated yields etc. This investigation revels us to trying an improved research for the synthesis of hexahydroacridine-1,8(*2H,5H*)-dione derivatives catalysed by activated fly-ash catalyst in water.

Experimental

Materials and methods

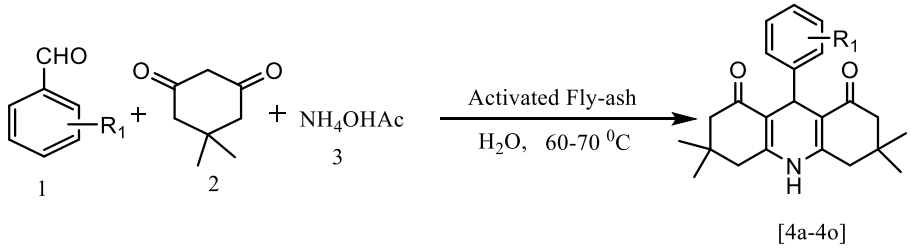
Fly-ash was collected from a local sugar factory Sugar factory Azamgarh, Uttar Pradesh, India. All chemicals were purchased from Aldrich and Alfa Aesar and were used without purification. NMR spectra were recorded on 500MHz for ^{1}HNMR and 125.75 MHz for ^{13}CNMR using TMS as an internal reference for both of the cases. Here chemical shifts were reported in parts per million (ppm) and melting points were monitored by open glass capillary method and were uncorrected. Physical and spectral data of the obtained products were compared with reported literature data.

Preparation of activated fly ash (Catalyst):

The fly-ash collected has been activated by the procedure reported in the literature[20]. The fly-ash sieved in a 100 mesh sieve to remove any coarser and foreign particles (external impurities) and then mechanically ground in a ball mill to fine powder. The particle size distribution was found to be between 40 and 90 μm. Finely the ground Fly ash was kept at a temperature of about 900-1000 ^{o}C in a silica crucible for 60 min for activation. The sulphur, carbon, and other impurities were removed by thermal activation and the resultant Fly ash used as catalyst.

General procedure for synthesis of Hexahydroacridine-1,8(2H,5H)-dione Derivatives

The mixture of substituted aromatic aldehydes **1** (1 mmol), dimedone **2** (2 mmol), ammonium acetate **3** (2 mmol), and activated fly-ash (0.5gm) in water (20 mL) was heated at 60-70°C for the appropriate time (1-1.5 h) as monitored by TLC. After completion of the reaction, water from the reaction mixture was evaporated. The desired product in the residue thus obtained was dissolved in ethanol and the insoluble catalyst was recovered by filtering it from the ethanolic solution. The filtrate was evaporated under vacuum and crude product thus obtained was recrystallized from aqueous ethanol to get the pure product (**4a–4o**). The recovered catalyst was washed with ethyl acetate (10mL) dried in an oven at 100°C for 1h and reused for further reactions. All synthesized compounds were characterized with spectral analysis and melting points were consistent with literature value.



Scheme 4. General reaction for synthesis of hexahydroacridine-1,8(2*H*,5*H*)-dione

Analytical data for the products

All obtained products are known and compared with literature data. The purity of the obtain materials are confirmed by spectral analysis. (m.p. IR, ^{1}H and ^{13}C NMR)[21-25].

3,3,6,6-tetramethyl-9-phenyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4a) : White solid; m.p: 289–290 ℃ IR ; IR (KBr, cm^{-1}): νmax; 1588, 2882 and 3066, ^{1}H-NMR ($CDCl_3$, 500 MHz) δ ppm:1.12 (s, 6H, 2x CH_3), 1.24 (s, 6H, 2xMe), 2.30-2.45 (m, 8H, 4xCH_2), 7.14-7.28 (m, 5H, Ar-H), 11.94 (s,1H, NH); ^{13}C-NMR ($CDCl_3$, 125 MHz) δ ppm: 27.44, 29.6, 31.48, 32.76, 46.42, 47.08, 115.64, 125.87, 126.77, 128.28, 138.00, 189.45, 190.55.

9-(4-chlorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4b) : White solid; m.p: 298–300 ℃; IR (KBr, cm^{-1}): νmax; 678, 1590 , 2098 and 3049 (NH); ^{1}H-NMR ($CDCl_3$, 500 MHz) δ ppm: 1.08 (s, 6H, 2xCH_3), 1.21 (s, 6H, 2xCH_3), 2.29-2.47 (m, 8H, 4xCH_2), 5.46 (s, 1H, CH), 7.05-7.00 (d, 2H, Ar-H, J=8.0 Hz), 7.22-7.25 (d, 2H, Ar-H, J= 8.0 Hz), 11.80 (s, 1H, NH); ^{13}C-NMR ($CDCl_3$, 125 MHz) δ ppm: 27.25, 27.40, 29.24, 29.55, 31.33, 31.40, 32.18, 32.45, 40.86, 46.42, 47.05, 50.68, 115.24, 115.34, 128.01, 128.33,129.78, 131.55, 131.90, 136.75, 142.60, 162.44, 189.38, 190.50, 196.28.

3,3,6,6-tetramethyl-9-(2-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4c): Yellow solid; m.p: 296–298 ℃; IR (KBr, cm^{-1}):νmax; 1345, 1526, 1728, 2948, 3378 ; ^{1}H-NMR ($CDCl_3$, 500 MHz) δ ppm: 1.04 (s, 6H,2xCH3), 1.14 (s, 6H, 2xCH3), 2.20-2.45 (m, 8H, 4xCH2), 6.04 (s, 1H, -CH), 7.24-7.35 (m, 4H, Ar-H), 11.66 (s, 1H, NH); ^{13}C-NMR ($CDCl_3$, 125 MHz) δ ppm: 28.18, 28.50, 30.10, 31.39, 46.22, 46.84, 114.62, 124.23,127.28, 129.63, 131.33, 132.12, 149.77, 189.14, 190.24.

3,3,6,6-Tetramethyl-9-(m-tolyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4f) : Yellow solid, mp 304-305 ℃, IR (KBr, cm^{-1}): νmax; 3393, 3280, 3208, 2950, 2881, 1724, 1610, 1475, 1382, 1221, 1141; ^{1}H-NMR ($CDCl_3$, 500 MHz) δ ppm: 0.95 (6H, s, 2x CH3), 1.1 (6H, s, 2xCH3), 1.65 (3H, s, CH3), 2.0–2.5 (8H, m, 4xCH2), 5.10 (1H, s, CH), 6.85, (1H, d, Ar-H), 7.09 (1H, m, Ar-H), 7.18 (1H, d, Ar-H), 7.44 (1H, s, Ar-H); ^{13}C-NMR ($CDCl_3$, 125 MHz) δ ppm: 20.55, 26.03, 31.57, 32.35, 39.29, 49.90, 112.38, 125.72, 127.19, 128.01, 129.44, 136.45, 145.46, 148.05, 194.88.

3,3,6,6-Tetramethyl-9-(3-chlorophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4g): white solid, mp 289-290 ℃, ; IR (KBr, cm^{-1}):νmax; 3298, 3194, 2957, 1648, 1625, 1594, 1560, 1465, 1382, 1222. ^{1}H-NMR ($CDCl_3$, 500 MHz) δ ppm: 1.18 (6H, s, 2xCH3), 1.34 (6H, s, 2xCH3), 2.32–2.60 (8H, m, 4xCH2), 5.78 (s, 1H, CH), 6.64–7.36 (4H, m, Ar-H), 11.75 (1H, s, NH), ^{13}C-NMR ($CDCl_3$, 125 MHz) δ ppm:26.50, 28.75, 32.46, 33.78, 45.35, 46.18, 112.95, 121.88, 123.90, 125.74, 128.82, 131.24, 142.19, 187.36, 191.28.

3,3,6,6-Tetramethyl-9-(4-fluorophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (4h): White solid, mp 247-248 ℃, IR (KBr, cm^{-1}):νmax; 3276, 3208, 3072, 2954, 1648, 1608, 1612, 1481, 1365, 1220, 1145. ^{1}H-NMR ($CDCl_3$, 500 MHz) δ ppm: 0.98 (6H, s, 2xCH3), 1.24 (6H, s 2xCH3), 2.06-2.15 (4H, dd, 2xCH2), 2.37 (4H, s, 2xCH2), 4.98 (1H, s, CH), 6.75-6.98 (2H, t, Ar-H), 7.15-7.25 (2H, d, Ar-H), 8.78 (1H, s, NH). ^{13}C-NMR ($CDCl_3$, 125 MHz) δ ppm:

28.19, 29.96, 31.64, 33.20, 49.45, 112.15, 125.22, 126.97, 128.23, 146.88, 148.18, 148.76, and 193.36.

4-(3,3,6,6-tetramethyl-1,8-dioxo-1,2,3,4,5,6,7,8,9,10-decahydroacridin-9-yl)benzonitrile(4j) Dark Yellow crystals, m.p. 270–271 ℃. IR (KBr, cm^{-1}):νmax; 3295, 2975, 2284, 1675, and 1612; ^{1}H-NMR ($CDCl_3$, 500 MHz) δ ppm: 11.95 (s, 1H, -NH), 7.85 (d, 2H,Ar), 7.68 (d, 2H,Ar), 4.84 (s, 1H, CH), 2.65 (s,4H, 2xCH2), 2.18 (s, 4H 2xCH2),1.08 (s, 6H, 2xCH3), and 1.06 (s, 6H, 2xCH3); ^{13}C-NMR ($CDCl_3$, 125 MHz) δ ppm:195.15, 151.14, 150.58, 136.28, 130.58, 119.10, 112.85, 111.28, 52.74, 41.15, 33.68, 33.79, and 28.78.

9-(3-Nitrophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-(2H,5H)-acridine-1,8-dione (4m) : White solid mp 286-288 ℃, ; IR (KBr, cm^{-1}):νmax; 3292, 2972, 1702, 1616; ^{1}H-NMR (DMSO d_6, 500 MHz) δ ppm: 0.92 (s, 6H, 2×CH3), 0.95 (s,6H, 2×CH3), 10.92 (s, 1H, NH), 7.85–8.75 (m, 2H, Ar-H), 7.217.37 (m, 2H, Ar-H), 5.38 (s, 1H, CH), 2.24–2.44(m, 8H, 4×CH2); ^{13}C NMR (125 MHz, DMSO-d6) δ: 27.45, 29.42, 31.28, 32.75, 44.3, 47.28, 115.18, 124.33,127.42, 128.86, 135.76, 143.50, 189.48, 192.50.

9-(3-Fluorophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-(2H,5H)-acridine-1,8-dione (4n): Yellowish solid mp 274-275 ℃, ; IR (KBr, cm^{-1}):νmax; 3285, 2982, 1698, 1612; ^{1}H NMR(500 MHz, DMSO-d6) δ: 0.90 (s, 6H, 2×CH3), 1.00 (s,6H, 2×CH3), 10.87 (s, 1H, NH), 7.10–7.20 (m, 4H, Ar-H), 5.52 (s, 1H, CH), 2.15–2.25 (m, 8H, 4×CH2); ^{13}CNMR (125 MHz, DMSO-d6) δ ppm: 27.25, 29.44, 31.10, 32.50,44.35, 49.28, 115.13, 127.45, 129.88, 131.20, 138.27, 188.19,191.90.

9-(4-Methoxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydro-(2H,5H)-acridine-1,8-dione (4o): White solid mp 272-273 ℃, ; IR (KBr, cm^{-1}):νmax; 3285, 2964, 1648, 1620; ^{1}H NMR(500 MHz, DMSO-d6) δ: 0.89 (s, 6H, 2×CH3), 0.99 (s,6H, 2×CH3), 10.92 (s, 1H, NH), 7.15–7.30 (m, 4H,Ar-H), 5.31 (s, 1H, CH), 2.15–2.25 (m, 8H, 4×CH2), 3.75 (s, 3H, OCH3), ^{13}C NMR (125 MHz, DMSO-d6) δ ppm: 21.85, 27.12, 28.22, 30.49, 32.65, 44.87, 48.22, 56.52, 1 13.60,115.35, 127.22, 128.55, 134.26, 140.70, 146.50, 189.33, 193.88.

CONCLUSION

We have developed a new, easy, efficient method for eco-compatible preparation of substituted hexahydroacridine-1,8(2*H*,5*H*)-dione in an aqueous medium with activated fly ash, (an industrial waste pollutant) as an efficient catalyst. The mildness of the conversion, the experimental simplicity, compatibility with various functional groups, excellent yield (up to 95%) of product and the easy work-up procedure make this approach attractive for synthesizing a variety of such derivatives.

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