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$Al_2(SO_4)_3.18H_2O$ as an Efficient and Environmentally Benign Natural Catalyst for Facile and Solvent-free Synthesis of Xanthene Derivatives

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Abstract

An environmentally friendly, facile and solvent-free synthetic route for aluminum sulfate octadecahydrate ($Al_2(SO_4)_3.18H_2O$) catalyzed one-pot multi-component synthesis of 12-aryl-tetrahydrobenzo[*α*]xanthene-11-ones, 1,8-dioxo-octahydroxanthenes and 14-aryl-14*H*-dibenzo[*α,j*]xanthenes has been developed. The readily, easy to handle, non-toxic, environmental friendly and low-cost catalyst, easily separated with no column chromatographic separation, short reaction times and high to excellent yields is an option for the economical, mild and facile synthesis of these rings.

Keywords: *Aluminum sulfate octadecahydrate ($Al_2(SO_4)_3.18H_2O$), Facile, eco-friendly and solvent-free synthesis, 12-aryl-tetrahydrobenzo[α]xanthene-11-ones, 1,8-dioxo-octahydroxanthenes, 14-aryl-14H-dibenzo[α,j]xanthenes.*

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Introduction

In recent years, organic chemists have focused their attention towards multi-component domino reactions (MCRs) [1-8] for the synthesis of heterocyclic compounds due to a broad range of notable advantages such as non-toxic substrate and environmental friendly, atom-economy, simple work-up, one-pot and low-cost.

Synthesis of heterocyclic compounds has attracted great interests due to their wide applicability in life and nature. The compounds with xanthene derivatives ring systems are reported as antiplasmodial [9], antiviral [10] and anti-inflammatory [11]. Besides, these heterocyclic molecules have been widely used as pH sensitive fluorescent materials for visualization of biomolecules [12, 13], laser technology [14, 15], luminescent dyes [16, 17] and sensitizers in photodynamic therapy [18, 19].

In recent decades, a number of methodologies for the preparation of these compounds have been reported that is including various catalysts [20-37]. Some of these methodologies have limitations such as difficult work-up, toxic and expensive catalysts, low yields, the use of strongly acidic conditions and long time reactions. As part of our ongoing research program on the development of eco-safe methodologies [38-41], herein, we report a clean and facile one-pot synthesis of 12-aryl-tetrahydrobenzo[*α*]xanthene-11-ones, 1,8-dioxo-octahydroxanthenes and 14-aryl-14*H*-dibenzo[*α,j*]xanthenes in the presence of catalytic amount of aluminum sulfate octadecahydrate ($Al_2(SO_4)_3.18H_2O$) as an eco-safe and highly efficient catalyst under solvent-free conditions. Short reaction times, high to excellent yields, eco-friendly, one-pot, efficient, readily, low-cost and non-toxic catalyst that makes our protocol alternative in comparison to some of the earlier reported methods.

Experimental

General

Melting points all compounds were determined using an Electro thermal 9100 apparatus. Also, nuclear magnetic resonance, ^{1}H NMR spectra were recorded on a Bruker DRX-400 Avance instrument with $CDCl_3$ as solvents. In this article, all reagents and solvents were purchased from Merck, Fluka and Acros chemical companies were used without further purification.

General procedure for preparation of 12-aryl-tetrahydrobenzo[α]xanthene-11-ones, 1,8-dioxo-octahydroxanthenes and 14-aryl-14H-dibenzo[α,j]xanthenes

Synthesis of 12-aryl-tetrahydrobenzo[α]xanthene-11-ones (**4**)

A mixture of *β*-naphthol (**1**, 1.0 mmol), aromatic aldehyde derivatives (**2**, 1.0 mmol), dimedone (**3**, 1.0 mmol) and $Al_2(SO_4)_3.18H_2O$ (10 mol %) was heated at 80 °C for appropriate time. After completion of the reaction (by thin layer chromatography TLC) the mixture was cooled to r.t. and ethanol was added and the precipitated was separated by filtration and solid was recrystallized from ethanol to afford the pure products (**4a- m**).

Synthesis of 1,8-dioxo-octahydroxanthenes (**6**)

A mixture of dimedone (**3**, 2.0 mmol), aromatic aldehyde derivatives (**5**, 1.0 mmol), and $Al_2(SO_4)_3.18H_2O$ (10 mol %) was heated at 80 °C for appropriate time. After completion of the reaction (by thin layer chromatography TLC) the mixture was cooled to r.t. and ethanol was added and the precipitated was separated by filtration and solid was recrystallized from ethanol to afford the pure products (**6a- m**).

Synthesis of 14-aryl-14H-dibenzo[α,j]xanthenes (**8**)

A mixture of *β*-naphthol (**1**, 2.0 mmol), aromatic aldehyde derivatives (**7**, 1.0 mmol) and $Al_2(SO_4)_3.18H_2O$ (10 mol %) was heated at 80 °C for appropriate time. After completion of the reaction (by thin layer chromatography TLC) the mixture was cooled to r.t. and ethanol was added and the precipitated was separated by filtration and solid was recrystallized from ethanol to afford the pure products (**8a- m**). The products have been characterized by melting points and ^1H NMR spectroscopy.

Spectra data some of known products are represented below:

9,9-dimethyl-12-(4-methoxyphenyl)-8,9,10,12-tetrahydrobenzo[a]xanthen-11-one ***(4l)*****:**

Yield: 85%; m.p. 204-206 °C.

^1HNMR (400 MHz, $CDCl_3$): 0.99 (3H, s, CH_3), 1.12 (3H, s, CH_3), 2.16-2.35 (2H, m, CH_2), 2.58 (2H, s, CH_2), 3.71 (3H, s,OCH_3), 5.68 (1H, s, CHAr), 6.72 (2H, d, *J* = 8.4 Hz, ArH), 7.21-7.47 (5H, m, ArH), 7.85 (2H, t, *J*= 9.2 Hz, ArH), 8.01 (1H, d, *J*= 8.4 Hz, ArH).

3,3,6,6-tetramethyl-9-(4-methoxyphenyl)-1,2,3,4,5,6,7,8-octahydroxanthene-1,8-dione ***(6l):***

Yield: 87%; m.p. 240-242 °C.

^1H NMR (400 MHz, $CDCl_3$): 1.01 (6H, s, $2CH_3$), 1.12 (6H, s, $2CH_3$), 2.16-2.27 (4H, q, *J*= 8.2 Hz, $2CH_2$), 2.47 (4H, s, $2CH_2$), 3.75 (3H, s, OCH_3), 4.72 (1H, s, CH), 6.77 (2H, d, *J*= 8.8Hz, ArH), 7.22 (2H, d, *J*= 8.8 Hz, ArH).

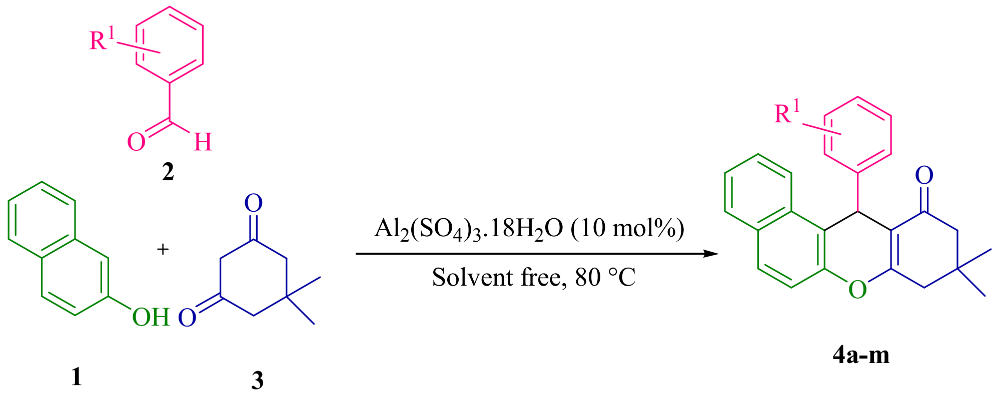
14-(4-methylphenyl)-7,14-dihydro-dibenzo[a,j]xanthenes ***(8e)*****:**

Yield: 90%; m.p. 225-227 °C.

^{1}H NMR (400 MHz, $CDCl_3$): 2.18 (3H, s, CH_3), 6.47 (1H, s, CH), 6.81(1H, d, *J*=7.6 Hz, ArH), 6.81(1H, t, *J*=7.6 Hz, ArH), 7.40-7.85 (12H, m, ArH), 8.42 (2H,d, *J*= 8.8 Hz, ArH).

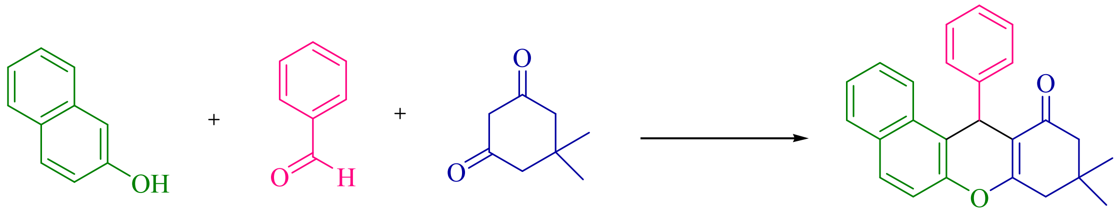
Results and discussion

By employing a reaction of *β*-naphthol (**1**), benzaldehyde (**2**) and dimedone (**3**), the reaction conditions were optimized. It is noticed that the reaction proceeded smoothly with 10 mol% $Al_2(SO_4)_3.18H_2O$. Thus, after 15 min at 80 °C, the desired product was obtained in 88% yield under solvent-free condition. Initially, a comparative study was carried out with various temperatures and various molar catalysts in solvent-free condition (Table 1). It is noteworthy that in the absence of catalyst a very low amount of the desired product was formed (Table 1, entry 1). Also taking reaction in the room temperature gave lower yield and higher reaction time (Table 1, entry 4). The yield increased sharply when the temperature was raised from room temperature to 90 °C. Higher amounts of the catalyst up to 10 mol% and of the reaction temperature did not improve the results to a greater extent (Table 1, entry 9). After optimizing the conditions, to demonstrate the diversity of the $Al_2(SO_4)_3.18H_2O$ catalyst and to explore the scope of the process, the optimized conditions were applied to reactions of *β*-naphthol (**1**, 1.0 mmol) and dimedone (**3**, 1.0 mmol) with various aromatic aldehydes (**2**, 1.0 mmol) (Scheme1). All desired products were formed as shown in Table 2.



Scheme 1. Synthesis of 12-aryl-tetrahydrobenzo[*α*]xanthene-11-ones.

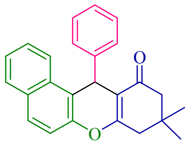
Table 1. Optimization of the reaction condition for the synthesis of **4a** [a]



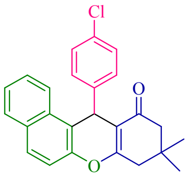
Entry	$Al_2(SO_4)_3.18H_2O$ (mol %)	Temperature (°C)	Time (min)	Isolated Yields (%)
1	Catalyst free	80	240	trace
2	5	80	30	59
3	10	80	15	88
4	10	rt	240	trace
5	10	40	40	37
6	10	60	35	61
7	10	70	20	74
8	10	90	15	88
9	15	80	15	90

[a] Reaction conditions: β-naphthol (1.0 mmol); benzaldehyde (1.0 mmol); dimedone (1.0 mmol) and $Al_2(SO_4)_3.18H_2O$ was heated under various temperatures for the appropriate time.

Table 2. $Al_2(SO_4)_3.18H_2O$ catalyzed synthesis of 12-aryl-tetrahydrobenzo[α]xanthene-11-ones.



4a



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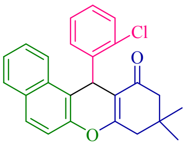

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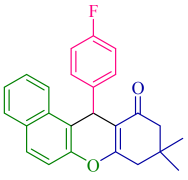

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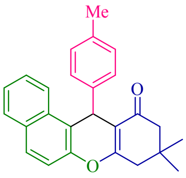

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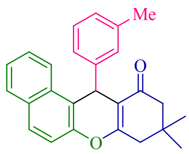

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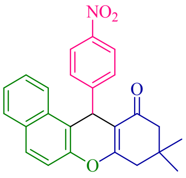

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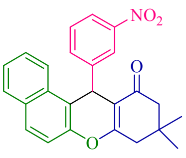
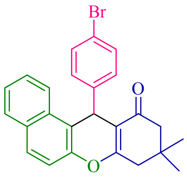
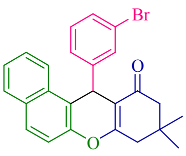


4h



4i



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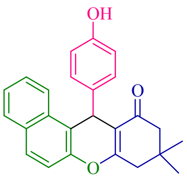

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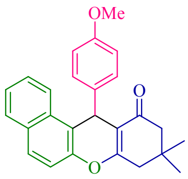

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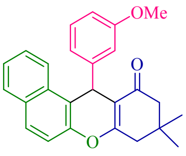
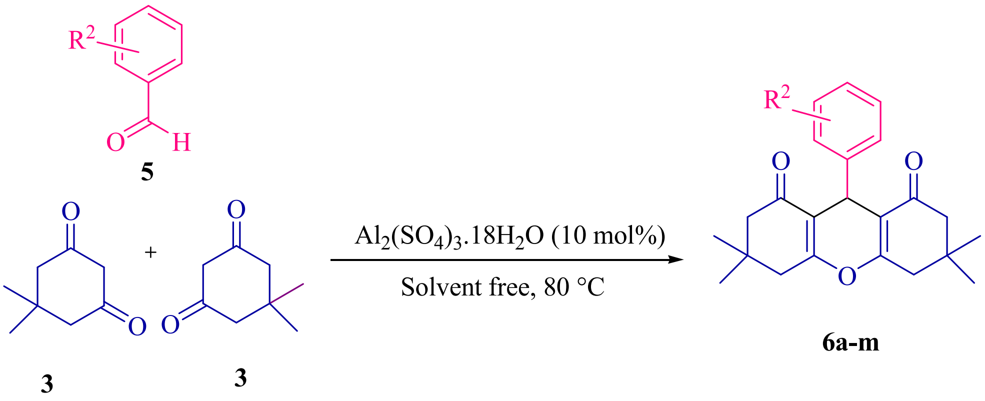


4m

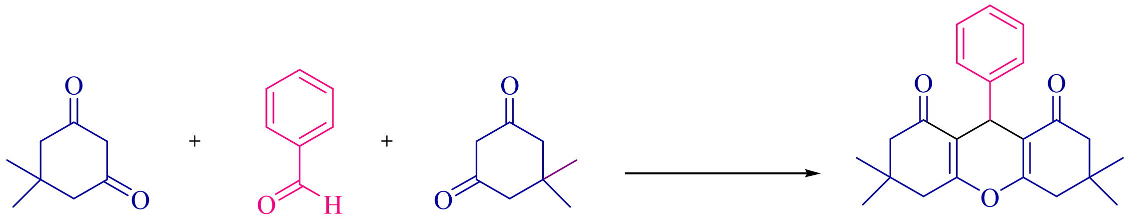
Entry	R^1	Product	Time (min)	Isolated Yields (%)	m.p.°C	m.p.°C Lit
1	C_6H_5	**4a**	15	88	150-152	148-150 [24]
2	4- Cl-C_6H_4	**4b**	20	83	174-176	176-178 [24]
3	2- Cl-C_6H_4	**4c**	20	85	179-181	179-180 [27]
4	4-F- C_6H_4	**4d**	10	92	183-185	184-185 [20]
5	4-Me- C_6H_4	**4e**	15	90	169-171	171-173 [24]
6	3-Me- C_6H_4	**4f**	15	91	180-182	178-180 [23]
7	4-O_2N-C_6H_4	**4g**	10	88	177-179	175-178 [26]
8	3-O_2N-C_6H_4	**4h**	10	91	169-171	167-169 [20]
9	4-Br- C_6H_4	**4i**	20	83	185-187	184-186 [24]
10	3-Br- C_6H_4	**4j**	20	85	164-166	161-164 [26]
11	4-OH- C_6H_4	**4k**	25	81	220-222	222-223 [24]
12	4-OMe- C_6H_4	**4l**	15	85	204-206	202-204 [24]
13	3-OMe- C_6H_4	**4m**	15	86	203-205	202-204 [24]

After the successful synthesis of 12-aryl-tetrahydrobenzo[α]xanthene-11-ones, we turned our attention toward the synthesis of 1,8-dioxo-octahydroxanthenes. Initially, in order to optimize the reaction conditions, the model reaction was carried out by using benzaldehyde (2.0 mmol) and dimedone (1.0 mmol) at various temperatures and in the presence of various molar $Al_2(SO_4)_3.18H_2O$ as catalyst and the results are listed in Table 3. However, only a trace amount of the product was formed in the absence of catalyst (Table 3, entry 1). Afterward, optimization of catalyst amounts was carried out in the model study by using different amounts of the $Al_2(SO_4)_3.18H_2O$. The highest yield was obtained by increasing the amount of catalyst from 5 mol% to 10 mol% (Table 3, entry 3). However, further increase of the molar amount of the catalyst from 10 mol% to 15 mol% did not significantly increase the yield of the product (Table 3, entry 9). Hence, the optimum concentration of $Al_2(SO_4)_3.18H_2O$ was chosen 10 mol% in the model to improve the yield of the target product, we carried out the test reaction in presence of various temperatures and the results are presented in Table 3. As Table 3 indicates, reasonable results were obtained when the reaction was performed using 10 mol % of the catalyst at 80 °C (Table 3, entry 3). To study the scope of the reaction, a series of aldehydes were employed. The results are shown in Table 4. In all cases, aromatic aldehydes substituted with either electron-donating or electron-withdrawing groups underwent the reaction smoothly and gave the products in high yields (Table 4) (Scheme 2).



Scheme 2. Synthesis of 1,8-dioxo-octahydroxanthenes.

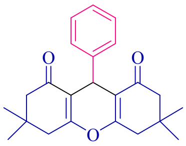
Table 3. Optimization of the reaction condition for the synthesis of **6a** [a]



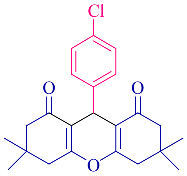
Entry	**$Al_2(SO_4)_3.18H_2O$ (mol %)**	**Temperature (°C)**	**Time (min)**	**Isolated Yields (%)**
1	Catalyst free	80	240	trace
2	5	80	25	65
3	**10**	**80**	**10**	**92**
4	10	rt	240	trace
5	10	40	45	47
6	10	60	25	61
7	10	70	15	79
8	10	90	10	94
9	15	80	10	93

[a] Reaction conditions: dimedone (2.0 mmol); benzaldehyde (1.0 mmol) and $Al_2(SO_4)_3.18H_2O$ was heated under various temperatures for the appropriate time.

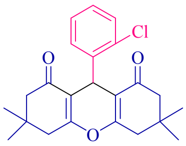
Table 4. $Al_2(SO_4)_3.18H_2O$ catalyzed synthesis of 1,8-dioxo-octahydroxanthenes.



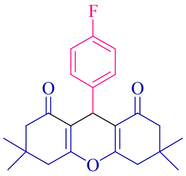
6a



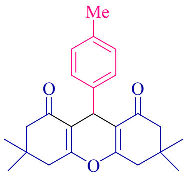
6b



6c



6d



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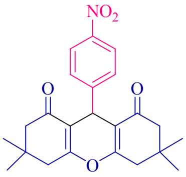

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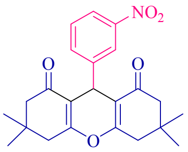

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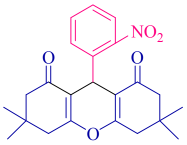

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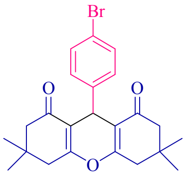
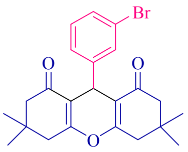


6i



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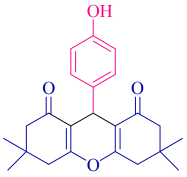

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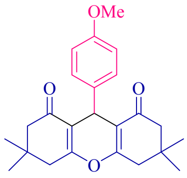

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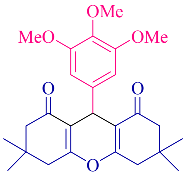
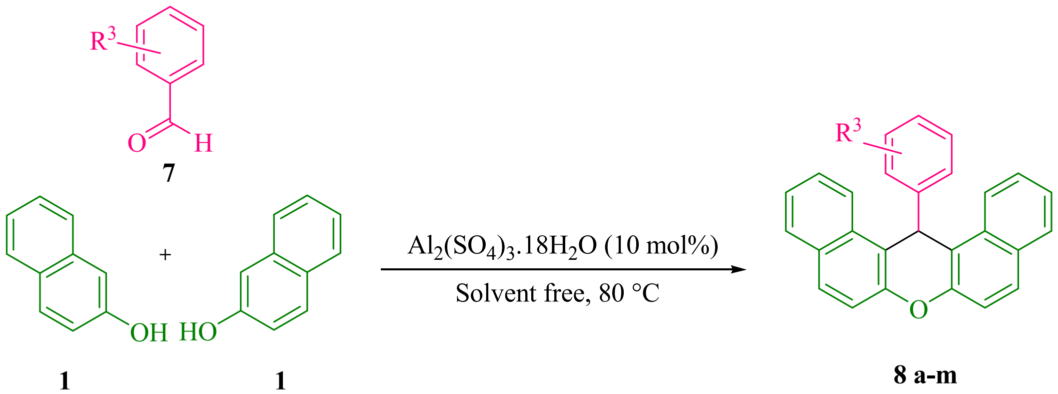


6m

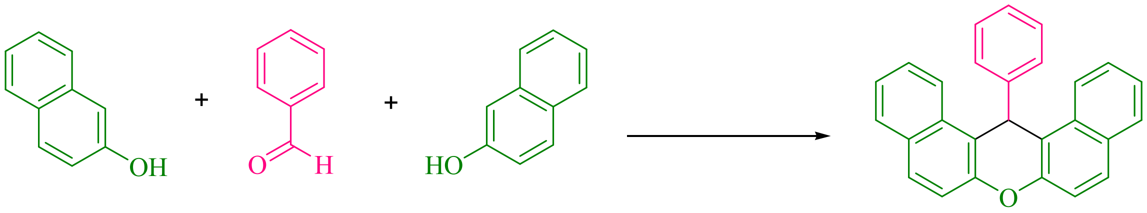
Entry	R^2	Product	Time (min)	Isolated Yields (%)	m.p.°C	m.p.°C
1	C_6H_5	**6a**	10	92	207-209	206-208 [24]
2	4- Cl-C_6H_4	**6b**	15	86	236-238	235-238 [20]
3	2- Cl-C_6H_4	**6c**	15	88	223-225	224-227 [20]
4	4-F- C_6H_4	**6d**	10	94	194-196	193-195 [24]
5	4-Me- C_6H_4	**6e**	10	89	215-217	216-218 [29]
6	4-O_2N-C_6H_4	**6f**	10	92	224-226	222-224 [29]
7	3-O_2N-C_6H_4	**6g**	10	93	172-174	171-172 [20]
8	2-O_2N-C_6H_4	**6h**	10	93	256-258	258-260 [29]
9	4-Br- C_6H_4	**6i**	15	82	240-242	239-241 [20]
10	3-Br- C_6H_4	**6j**	15	85	191-193	192-194 [29]
11	4-OH- C_6H_4	**6k**	20	81	245-247	246-248 [29]
12	4-OMe- C_6H_4	**6l**	15	87	240-242	241-243 [20]
13	3,4,5-$(OMe)_3$-C_6H_2	**6m**	20	89	188-200	186-188 [29]

As a part of our ongoing efforts towards the development of eco-safe procedure for the xanthene derivatives through multi-component reactions using $Al_2(SO_4)_3.18H_2O$ as catalyst. We have discovered an efficient and environmental friendly procedure for the synthesis of 14-aryl-14*H*-dibenzo[*α,j*]xanthenes. Initially, our investigation began with the evaluation of $Al_2(SO_4)_3.18H_2O$ as a catalyst in the reaction of *β*-naphthol (2.0 mmol) and benzaldehyde (1.0 mmol) at 80 °C in solvent-free conditions. In the absence of catalyst, the reaction proceeded sluggishly (Table 5, entry 1). The use of 5 mol% of $Al_2(SO_4)_3.18H_2O$ in this condition afforded a 61% yield (Table 5, entry 2) of the desired product. Optimization of the reaction conditions was undertaken to increase the yield employing different catalyst loadings in solvent-free conditions. The results were summarized in table 5. The yield was increased by using 10 mol% of $Al_2(SO_4)_3.18H_2O$ (Table 5, entry 3). However, the addition of 15 mol% of the catalyst was found to have a weak effect on the formation of 3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-9-phenyl-2*H*-xanthene-1,8(5*H*,9*H*)-dione (Table 5, entry 9). The effects of temperature were also examined on model reaction. Increasing of temperature to the point of 80 °C causes increase the yield or reaction, but in temperature higher than 80 °C, the results showed that a significant increase in the yield has not been observed (Table 5, entry 8). And, in continuation of our effort to develop xanthene derivatives synthesis, we report herein, a simple, mild and eco-safe synthesis of 14-aryl-14*H*-dibenzo[*α,j*]xanthenes *via* *β*-naphthol (**1**, 2.0 mmol) and aromatic aldehyde derivatives (**7**, 1.0 mmol) in high yields using $Al_2(SO_4)_3.18H_2O$ as a catalyst at 80 °C in solvent-free conditions (Scheme 3).



Scheme 3. Synthesis of 14-aryl-14*H*-dibenzo[*α*,*j*]xanthenes.

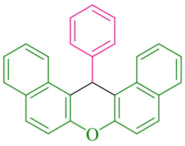
Table 5. Optimization of the reaction condition for the synthesis of **8a** [a]



Entry	$Al_2(SO_4)_3.18H_2O$ (mol %)	Temperature (°C)	Time (min)	Isolated Yields (%)
1	Catalyst free	80	240	trace
2	5	80	25	61
3	**10**	**80**	**15**	**89**
4	10	rt	240	Trace
5	10	40	55	43
6	10	60	30	68
7	10	70	20	72
8	10	90	15	88
9	15	80	15	91

[a]Reaction conditions: *β*-naphthol (2.0 mmol); benzaldehyde (1.0 mmol) and $Al_2(SO_4)_3.18H_2O$ was heated under various temperatures for the appropriate time.

Table 6. $Al_2(SO_4)_3.18H_2O$ catalyzed synthesis of 14-aryl-14*H*-dibenzo[*α*,*j*]xanthenes.



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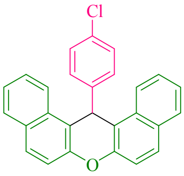

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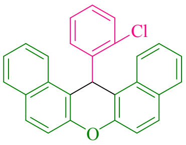

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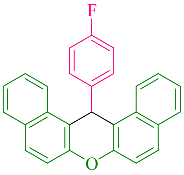

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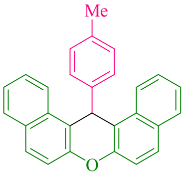

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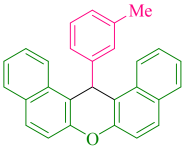

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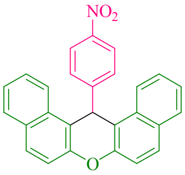

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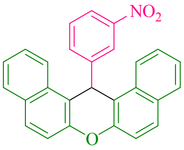

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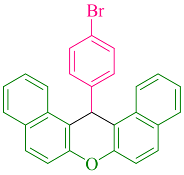

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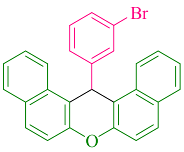

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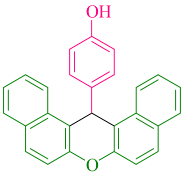

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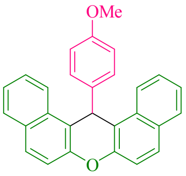

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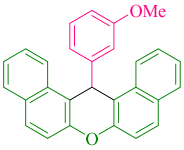
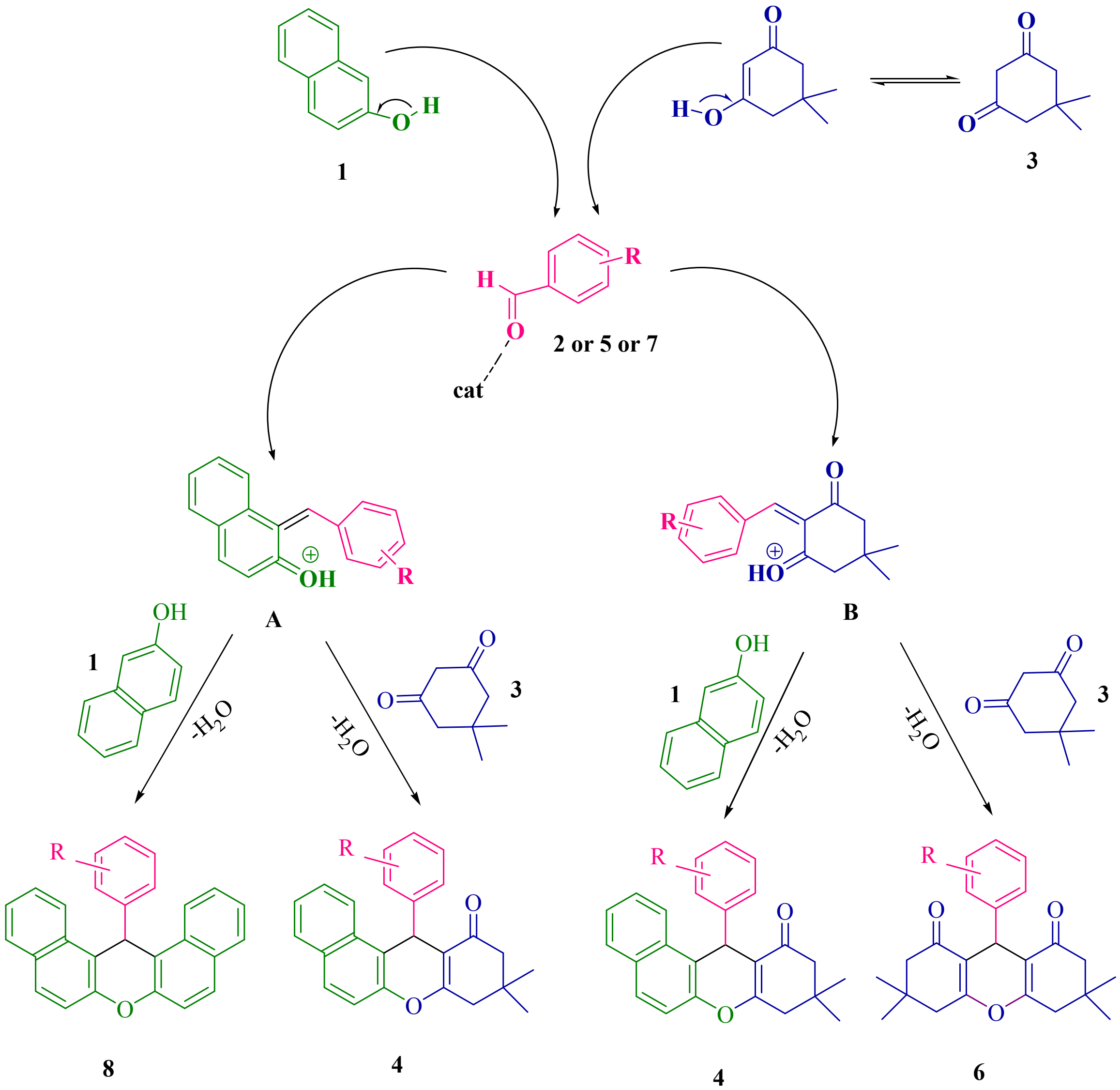


8m

Entry	R^3	Product	Time (min)	Isolated Yields (%)	m.p. °C	m.p. °C
1	C_6H_5	**8a**	15	89	184-186	183-184 [37]
2	4-Cl-C_6H_4	**8b**	20	83	290-292	289-290 [37]
3	2-Cl-C_6H_4	**8c**	20	85	212-214	212-213 [37]
4	4-F- C_6H_4	**8d**	10	92	238-240	240-242 [30]
5	4-Me-C_6H_4	**8e**	10	90	225-227	227-228 [24]
6	3-Me-C_6H_4	**8f**	10	91	198-200	197-198 [36]
7	4-O_2N-C_6H_4	**8g**	15	89	307-309	308-309 [36]
8	3-O_2N-C_6H_4	**8h**	15	91	213-215	212-213 [36]
9	4-Br-C_6H_4	**8i**	20	83	295-297	297-298 [24]
10	3-Br-C_6H_4	**8j**	20	86	190-192	191-193 [20]
11	4-OH-C_6H_4	**8k**	20	81	137-139	138-140 [36]
12	4-OMe-C_6H_4	**8l**	15	85	202-204	204-205 [24]
13	3-OMe-C_6H_4	**8m**	15	87	173-175	174-176 [34]

Proposed mechanistic route for the synthesis of 12-aryl-tetrahydrobenzo[*α*]xanthene-11-ones (**4**), 1,8-dioxo-octahydroxanthenes (**6**) and 14-aryl-14*H*-dibenzo[*α*,*j*]xanthenes (**8**) in the presence of $Al_2(SO_4)_3.18H_2O$ are shown in scheme 4. On the basis of this mechanism, $Al_2(SO_4)_3.18H_2O$ activates aldehyde. Then, nucleophilic *β*-naphthol (**1**) or dimedone (**3**) attacks the carbonyl group of the activated aldehyde and by removing H_2O, the knoevenagel products (**A** or **B**) are generated. The following addition of these intermediates to **1** or **3**, gives the acyclic adduct intermediate, which undergoes an intramolecular cyclization with the participation of two hydroxyl groups to afford the xanthene derivatives (Scheme 4).



Scheme 4. Proposed mechanistic route for the synthesis of 12-aryl-tetrahydrobenzo[*α*]xanthene-11- (**4**), 1,8-dioxo-octahydroxanthenes (**6**) and 14-aryl-14*H*-dibenzo[*α,j*]xanthenes (**8**).

Comparison of catalytic ability some of catalysts reported in the literature for synthesis of 12-aryl-tetrahydrobenzo[*α*]xanthene-11-one (**4**), 1,8-dioxo-octahydroxanthenes (**6**) and 14-aryl-14*H*-dibenzo[*α,j*]xanthenes (**8**) are shown in Table 7- 9.

Table 7. Comparison of catalytic ability some of catalysts reported in the literature for synthesis of 12-aryl-tetrahydrobenzo[*α*]xanthene-11-ones [a]

Entry	Catalyst	Conditions	Time/Yield (%)	References
1	Fe_3O_4@SiO_2-SO_3H	Solvent-free, 110 °C	30 min/95	[20]
2	$NaHSO_4/SiO_2$	CH_2Cl_2, Reflux	300 min/91	[21]
3	NO_2−FePc/C	EtOH, Reflux	30 min/91%	[22]
4	DSIMHS	Solvent-free, 55 °C	20 min/93	[24]
5	CAN	Microwave irradiation, 120 °C	120 min/85	[25]
6	$Sr(OTf)_2$	1,2-Dichloroethane, 80 °C	300 min/85	[28]
7	$Al_2(SO_4)_3.18H_2O$	Solvent-free, 80 °C	15 min/88	This work

[a]Based on the three-component reaction of *β*-naphthol (1.0 mmol); benzaldehyde (1.0 mmol) and dimedone (1.0 mmol).

Table 8. Comparison of catalytic ability some of catalysts reported in the literature for synthesis of 1,8-dioxo-octahydroxanthenes [a]

Entry	Catalyst	Conditions	Time/Yield (%)	References
1	Fe_3O_4@SiO_2-SO_3H	Solvent-free, 110 °C	4 min/94	[20]
2	DSIMHS	Solvent-free, 55 °C	4 min/95	[24]
3	[cmmim][BF_4]	Microwave irradiation	2 min/92	[29]
4	[Hbim]BF_4	Microwave irradiation	45 min/85	[31]
5	[BMim][BF_4]	$Mg(BF_4)_2$, 80 °C	30 min/97	[32]
6	$Al_2(SO_4)_3.18H_2O$	Solvent-free, 80 °C	10 min/92	This work

[a] Based on the three-component reaction of dimedone (2.0 mmol) and benzaldehyde (1.0 mmol).

Table 9. Comparison of catalytic ability some of catalysts reported in the literature for synthesis of 14-aryl-14*H*-dibenzo[*α,j*]xanthenes [a]

Entry	Catalyst	Conditions	Time/Yield (%)	References
1	Fe_3O_4@SiO_2-SO_3H	Solvent-free, 110 °C	30 min/94	[20]
2	DSIMHS	Solvent-free, 90 °C	3 min/94	[24]
3	[BMim][BF_4]	$Mg(BF_4)_2$, 80 °C	15 min/95	[32]
4	SFP	Solvent-free, 90 °C	30 min/98	[33]
5	SiO_2-Pr-SO_3H	Solvent-free, 125 °C	20 min/98	[34]
6	[H-NMP][HSO_4]	Solvent-free, 110 °C	12 min/94	[36]
7	Diatomite-SO_3H	Solvent-free, 90 °C	10 min/93	[37]
8	$Al_2(SO_4)_3.18H_2O$	Solvent-free, 80 °C	15 min/89	This work

[a] Based on the three-component reaction of *β*-naphthol (2.0 mmol) and benzaldehyde (1.0 mmol).

Conclusion

In conclusion, a facile, efficient and eco-friendly procedure for the one-pot, clean synthesis of 12-aryl-tetrahydrobenzo[*α*]xanthene-11-ones, 1,8-dioxo-octahydroxanthenes and 14-aryl-14*H*-dibenzo[*α,j*]xanthenes by using of aluminum sulfate octadecahydrate ($Al_2(SO_4)_3.18H_2O$) as an environmental friendly and readily catalyst is reported. The present procedure provides economic and simple methodology for the synthesis of these heterocyclic compounds and offers several notable advantages over the exiting methods, including efficient, low-cost, available and non-toxic catalyst, solvent-free, environmental friendly, short reaction times, high to excellent yields and simple work-up.

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