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An efficient synthesis of 12-aryl-8,9,10,12-tetrahydrobenzo[*a*]xanthen-11-ones using (4-sulfobutyl)tris(4-sulfophenyl)phosphonium hydrogen sulphate as catalyst under neat conditions

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ABSTRACT

Multicomponent one-pot synthesis of 12-aryl-8,9,10,12-tetrahydrobenzo[a]xanthen-11-ones were achieved by the condensation of β-naphthol, aromatic aldehydes and cyclic 1,3-dicarbonyl compounds under neat conditions employing an efficient and eco-friendly Brønsted acidic ionic liquid (4-sulfobutyl)tris(4-sulfophenyl)phosphonium hydrogen sulphate as catalyst. The reaction work-up is very simple and catalyst can be easily separated from the reaction mixture and reused at least five additional times without any noticeable decrease in catalytic activity.

Key Words: 12-aryl-8,9,10,12-tetrahydrobenzo[*a*]xanthen-11-one; Cyclic 1,3-dicarbonyl compounds; Neat conditions; (4-sulfobutyl)tris(4-sulfophenyl)phosphonium hydrogen sulphate.

INTRODUCTION

The development of environmentally benign, efficient and economical methods for the synthesis of biologically interesting compounds remains a significant challenge in synthetic Chemistry. Green Chemistry emphasizes the need for environmentally clean synthesis, which involves improvement in selectivity, high atom efficiency, elimination of hazardous reagents and easy separation with recovery and reuse of reagents. As a result, volatile organic solvents are being replaced by non-toxic, non-volatile media such as ionic liquids [1], polyethylene glycol and water. Alternatively, the reactions are carried out under solvent-free conditions [2-5].

Xanthenes and benzoxanthenes have drawn much attention in the field of medicinal Chemistry. These oxygen heterocycles have been reported to exert various biological and therapeutic properties such as anti-proliferative [6], antineoplastic [7], analgesic and anti-inflammatory activities [8], as well as their use in photodynamic therapy [9]. These are being utilized as antagonists for paralyzing action of zoxazolamine [10]. Besides, these heterocyclic molecules have been widely used as dyes [11], pH-sensitive fluorescent materials for visualization of biomoleculs [12] and also found in laser technologies [13]. Thus a broad utility range has made xanthenes prime synthetic candidates there by accentuating the need to develop newer synthetic routes for scaffold manipulation of xanthene derivatives. Various methods have been reported for the synthesis of 12-aryl-8,9,10,12-tetrahydrobenzo[*a*]xanthen-11-ones using different catalysts such as Strontium triflate [14], $NaHSO_4$-SiO_2 under reflux in halogenated solvents [15], p-Toluenesulfonic acid [16], Cu/SiO_2 [17], Ruthenium chloride [18], $InCl_3$ / P_2O_5 [19], Phenylboronic acid [20] and Caro's acid-silica gel [21]. However these methods are suffering from one or severe drawbacks, such as long reaction time, low yield, tedious workup, use of toxic solvents and expensive reagents. Therefore, to overcome these disadvantages, we developed a simple, efficient and environmentally benign methodology using Brønsted acidic ionic liquid (4-sulfobutyl)tris(4-sulfophenyl) phosphonium hydrogen sulphate as catalyst under neat conditions.

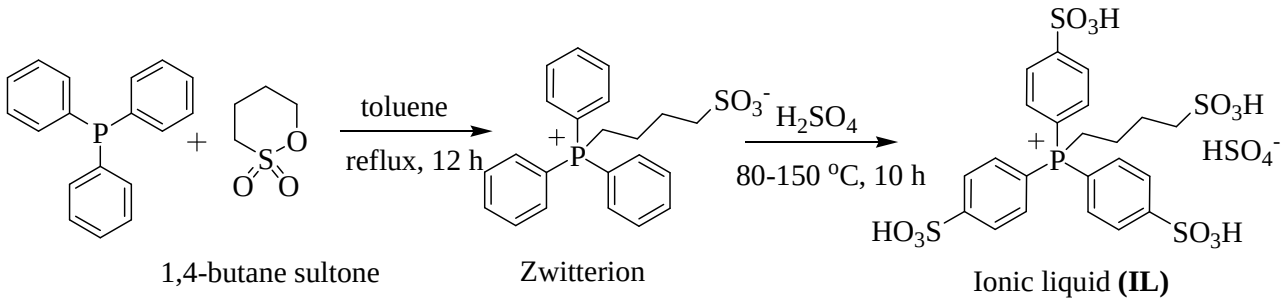
EXPERIMENTAL SECTION

The melting points were determined in open capillaries using Stuart SMP-30 instrument and are uncorrected. The progress of the reaction was monitored by TLC. IR spectra were recorded on Thermo Nicolet Nexus 670 spectrometer using KBr pellet, values are expressed in cm^{-1}. ^1H NMR spectra were recorded on Brucker 300 MHz spectrometer using TMS as internal standard and Chemical shifts are expressed in ppm. The C, H and N analysis of the compounds were done on a Carlo Erba modal EA1108. Mass spectra were recorded on a Jeol JMSD-300 spectrometer.

Preparation of (4-sulfobutyl)tris(4-sulfophenyl)phosphonium hydrogen sulphate:
The Brønsted acidic ionic liquid (4-sulfobutyl)tris(4-sulfophenyl)phosphonium hydrogen sulphate has prepared according to the literature procedure [22] as given below (Scheme-1) and confirmed by the spectral data.

1. Preparation of zwitterions:
Magnetically stirred a mixture of triphenylphosphine (20 mmol) and 1,4-butane sulfonate (20 mmol) in toluene for 12 h at the refluxed temperature. Then, a white solid zwitterion was formed, washed repeatedly with ether and dried in vacuum (110 °C). The product (zwitterion) was obtained in good yield (> 90%) with high purity as assessed by NMR spectroscopy. ^1H NMR (300 MHz, D_2O): δ (ppm) = 1.20 (s, 2H), 2.34 (s, 2H), 3.80 (s, 2H), 4.12 (s, 2H), 7.43–8.51 (m, 15H); ^{13}C NMR (75 MHz, D_2O): δ (ppm) = 143.90, 134.77, 131.42, 127.46, 37.97, 32.82, 23.74, 19.87.

2. Preparations of (4-sulfobutyl)tris(4-sulfophenyl)phosphonium hydrogen sulfate (IL)
Quadruple molar sulfuric acid (40 mmol) was added to the zwitterions (10 mmol), then the mixture was stirred for 12 h at temperature from 80 to 150 °C to form the ionic liquid. Then, the IL phase was washed repeatedly with toluene and ether to remove non-ionic residues and dried in vacuum (110 °C). A black viscous liquid was formed quantitatively in high purity as assessed by NMR spectroscopy. ^1H NMR (300 MHz, D_2O): δ (ppm) = 1.87 (s, 2H), 1.28–1.39 (m, 4H), 1.80– 1.92 (m, 2H), 3.52 (t, *J*=7.6 Hz, 3H), 7.94 (d, *J*=6.4 Hz, 6H), 7.53 (d, *J*=6.8 Hz, 6H); ^{13}C NMR (75 MHz, D_2O): δ (ppm) = 149.83, 135.42, 128.14, 122.43, 52.05, 26.64, 22.62, 20.63.



Scheme-1. Synthesis of (4-sulfobutyl)tris(4-sulfophenyl)phosphonium hydrogen sulphate (IL)

3. General procedure for the synthesis of 12-aryl-8,9,10,12-tetrahydrobenzo [a]xanthen-11-one derivatives (4 a-l)
To a mixture of β-naphthol (1 mmol), aromatic aldehyde (1 mmol) and dimedone /cyclohexane-1,3-dione (1.2 mmol); ionic liquid (10 mol%) was added and heated at 80 °C for an appropriate time. After completion of the reaction indicated by TLC, 5 ml of water was added and stirred at room temperature for about 5 min. The solid obtained was filtered, washed with water, dried and purified by recrystallization with ethanol. The recovered catalyst was reused for subsequent reactions.

RESULTS AND DISCUSSION

In continuation of our ongoing research on the development of more efficient and environmentally friendly procedures for the synthesis of xanthenes [23], herein we wish to describe a new and convenient protocol for the synthesis of 12-aryl-8,9,10,12-tetra hydrobenzo[*a*]xanthen-11-ones via one-pot three component condensation of β-naphthol, aromatic aldehydes and cyclic 1,3-dicarbonyl compounds in the presence of ionic liquid (IL) as catalyst.

In order to set the optimum reaction conditions in terms of solvent and temperature, a model reaction was performed with β-naphthol (1 mmol), benzaldehyde (1 mmol) and dimedone (1.2 mmol) using ionic liquid (5 mol%) as catalyst in different solvents (EtOH, AcOH, CH_3CN, H_2O) and in solvent-free conditions at different temperatures. At room

temperature there is no formation of product was observed in any of the above solvents even after 24 hrs, but under solvent-free conditions only trace amount of compound was observed. As the temperature rises to 80 ^{o}C, maximum amount of the product was observed under solvent-free conditions and minimum was observed in water. Further increment of temperature doesn't much effect on the yield of the product. Optimum reaction conditions were obtained when reaction was carried out at 80 °C under solvent-free conditions (Table 1).

Table-1. Effect of solvent and temperature on the yield of 9,9-dimethyl-12-phenyl-8,9,10,12-tetraydrobenzo[*a*]xanthen-11-one.

Entry	Solvent	Temp. (^{o}C)	Time (h)	Yield[a] (%)
1	EtOH	RT	24	-
2	AcOH	RT	24	-
3	CH_3CN	RT	24	-
4	H_2O	RT	24	-
5	Solvent-free	RT	24	Trace
6	EtOH	80	3	58
7	AcOH	80	3	55
8	CH_3CN	80	3	61
9	H_2O	80	3	37
10	Solvent-free	80	1	67
11	EtOH	Reflux	2	60
12	AcOH	Reflux	2	59
13	CH_3CN	Reflux	2	65
14	H_2O	Reflux	2	46
15	Solvent-free	120	1	67

Reaction conditions: *β-naphthol (1 mmol), benzaldehyde (1 mmol), dimedone (1.2 mmol) and ionic liquid (5 mol%).*
[a] Isolated Yields.

In order to test the efficiency of the catalyst, the reaction of β-naphthol (1 mmol), benzaldehyde (1 mmol) and dimedone (1.2 mmol) was carried out at 80 ^{o}C under solvent-free conditions by changing the amount of the ionic liquid. As the amount of the ionic liquid increases the yield of the product increases and observed quantitative yield with 10 mol% in shorter reaction time (30 min). Further increment of ionic liquid quantity does not affect the yield and time of the product (Table-2). After completion of the reaction indicated by TLC, the catalyst was recovered by evaporating the water, washed with ether, dried and reused for subsequent reactions at least five additional times without significant loss in its activity (Table-3).

Table-2. Optimizing the amount of Ionic liquid (IL).

Entry	Amount IL (mol%)	Time (min)	Yield[b] (%)
1	5	60	67
2	7.5	60	84
3	10	30	96
4	12.5	30	96

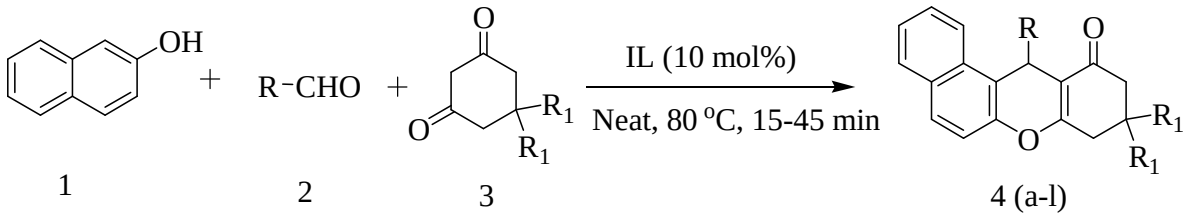
Reaction conditions: *β-naphthol (1 mmol), benzaldehyde (1 mmol) and dimedone (1.2 mmol); solvent-free, 80 ^{o}C.*
[a] Isolated Yields.

Table-3: The effect of reusability of Ionic liquid (IL) on 9,9-dimethyl-12-phenyl-8,9,10,12-tetraydrobenzo[*a*]xanthen-11-one yield

Run	Cycle	Yield[a] (%)
1	0	96
2	1	96
3	2	95
4	3	94
5	4	94
6	5	92

Reaction conditions: *β-naphthol (1 mmol), benzaldehyde (1 mmol), dimedone (1.2 mmol) and SBSANs (0.12 g); Neat 80 ^{o}C; 30 min.*
[a] Yields refer to the pure isolated recovered catalyst.

At these optimistic conditions (amount of IL 10mol%, solvent-free, 80 ^{o}C) various 12-aryl-8,9,10,12-tetrahydrobenzo[*a*]xanthen-11-ones were synthesized with the average of 93% yield in shorter reaction times (Scheme-1, Table-4). All the compounds were confirmed by IR, ^{1}H NMR and Mass spectral analysis and compared with the authentic samples.



1 2 3 4 (a-l)

R_1 = CH_3, H; R = Aryl

Scheme-2. **Synthesis of 12-aryl-8,9,10,12-tetrahydrobenzo[*a*]xanthen-11-ones**

Table-4. Ionic liquid catalyzed solvent-free synthesis of 12-aryl-8,9,10,12-tetrahydrobenzo[*a*] xanthen-11-one derivatives

Product	R	R_1	Time (min)	Yield[a] (%)	Melting Points (^{o}C)	
					Observed	Literature [Ref]
4a	C_6H_5	CH_3	30	96	149-150	151-153 [19]
4b	4-ClC_6H_4	CH_3	15	98	180	180-182 [19]
4c	4-$OCH_3C_6H_4$	CH_3	30	94	203-205	204-205 [19]
4d	4-$CH_3C_6H_4$	CH_3	30	96	175-177	176-178 [19]
4e	3-$NO_2C_6H_4$	CH_3	45	94	168-170	168-170 [19]
4f	4-OHC_6H_4	CH_3	30	90	222-224	223-225 [19]
4g	4-$N(CH_3)_2C_6H_4$	CH_3	30	89	198-200	200-202 [24]
4h	C_6H_5	H	30	90	189-191	189-190 [26]
4i	4-ClC_6H_4	H	20	97	208	206-208 [26]
4j	3-$NO_2C_6H_4$	H	45	92	232-233	235-236 [26]
4k	4-$OCH_3C_6H_4$	H	30	92	179-181	180-182 [26]
4l	4-$CH_3C_6H_4$	H	30	88	202-203	205-206 [27]

Reaction conditions: *β-naphthol (1 mmol), aromatic aldehyde (1 mmol), 1,3-dicarbonyl compounds (1.2 mmol) and ionic liquid (10mol%); Neat 80 ^{o}C; 15-45 min.*

[a] *Isolated Yields.*

A plausible mechanism for the synthesis of 12-aryl-8,9,10,12-tetrahydrobenzo[*a*]xanthen-11-ones catalyzed by ionic liquid has been proposed (Scheme-2). The aldehydic carbonyl oxygen gets activated by the acid part of ionic liquid through intermolecular hydrogen bonding and subsequent condensation with 1,3-dicarbonyl compounds give chalcone, which on reaction with β-naphthol followed by cyclization and dehydration afforded the final product.

Characterisation data:

9,10-Dihydro-9,9-dimethyl-12-phenyl-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4a):

White solid; ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm) = 0.96 (s, 3H), 1.12 (s, 3H), 2.22-2.32 (m, 2H), 2.57 (s, 2H), 5.67 (s, 1H), 7.15-7.49 (m, 7H), 7.68–7.80 (m, 3H), 7.79 (d, *J* = 8.1 Hz, 1H); IR (KBr, cm^{-1}): 1654 (C=O), 1175 (C-O-C); EIMS, 70ev, *m/z*: 354 (M^{+}); Anal. Calcd. For $C_{25}H_{22}O_2$: C, 84.72; H, 6.26. Found: C, 84.37; H, 6.54.

12-(4-Chlorophenyl)-9,10-dihydro-9,9-dimethyl-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4b):

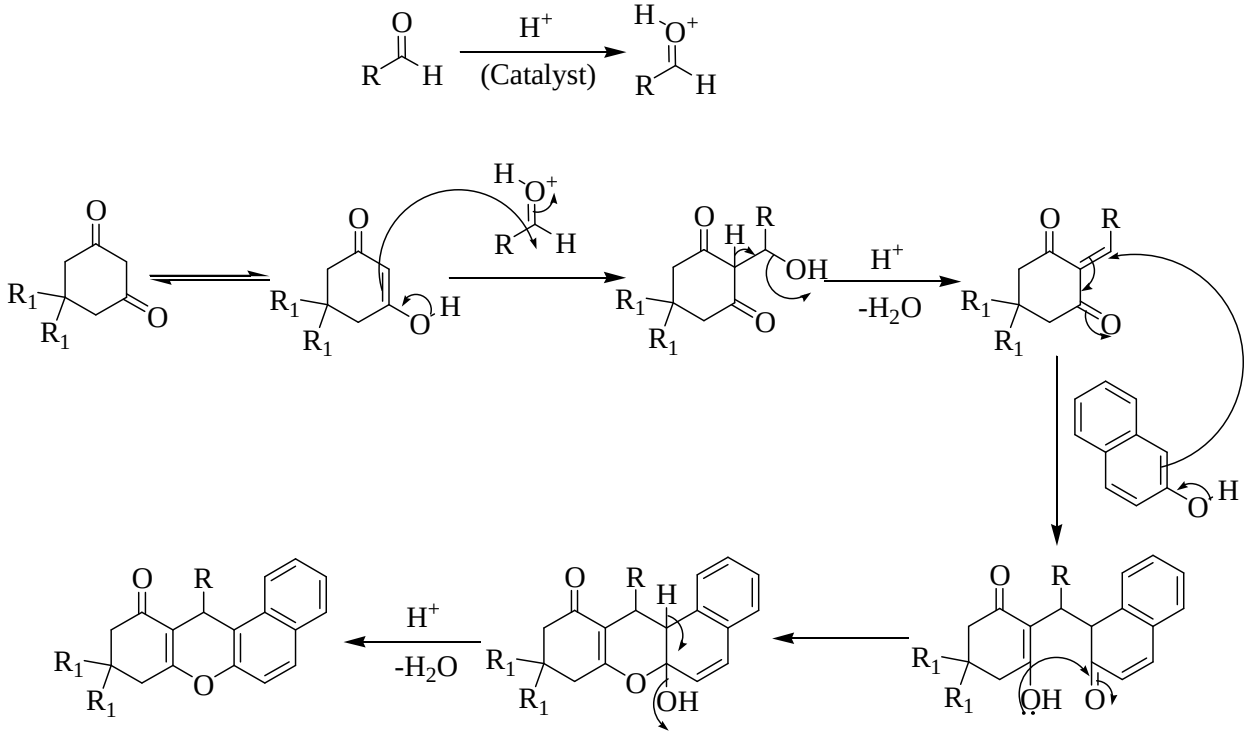
White solid; ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm) = 0.95 (s, 3H), 1.11 (s, 3H), 2.21-2.33 (m, 2H), 2.56 (s, 2H), 5.68 (s, 1H), 7.12-7.45 (m, 7H), 7.76 (d, *J* = 8.4 Hz, 2H), 7.90 (d, *J* = 8.4 Hz, 1H); IR (KBr, cm^{-1}): 1649, 1178; EIMS, 70ev, *m/z*: 388 (M^{+}); Anal. Calcd. For $C_{25}H_{21}ClO_2$: C, 77.21; H, 5.44. Found: C, 77.03; H, 5.57.

9,10-Dihydro-12-(4-methoxyphenyl)-9,9-dimethyl-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4c):

Pale yellow solid; ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm) = 0.97 (s, 3H), 1.14 (s, 3H), 2.20-2.31 (m, 2H), 2.55 (s, 2H), 3.68 (s, 3H), 5.64 (s, 1H), 7.11-7.42 (m, 7H), 7.78 (d, *J* = 8.2 Hz, 2H), 7.91 (d, *J* = 8.2 Hz, 1H); IR (KBr, cm^{-1}): 1662 (C=O), 1194, 1178 (C-O-C); EIMS, 70ev, *m/z*: 384 (M^{+}); Anal. Calcd. For $C_{26}H_{24}O_3$: C, 81.22; H, 6.29. Found: C, 81.58; H, 6.42.

9,10-Dihydro-9,9-dimethyl-12-*p*-tolyl-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4d):
White solid, ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm) = 0.97 (s, 3H), 1.11 (s, 3H), 2.19 (s, 3H), 2.22-2.32 (m, 2H), 2.56 (s, 2H), 5.67 (s, 1H), 6.96-7.45 (m, 7H), 7.73-7.78 (t, *J* = 8.4 Hz, 2H), 8.01 (d, *J* = 8.4 Hz, 1H); IR (KBr, cm^{-1}): 1657, 1185; EIMS, 70ev, *m/z*: 368 (M^+); Anal. Calcd. For $C_{26}H_{24}O_2$: C, 84.75; H, 6.57. Found: C, 84.53; H, 6. 62.

9,10-Dihydro-9,9-dimethyl-12-(3-nitrophenyl)-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4e):
White solid; ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm) = 0.99 (s, 3H), 1.15 (s, 3H), 2.24-2.30 (m, 2H), 2.58 (s, 2H), 5.68 (s, 1H), 6.92-7.21 (m, 2H), 7.34–7.46 (m, 5H), 7.70-7.81 (m, 2H), 7.92 (d, *J*= 8.1 Hz, 1H); IR (KBr, cm^{-1}): 1655 (C=O), 1549 (-NO_2), 1178 (C-O-C); EIMS, 70ev, *m/z*: 399 (M^+); Anal. Calcd. For $C_{25}H_{21}NO_4$: C, 75.17; H, 5.30; N, 3.51, Found: C, 75.46; H, 5.62; N, 3.44.



R_1 = CH_3, H and R = Aryl

Scheme-3. Plausible mechanism for the synthesis of 12-aryl-8,9,10,12-tetrahydrobenzo[*a*] xanthen-11-one derivatives.

9,10-Dihydro-12-(4-hydroxyphenyl)-9,9-dimethyl-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4f):
Pale yellow solid; ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm) = 0.99 (s, 3H), 1.16 (s, 3H), 2.32-2.37 (m, 2H), 2.61 (s, 2H), 5.72 (s, 1H), 7.15-7.47 (m, 7H), 7.75 (d, *J* = 8.2 Hz, 2H), 7.91 (d, *J* = 8.2 Hz, 1H), 9.12 (s, 1H); IR (KBr, cm^{-1}): 3430 (OH), 1643 (C=O), 1182 (C-O-C); EIMS, 70ev, *m/z*: 370 (M^+); Anal. Calcd. For $C_{25}H_{22}O_3$: C, 81.06; H, 5.99. Found: C, 81.43; H, 6.21.

12-(4-(Dimethylamino)phenyl)-9,10-dihydro-9,9-dimethyl-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4g):
White solid; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 0.96 (s, 3H), 1.14 (s, 3H), 2.20-2.32 (m, 2H), 2.55 (s, 2H), 3.07 (s, 6H), 5.66 (s, 1H), 7.10-7.41 (m, 7H), 7.66 (d, *J* = 8.4 Hz, 2H), 7.89 (d, *J* = 8.4 Hz, 1H); IR (KBr, cm^{-1}): 1651 (C=O), 1183 (C-O-C); EIMS, 70ev, *m/z*: 397 (M^+); Anal. Calcd. For $C_{27}H_{27}NO_2$: C, 81.58; H, 6.85; N, 3.52. Found: C, 81.26; H, 7.09; N, 3.74.

9,10-Dihydro-12-phenyl-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4h):
White solid, ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm) = 1.89-2.11 (m, 2H), 2.21-2.50 (m, 2H), 2.59-2.75 (m, 2H), 5.69 (s, 1H), 7.01-7.19 (m, 2H), 7.24-7.46 (m, 5H), 7.75-7.80 (m, 3H), 7.89 (d, *J* = 8.1 Hz, 1H); IR (KBr, cm^{-1}): 1662 (C=O), 1159 (C-O-C); EIMS, 70ev, *m/z*: 326 (M^+); Anal. Calcd. For $C_{23}H_{18}O_2$: C, 84.64; H, 5.56. Found: C, 84.88; H, 5.73.

12-(4-Chlorophenyl)-9,10-dihydro-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4i):
White solid, ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm) = 1.90-2.13 (m, 2H), 2.24-2.53 (m, 2H), 2.60-2.76 (m, 2H), 5.70 (s, 1H), 6.91-7.14 (m, 2H), 7.21-7.44 (m, 5H), 7.79-7.84 (m, 2H), 7.91 (d, *J* = 8.2 Hz, 1H); IR (KBr, cm^{-1}): 1655 (C=O), 1185 (C-O-C); EIMS, 70ev, *m/z*: 360 (M^+); Anal. Calcd. For $C_{23}H_{17}ClO_2$: C, 76.56; H, 4.75. Found: C, 76.32; H, 4.98.

9,10-Dihydro-12-(3-nitrophenyl)-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4j):
White solid, ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm) = 1.90-2.12 (m, 2H), 2.22-2.52 (m, 2H), 2.60-2.77 (m, 2H), 5.71 (s, 1H), 6.90-7.17 (m, 2H), 7.23-7.46 (m, 5H), 7.76-7.79 (m, 2H), 7.89 (d, *J* = 8.1 Hz, 1H); IR (KBr, cm^{-1}): 1649 (C=O), 1594 (-NO_2), 1189 (C-O-C); EIMS, 70ev, *m/z*: 371 (M^+); Anal. Calcd. For $C_{23}H_{17}NO_4$: C, 74.38; H, 4.61; N, 3.77, Found: C, 74.71; H, 4.84; N, 4.02.

9,10-Dihydro-12-(4-methoxyphenyl)-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4k):
White solid, ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm) = 1.91-2.08 (m, 2H), 2.31-2.48 (m, 2H), 2.57-2.76 (m, 2H), 3.68 (s, 3H), 5.69 (s, 1H), 6.70 (d, *J* = 8.0 Hz, 2H), 7.20-7.44 (m, 5H), 7.77 (d, *J* = 8.4 Hz, 2H), 7.95 (d, *J* = 8.4 Hz, 1H); IR (KBr, cm^{-1}): 1651, 1188; EIMS, 70ev, *m/z*: 356 (M^+); Anal. Calcd. For $C_{24}H_{20}O_3$: C, 80.88; H, 5.66. Found: C, 80.64; H, 5.51.

9,10-Dihydro-12-*p*-tolyl-8*H*-benzo[*a*]xanthen-11(12*H*)-one (4l):
White solid, ^{1}H NMR (300 MHz, $CDCl_3$): δ (ppm) = 1.92-2.12 (m, 2H), 2.21 (s, 3H), 2.23-2.52 (m, 2H), 2.61-2.76 (m, 2H), 5.70 (s, 1H), 6.90-7.18 (m, 2H), 7.23-7.47 (m, 5H), 7.76-7.78 (m, 2H), 7.90 (d, *J* = 8.1 Hz, 1H); IR (KBr, cm^{-1}): 1661 (C=O), 1187 (C-O-C); EIMS, 70ev, *m/z*: 340 (M^+); Anal. Calcd. For $C_{24}H_{20}O_2$: C, 84.68; H, 5.92. Found: C, 84.93; H, 6.27.

CONCLUSION

In conclusion, we have developed a convenient, highly efficient and environmentally benign synthetic procedure for the synthesis of 12-aryl-8,9,10,12-tetrahydrobenzo[*a*]xanthen-11-ones employing a novel acidic ionic liquid (4-sulfobutyl)tris(4-sulfophenyl)phosphonium hydrogen sulphate as catalyst under solvent-free conditions in excellent yields. Straight forward workup, reusability of the catalyst and green conditions are the most obvious advantages of this methodology. Therefore, we believe that our procedure is more suitable for the synthesis of 12-aryl-8,9,10,12-tetrahydrobenzo[*a*] xanthen-11-ones than the literature methods.

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