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# Superoxide Mediated New, Convenient Multicomponent Synthesis of Xanthene and 4H-benzochromene Derivatives Using Tetraethylammonium Bromide as Phase Transfer Catalyst

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**ABSTRACT**: A new, convenient one-pot multicomponent synthesis of xantheneand 4*H*-benzochromene derivatives has been achieved in the presence of superoxide ion using tetraethylammonium bromide as phase transfer catalyst. The three-component reaction of various aromatic aldehydes,  $\beta$ -naphthol and dimedone / malononitrile yielded Xanthene and 4*H*-benzochromenederivatives under the mild reaction conditions at room temperature. The present report demonstrates the applicability of tetraethylammonium bromide as a phase transfer catalyst for efficient superoxide studies.

KEYWORDS: Xanthene, heterocyclic compounds, multicomponent synthesis, PTC, superoxide ion, TEAB.

### I. INTRODUCTION

Multicomponent reactions (MCRs) by their convergence, productivity, high yields and ease of execution have recently recognised as a worthless synthetic route in current organic synthesis[1].MCRs usually benefit from other aspects such as atom economy, the use of readily available starting materials, resource effectiveness and bond-forming efficiency, which renders these reactions green and environmentally friendly, given greener perspective[2-4].Also, because MCRs are one-pot processes with milder experimental conditions that do not require the isolation of intermediates, they are perfect candidates for combinatorial, automated synthesis and drug discovery. The chemistry of superoxide ion has come to the forefront of current interdisciplinary research owing to its demonstrated biochemical implications and as a species of relatively unexplored chemical reactivity[5, 6]. From the standpoint of synthetic considerations, the ability of superoxide ion to function as a multipotent reagent has made the chemistry of superoxide ion, an important aspect involving the use of superoxideradical ion in multicomponent organic synthesis remains untouched and warrants its study in thesynthesis of biologically active unit'sdirection [10-15].

Xanthene and 4*H*-benzochromeneits derivatives are a centralclass of oxygen-containing heterocyclic compounds that are known to partake important biological and pharmacological activities such as anti-inflammatory[16-18]anti-bacterial[19] anti-viral[20] and also provide efficacy in photodynamic therapy[21, 22]. They are also used as pH sensitive fluorescent materials[23, 24]dyes and also in laser technologies[25, 26]. Therefore, the syntheses of xanthene derivatives due to a broad range of applications have always been a fascinating field in organic synthesis. Three component reactions of aldehydes,  $\beta$ -naphthol and dimedone/malononitrile compoundsareone of the most attractive



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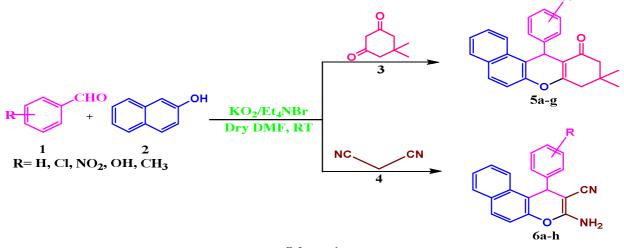
methodologies for the synthesis of xanthene and 4H-benzochromene[27-33]. A few catalysts have been used to stimulate this transformation, as well as Co/Mn supported on nanoZnO[34], TiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>3</sub>O<sub>4</sub> nanoparticles[35],KHSO<sub>4</sub>[36] PEG-400[37], Fe<sub>3</sub>O<sub>4</sub>[38] titanium dodecyl amino phosphate[39], trityl Chloride[40], imidazole and isoquinoline[41], H<sub>4</sub>SiW<sub>12</sub>O<sub>40</sub>[42], Ce(SO<sub>4</sub>)<sub>2</sub>.4H<sub>2</sub>O[43], ionic liquid DDPA[HSO<sub>4</sub>][44], [Pyridinesulfuric SO<sub>3</sub>H]Cl[45],silica acid(SSA)[46], dodecylbenzenesulfonicacid[47], TTAB[48], guanidinehydrochloride[49], Fe<sub>3</sub>O<sub>4</sub>@MCM-41-SO<sub>3</sub>H@[HMIm][HSO<sub>4</sub>][50], ammonium oxalate[51], grinding and CSA[52], nano-TiCl<sub>4</sub>/SiO<sub>2</sub>[53], InCl<sub>3</sub> and P<sub>2</sub>O<sub>5</sub>[54], *p*-toluene sulfonic acid (*p*-TSA)/ liquid[55], silicasulfamicacid[56], NaHSO<sub>4</sub>-SiO<sub>2</sub>under reflux halogenated ionic solvents[57], Cu/SiO<sub>2</sub>[58] ruthenium chloride[59], iodine[60], phenyl boronic acid[61],K<sub>5</sub>CoW<sub>12</sub>O<sub>40</sub>•3H<sub>2</sub>O[62],LiBr[63], KAl(SO<sub>4</sub>)<sub>2</sub>• 12H<sub>2</sub>O[64] and Yb(OTf)<sub>3</sub>[65]. Respectively of these methods has its own merit, but some of these methods are plagued by poor yields, difficult workup, and effluent pollution. Thus there is a need for further enlargement in the synthesis of xanthene derivative. To investigating the behavior of the superoxide ion in multicomponent organic synthesis, which is of significance in itself and further to assess its synthetic scope, the reaction of this novel reagent was studied.

#### **II. MATERIALS AND METHODS**

All chemicals were obtained from Aldrich, the USA, and E. Merck, Germany and used without further purification. TLC was carried out on the SiO<sub>2</sub> gel (HF254, 200 mesh). The solvent system employed was ethyl acetate: n-hexane (2: 1) and the spots were identified by Iodine chamber. IR spectra were taken on a PerkinElmer FT/IR version10.03.05 spectrometer. NMR spectra were run on a Bruker Advance 500 MHz FT-NMR at 500 MHz spectrometers; chemical shifts are given in  $\delta$ ppm, relative to TMS as an internal standard. An open capillaries were used for measuring melting points and are uncorrected.

### **III.RESULTS AND DISCUSSION**

In continuation of our ongoing programof superoxide research, it is our current endeavor to extend the applicability of  $Et_4NO_2$  for the one-pot synthesis of multi-substituted xanthene and 4*H*-benzochromene derivatives by the reaction of aromatic aldehydes,  $\beta$ -naphthol and dimedone/malononitrile (Scheme 1). Tetraethylammonium superoxide was generated in situ by phase transfer reaction of potassium superoxide and tetraethylammonium bromide in dry DMF at room temperature.







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In order to achieve the optimum yield of the products, the effect of manyparameters such astheeffect of solvent and molar proportion of the reactants were investigated in detail. The multicomponent reaction of benzaldehydes,  $\beta$ -naphthol and dimedone/malononitrile was taken as reference reaction. A perusal of the table clearly indicates that the best result was obtained using benzaldehydes,  $\beta$ -naphthol ,dimedone/malononitrile, KO<sub>2</sub> and Et<sub>4</sub>NBr in the molar proportion 1: 1: 1: 4: 2 at room temperature in dry DMF in 4 h (Entry 5, Table 1).

 Table 1: Effect of molar ratio on the % yield of compound 5a/6a.

Entry		]	Reactants molar ratio		%Yield, 5a/6a	
	Benzaldehy	ydesβ-naphthol	DimedoneKO <sub>2</sub>	Et <sub>4</sub> NBr		
1	1.0	1.0	1.0	1.0	1.0	38
2	1.0	1.0	1.0	1.0	0.5	37
3	1.0	1.0	1.0	1.0	0.25	25
4	1.0	1.0	1.0	2.5	1.25	68
5	1.0	1.0	1.0	4.0	2.00	90
6	1.0	1.0	1.0	6.0	3.00	90

To investigate the effect of solvents, the multicomponent reaction of benzaldehydes,  $\beta$ -naphthol and dimedone/malononitrile, as carried out in different aprotic solvents. The results obtained were clearly indicate that dimethylformamide (DMF) was the best solvent among all investigated solvents in the terms of product yield and the reaction time (Table 2, Entry 5).

Entry	Solvents	Time	%Yield	
1	Dichloromethane	12h	trace	
2	Acetonitrile	8h	69	
3	Tetrahydrofuran	14h	45	
4	Dimethylsulfoxide	20h	trace	
5	Dimethylformamide	<b>4h</b>	90	

Under the optimized set of reaction condition, various aromatic aldehydes **1** were made to undergo smooth coupling with  $\beta$ -naphthol **2** and dimedone/malononitrile**3**&**4** in the presence of tetraethylammonium superoxide(Et<sub>4</sub>NO<sub>2</sub>) at room temperature to afford naphthopyranderivatives in good to excellent yield5a-g/6a-h(Table1).

Based on product isolation and current literature on the chemistry of superoxide ion in aprotic media, the following mechanism may be hypothesized (Scheme 2).In these processes, superoxide ion plays an important role in accelerating the reaction. Superoxide is assumed to initiate the reaction by proton abstraction from the substrate



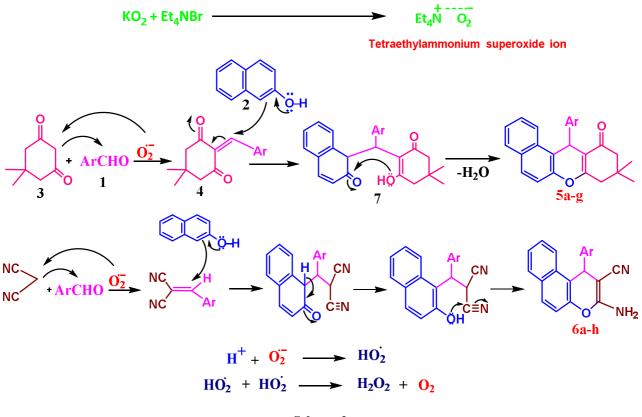
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dimedone followed by anucleophilic attack on aldehyde and subsequently, by dehydration, theolefin4 is formed. The subsequentMichael-type addition of  $\beta$ -naphthol2 to the olefin and produces7, which tautomerizes and cyclized to give the corresponding products 5a-g.

Mechanism of the reaction has described in the Scheme 2.



### Scheme 2

### **IV.EXPERIMENTAL**

### A.General procedure for synthesis of compounds 5a-g/6a-h

Potassium superoxide (1.42 g, 0.02mol) and tetraethylammonium bromide (2.10 g, 0.01mol) were weighed under nitrogen atmosphere using an atmosbag and were transferred into a three-necked R. B. flask, dry DMF (20 mL) was added to it and the mixture was agitated magnetically for 15 min to facilitate the formation of tetraethylammoniumsuperoxide. To the stirred reaction mixture, were added dimedone /malononitrile3/4(0.01 mol). After 10 min, the various aromatic aldehydes 1 (0.01 mol) and  $\beta$ -naphthol 2(0.01mol) were introduced, and the stirring was continued 4-6h. After the reaction was over as indicated by TLC, themixturewas treated with cold brine solution (2 mL) followed by saturated sodium hydrogen carbonate solution (2 mL) to decompose the unreacted KO<sub>2</sub>. The mixture was then extracted with dichloromethane (3× 15 mL), and the combined organic phase was dried over anhydrous Na<sub>2</sub>SO<sub>4</sub>, then filtered, and evaporated to give the products 5a-g/6a-h, which was purified by column chromatography.



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**B.**Characterization data for synthesized compounds of xanthene derivatives:

9,9-Dimethyl-12-phenyl-9,10-dihydro-8H-benzo[a]xanthen-11(12H)-one(**5***a*): mp:150–152<sup>0</sup>C. IR (KBr, cm<sup>-1</sup>): 3053, 2950, 2885, 1650, 1373, 1230, 1176, 1077, 813., <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>):  $\delta$ (ppm) 7.94-7.90 (d, J = 8.4, 1H), 7.72-7.68 (t, J = 7.4, 2H), 7.39-7.24 (m, 5H), 7.12-7.08 (t, J = 7.8, 2H), 7.05-7.03 (t, J = 7.4, 1H), 5.64 (s, 1H), 2.54 (s, 2H), 2.34-2.26 (m, 2H), 1.07 (s, 3H), 0.93(s, 3H), <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>):  $\delta$  (ppm) 196.2, 164.1, 147.4, 145.1, 131.2, 131.0, 129.3, 129.1, 128.4, 128.4, 127.6, 126.5, 125.3, 123.4, 117.5, 117.3, 113.2, 50.1, 40.6, 34.1, 32.1, 29.2, 26.2.

*9,9-dimethyl-12-(3-Chlorophenyl)-9,10-dihydro-8H-benzo[a]xanthen-11(12H)-one(5b)*: mp:178–180<sup>0</sup>C. IR(KBr, cm<sup>-1</sup>): 3073, 2932, 1646, 1371, 1225, 1175, 1030, 748.<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm) 7.90-7.84 (d, J = 8.4, 1H), 7.76-7.73 (m, 2H), 7.43-7.22 (m, 5H), 7.18-7.05 (t, J = 7.8, 1H), 7.01-7.00 (d, J = 8.8, 1H), 5.64 (s, 1H), 2.54(s, 2H), 2.29-2.20(m, 2H), 1.10(s, 3H), 0.95 (s, 3H), <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm) 196.6. 165.0, 148.05, 148.03, 133.3, 131.8, 131.2, 130.8, 130.2, 129.3, 128.7, 128.2, 127.5, 127.3, 125.7, 124.0, 118.0, 117.2, 113.5, 50.8, 41.1, 34.6, 32.5, 29.5, 27.0.

9,9-Dimethyl-12-(3-nitrophenyl)-9,10-dihydro-8H-benzo[a]xanthen-11(12H)-one(5c) : mp:232–234<sup>0</sup>C. IR(KBr, cm<sup>-1</sup>): 3062, 2950, 2889, 1650, 1593, 1372.<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm) 7.93–8.09 (2H, m, Ar.), 7.79–7.84 (4H, m, Ar.), 7.35–7.46 (4H, m, Ar.), 5.84 (1H, s, CH), 2.66–2.82 (2H, m, CH<sub>2</sub>), 2.33–2.45 (2H, m, CH<sub>2</sub>), 1.93–2.12 (2H, m, CH<sub>2</sub>), <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm) 197.0, 147.6, 146.6, 135.7, 134.7, 131.4, 129.0, 128.4, 127.1, 125.0, 123.1, 123.0, 122.3, , 121.4, 117.0, 116.0, 114.2, 36.0, 34.5, 27.3, 20.2.

*9*,9-*Dimethyl-12-(4-nitrophenyl)-9*,10-*dihydro-8H-benzo[a]xanthen-11(12H)-one* (*5d*) : mp:234–236<sup>0</sup>C. IR(KBr, cm<sup>-1</sup>): 3104, 3064, 2944, 2883, 1653, 1591, 1453.<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm)7.78–7.81 (3H, m, Ar.), 7.34–7.50 (5H, m, Ar.), 5.83 (1H, s, CH), 2.63–2.80 (2H, m, CH<sub>2</sub>), 2.33–2.50 (2H, m, CH<sub>2</sub>), 1.91–2.13 (2H, m, CH<sub>2</sub>), 13C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm)196.7. 151.9, 147.5, 143.1, 131.5, 131.0, 129.3, 129.1, 128.3, 127.4, 125.0, 123.3, 123.1, 123.0, 117.1, 116.0, 114.0, 36.7, 34.6, 27.5, 20.1.

*9*,9-*dimethyl*-12-(2-*Hydroxyphenyl*)-9,10-*dihydro*-8*H*-*benzo[a]xanthen*-11(12*H*)-one (**5***e*) : mp:220–222<sup>0</sup>C.IR(KBr,cm<sup>-1</sup>): 3351, 3054, 3022, 2948, 2872, 1651, 1593, 1466, 1369.<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm)9.23(s, 1H), 7.75-7.71 (m, 2H), 7.63-7.61 (d, J = 8, 1H), 7.38-7.27 (m, 3H), 7.00-6.96 (m, 2H), 6.60-6.54 (m, 2H), 5.72 (s, 1H), 2.54 (s, 2H), 2.40-2.30 (m, 2H), 1.10 (s, 3H), 0.97 (s, 3H), <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm) 197.2, 162.0, 153.9, 147.1, 135.9, 131.1, 128.9, 128.3, 128.1, 126.3, 124.4, 123.9, 117.3, 117.0, 115.2, 114.0, 109.0, 50.4, 41.0, 33.3, 29.4, 27.1.

*9,9-dimethyl-12-(3-Hydroxyphenyl)-9,10-dihydro-8H-benzo[a]xanthen-11(12H)-one(5f)* : mp:240–242<sup>0</sup>C.IR(KBr,cm<sup>-1</sup>): 3410, 3052, 2951, 1640, 1589, 1373, 1222, 1170, 1023.<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm) 9.23(s, 1H), 7.75-7.71 (m, 2H), 7.63-7.61 (d, J = 8, 1H), 7.38-7.27 (m, 3H), 7.00-6.95 (m, 2H), 6.60-6.54 (m, 2H), 5.72 (s, 1H), 2.52(s, 2H), 2.40-2.30 (m, 2H), 1.13 (s, 3H), 0.93 (s, 3H), <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm) 196.7, 164.5, 157.9, 148.0, 147.0, 131.9, 131.6, 129.8, 129.3, 127.9, 125.8, 124.1, 119.8, 118.0, 115.9, 114.1, 51.0, 41.1, 34.8, 32.7,29.6, 27.1.

*9*,9-*dimethyl*-12-(4-Hydroxyphenyl)-9,10-*dihydro*-8H-benzo[a]xanthen-11(12H)-one (**5g**) : mp:210–212<sup>0</sup>C.IR(KBr,cm<sup>-1</sup>): 3356, 3057, 3024, 2950, 2871, 1650, 1592, 1468, 1371.<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm)7.98–7.67 (3H, m, Ar.), 7.45–7.30 (3H, m, Ar.), 7.20–7.13 (2H, m, Ar.), 6.66–6.60 (2H, m, Ar.), 5.64 (1H, s, CH), 4.81 (1H, s, OH), 2.57 (2H, s, CH<sub>2</sub>), 2.34–2.20 (2H, m, CH<sub>2</sub>), 1.11 (3H, s, CH<sub>3</sub>), 0.97 (3H, s, CH<sub>3</sub>), <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm)197.5, 162.1, 154.0, 147.3, 136.8, 131.3, 129.2, 128.5, 128.1, 126.7, 124.7, 123.5, 117.5, 117.0, 115.0, 114.2, 109.2, 50.8, 41.2, 33.6, 29.0, 27.0.

*3-Amino-1-(phenyl)-1H–benzo[f]chromene-2-carbonitrile* (*6a*): mp: 288-290  $^{\circ}$ C. IR (KBr, cm<sup>-1</sup>): 3448, 3346, 2179, 1641, 1578, 1420. <sup>1</sup>HNMR (500 MHz, CDCl<sub>3</sub>):  $\delta$  (ppm) 7.64 (d, 1H, J = 8.4, Ar), 7.40 (d, 1H, J = 8.4, Ar), 7.60-7.56 (m, 3H, Ar), 7.17-7.12 (m, 5H, Ar), 6.80 (d, 1H, Ar), 6.59 (brs, 2H, NH<sub>2</sub>), 4.69 (s, 1H, CH). <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>):  $\delta$  (ppm) 177.6, 161.6, 140.2, 136.9, 126.4, 124.2, 124.1, 123.3, 123.1, 122.4, 122.3, 122.1, 119.6, 119.3, 118.3, 60.1, 29.1.

*3-Amino-1-(4-chlorophenyl)-1H-benzo[f]chromene-2-carbonitrile (6b)*: mp: 206–208<sup>0</sup>C. IR (KBr, cm<sup>-1</sup>): 3408, 3320, 3029, 2224, 1630, 1585. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm) 7.82 (d, 2H, Ar), 7.74-7.60 (m, 4H, Ar), 7.50 (d, 2H, Ar), 7.43-7.40 (m, 1H, Ar), 7.30-7.27 (m, 1H, Ar), 7.13 (brs, 2H, NH<sub>2</sub>), 5.01 (s, 1H, CH). <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>):δ (ppm) 210.1, 145.4, 141.9, 134.4, 131.2, 131.0, 129.4, 128.2, 127.4, 126.1, 125.3, 124.4, 122.9, 121.2, 118.9, 118.5, 60.2, 40.4.



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3-Amino-1-(3-chlorophenyl)-1H-benzo[f]chromene-2-carbonitrile (**6***c*): mp: 202–204<sup>0</sup>C. IR (KBr, cm<sup>-1</sup>): 3430, 3329, 2180, 1648, 1590. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm) 7.96–7.86 (m, 2H, Ar), 7.45–7.41 (m, 2H, Ar), 7.35–7.18 (m, 5H, Ar), 6.36 (brs, 2H, NH<sub>2</sub>), 5.34 (s, 1H, CH).<sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm) 159.9. 147.5, 145.6, 143.0, 137.1, 134.2, 127.1, 126.5, 126.3, 126.1, 123.9, 123.5, 120.9, 120.8, 119.5, 118.5, 115.0, 111.2, 59.1.

3-Amino-1-(2-chlorophenyl)-1H-benzo[f]chromene-2-carbonitrile (6d): mp: 268–270°C. IR (KBr, cm<sup>-1</sup>): 3342, 3264,

2192, 1636, 1543. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm)7.15-7.12 (1H, d, Ar), 7.76-7.73 (1H, d, Ar), 7.03-7.00 (1H, d, Ar), 7.08 (4H, s, Ar), 7.55-7.44 (3H, m, Ar), 6.53 (brs, 2H, NH<sub>2</sub>), 4.00 (s, 1H, CH). <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm)178.5, 175.4, 151.9, 145.5, 145.3, 132.9, 132.4, 129.1, 128.9, 128.3, 128.1, 127.8, 127.2, 126.5, 119.6, 118.1, 58.9, 17.9.

*3-Amino-1-(3-nitrophenyl)-1H-benzo[f]chromene-2-carbonitrile*(*6e*): mp: 240–242<sup>0</sup>C. IR (KBr, cm<sup>-1</sup>): 3465, 3356, 2193, 1659, 1590. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm) 8.00 (m, 1H, Ar). 7.98 (d, J=6.5 Hz, 1H, Ar), 7.94–7.88 (m, 2H, Ar), 7.84 (d, J= 7.5 Hz, 1H, Ar), 7.63 (m, 1H, Ar), 7.57 (t, J= 7.5 Hz, 1H, Ar), 7.45–7.40 (m, 2H, Ar), 7.36 (d, J= 8.0 Hz, Ar), 7.16 (brs, 2H, NH<sub>2</sub>), 5.02 (s, 1H, CH). <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm) 169.4, 158.4, 148.9, 147.8, 134.5, 131. 9, 130.9, 130.5, 130.1, 129.4, 127.9, 125.1, 123.0, 122.9, 121.1, 120.0, 117.1, 115.1, 57.9.

3-*Amino-1-(4-nitrophenyl)-1H-benzo[f]chromene-2-carbonitrile* (*6f*): mp: 184–186<sup>0</sup>C. IR (KBr, cm<sup>-1</sup>):3576, 3452, 2238, 1659, 1533.<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm)7.98 (d, 1H, Ar), 7.79 (d, 1H, Ar), 7.66-7.60 (m, 3H, Ar), 7.61-7.50 (m, 2H, Ar), 7.37-7.32 (m, 2H, Ar), 7.19 (d, 1H, Ar), 6.96 (brs, 2H, NH<sub>2</sub>), 4.31 (s, 1H, CH). <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm)188.9, 167.2, 154.9, 146.1, 129.2, 128.3, 128.1, 127.9, 127.5, 126.9, 126.7, 125.9, 125.1, 124.4, 122.2, 119.3, 67.1, 34.7.

*3-Amino-1-(3-hydroxyphenyl)-1H-benzo[f]chromene-2-carbonitrile*(*6g*): mp: 280–282<sup>0</sup>C. IR (KBr, cm<sup>-1</sup>): 3419, 3325, 2186, 1642, 1586. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm) 8.38 (brs, 1H). 7.89 (t, J= 8.5 Hz, 3H, Ar), 7.46–7.40 (m, 2H, Ar), 7.31 (d, 1H, Ar), 7.09 (t, J= 8.5 Hz, 1H, Ar), 6.69 (d, 1H, Ar), 6.64–6.60 (m, 2H, Ar), 6.56 (brs, 2H, NH<sub>2</sub>), 4.56 (s, 1H, CH). <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm) 161.1. 159.8, 148.3, 147.1, 132.0, 130.9, 129.9, 129.7, 128.8, 126.8, 124.9, 123.9, 119.1, 118.9, 116.9, 115.1, 114.1, 113.0, 59.9.

*3-Amino-1-(4-methylphenyl)-1H-benzo[f]chromene-2-carbonitrile* (*6h*): mp: 268–270<sup>0</sup>C. IR (KBr, cm<sup>-1</sup>): 3482, 3368, 2190, 1638, 1571. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>): δ (ppm)2.35 (s, 3H, CH<sub>3</sub>),δ 7.81 (d, 2H, Ar) ; 7.75-7.61 (m, 4H, Ar), 7.51 (d, 2H, Ar), 7.41-7.38 (m, 1H, Ar), 7.30- 7.27 (m, 1H, Ar), 7.00 (s, 2H, NH<sub>2</sub>), 4.46 (s, 1H, CH). <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): δ (ppm)187.3, 174.9, 152.9, 144.9, 144.5, 143.1, 134.9, 132.7, 131.9, 129.5, 129.3, 129.0, 128.1, 127.3, 127.1, 59.1, 24.3, 17.9.

#### V. CONCLUSION

A new alternative method for synthesis of some xantheneand 4*H*-benzochromene derivatives from three-component, one-pot condensation of an aldehyde,  $\beta$ -naphthol and dimedone / malononitrileunder the mild reaction conditions at room temperature using tetraethylammonium superoxide is given. The development includes the use oftetraethylammonium bromide as a phase transfer catalyst. The advantages of this method over other existingmethods are higher yields, mild reaction condition, easy purification and economic viability of the catalyst.

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