



## A facile one-pot synthesis of tetrahydrodibenzo[a,i]phenanthridines and novel hexahydrodicyclopenta[b,d]pyridines

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### ABSTRACT

A series of eight 5-aryl/heteryl-7,8,13,14-tetrahydrodibenzo[a,i]phenanthridines, another series of seven novel 5-aryl/heteryl-1,2,3,6,7,8-hexahydrodicyclopenta[b,d]pyridines and two hitherto unknown bridged 2,4-diaryl-3-azabicyclo[3.2.1]octan-8-ones have been synthesised by the condensation of  $\alpha$ -tetralone and various aryl/heteryl aldehydes, cyclopentanone and same aryl/heteryl aldehydes and cyclopentanone and two ortho substituted aryl aldehydes respectively in presence of ammonium acetate under methanol and ethanol media under different conditions. All the compounds have been screened for their antimicrobial and antifungal activity with promising results at the outset.

**Keywords:**  $\alpha$ -tetralone, cyclopentanone, aryl/heteryl aldehydes, dibenzo[a,i]phenanthridines and dicyclopenta[b,d]pyridines.

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### INTRODUCTION

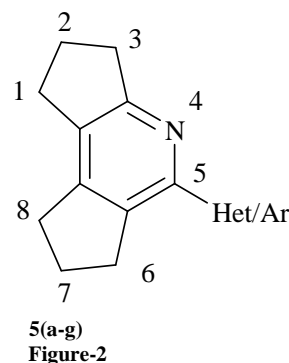
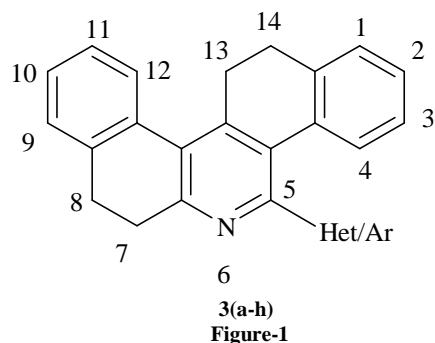
One pot multi component reactions are emerging as a new area of interest for organic chemists as these have tremendous advantages over conventional multistep synthesis as well as their potential applications in medicinal chemistry for the generation of diverse scaffolds [1-2]. As a part of our program to synthesise biologically active heterocyclic compounds using multi component coupling reactions we wish to report here an efficient one pot, three component synthesis of dibenzophenanthridines using aromatic/heteroaromatic aldehydes, cyclic ketones and ammonium acetate.

Phenanthridines are an important class of nitrogen containing tricyclic compounds formed by replacing a carbon atom by a nitrogen atom in the central ring of phenanthrene. Their derivatives exhibit significant synthetic potential and several biological and technological applications [3]. Their wide spread spectrum covers activities such as antitumour [4], antibacterial [5] and antiviral activities [6-7]. They are readily used in the treatment of biological disorders such as trypsinemia [8]. Various drugs such as isometadum, dimidium and holium contain phenanthridine as their pharmacophore [9]. Apart from these, their uses in the physical domain include photoconducting [10] and photovoltaic activities [11]. Their utility lies in the generation of optical materials such in holography, lithographic plates for printing and electric equipments [12]. Their synthesis possesses problems like lack of generality, generation from simple available shelf precursors and often low yields are encountered in their synthesis [13-14]. Thus, only a limited number of strategies and routes are available which as such or in some modified form have been successfully applied in the generation of heteropolycyclic scaffolds. The development of new, rapid and clean synthetic routes, trends and techniques towards focussed library of such compounds is therefore, of great importance to both medicinal and synthetic chemists [15].

Pyridines include compounds used as water repellents, herbicides and various drugs. This ring is in fact, embedded in a large number of compounds including niacin, pyridoxal and nicotine [16]. It is the basic nucleus in a large number of organic compounds and is associated with diverse pharmacological properties such as antimicrobial [17],

anticonvulsant [18], antiviral [19], anti-HIV and antifungal [20]. In the pharmaceutical industry it forms the key nucleus of over more than 700 existing drugs [21]. Describing various pharmacological properties of pyridines will be putting restriction on the large range and sweep of this vital nucleus. Keeping in view the biological importance of these two nuclei we contemplated the synthesis of two categories of condensed polycyclic heterocyclic systems containing embedded in them these two nuclei of phenanthridine and pyridine.

Initially we started with the generation of dibenzophenanthridines and encouraged with the obtention of these partially reduced dibenzo[*a,i*]phenanthridines **3** from  $\alpha$ -tetralone and aryl/heteryl aldehydes in very good yield using this one step synthesis, this method was extended for the generation of partially reduced dicyclopenta[*b,d*]pyridines **5** starting from cyclopentanone and different aryl/heteryl aldehydes. These two novel heterocyclic systems were prepared using **Scheme-1** and **Scheme-2** respectively.



## EXPERIMENTAL SECTION

**General Experimental:** The melting points were determined in open capillary tubes on Perfit melting point apparatus and are uncorrected. The purity of the products was checked on TLC plates coated with BDH silica gel-G. Visualization of spots was effected by exposure to iodine vapours and Draggen dorff reagent. The IR spectra were recorded on Perkin-Elmer FTIR spectrophotometer ( $\nu_{\max}$  in  $\text{cm}^{-1}$ ).  $^1\text{H}$  and  $^{13}\text{C}$  NMR Spectra were recorded on Bruker Ac-400 (400 & 100 MHz respectively). EIMS were recorded on Bruker Micro mass VG-7070 mass spectrometer. Elemental analysis was performed on Leco CHNS 932 analyser.

### Procedure for the Synthesis of 5-phenyl-7,8,13,14-tetrahydrodibenzo[*a,i*]phenanthridine (3a):

2 mmol of  $\alpha$ -tetralone (**1**) was taken in a round bottom flask and was treated with 1mmol of the concerned araldehyde/heteraldehyde (**2a**) and 1.5mmol of ammonium acetate in 25ml in anhydrous ethanol. The mixture was refluxed on water bath for 3 hours when colour changed to yellow. Reaction mixture was kept overnight at room temperature. The progress of reaction was monitored by TLC. The solid, thus, settled was separated and the crude product was purified by running it over silica gel column using petroleum ether and ethyl acetate as eluent (9:1). However, when the reaction was done in ethanol yield was 90% and when done in methanol it was 75%.

The compounds **3b-h** were obtained by a similar procedure.

### Spectral data

#### 5-Phenyl-7,8,13,14-tetrahydrodibenzo[*a,i*]phenanthridine (3a)

Black solid, IR (KBr,  $\nu$   $\text{cm}^{-1}$ ): 3035, 2834, 1599, 1418, 1382, 1235.  $^1\text{H}$  NMR ( $\text{CDCl}_3$ , 400MHz):  $\delta$  2.73-3.18(m, 8H), 7.3-7.89(m, 13H).  $^{13}\text{C}$  NMR ( $\text{CDCl}_3$ , 100MHz):  $\delta$  24.35, 27.39, 29.42, 31.34, 124.58, 125.8, 126.2, 126.8, 127.3, 127.2, 127.2, 128.3, 128.65, 129.3, 129.81, 130.6, 132.78, 132.9, 136.8, 138.5, 139.2, 139.6, 140.2, 142.5, 145.6, 153.4, 157.6. EIMS  $m/z$  = 360 ( $M+1$ ) $^+$ .

#### 5-(4-Methoxyphenyl)-7,8,13,14-tetrahydrodibenzo[*a,i*]phenanthridine (3b)

Orange solid, IR (KBr,  $\nu$   $\text{cm}^{-1}$ ): 3032, 2829, 1603, 1564, 1423, 1354, 1223.  $^1\text{H}$  NMR ( $\text{CDCl}_3$ , 400MHz):  $\delta$  2.56- 2.88 (m, 6H), 3.21(d, 2H), 3.73(s, 3H), 6.87-6.91(m, 4H), 7.28-7.32(m, 8H).  $^{13}\text{C}$  NMR ( $\text{CDCl}_3$ , 100MHz):  $\delta$  23.9, 27.2, 29.6, 30.2, 55.8, 114.5, 114.5, 126.4, 126.4, 127.3, 127.6, 128.1, 128.3, 128.3, 128.59, 128.6, 128.62, 128.9, 128.7, 132.3, 132.42, 136.3, 136.5, 142.3, 148.5, 151.2, 156.4, 159.8. EIMS  $m/z$  = 390 ( $M+1$ ) $^+$ .

**5-(4-Chlorophenyl)-7,8,13,14-tetrahydrodibenzo[a,i]phenanthridine (3c)**

Light grey solid, IR (KBr,  $\nu$   $\text{cm}^{-1}$ ): 3029, 2829, 1605, 1532, 1427, 1351, 1235, 752.  $^1\text{H NMR}$  (DMSO- $d_6$ , 400MHz):  $\delta$  2.53-3.09(m, 8H), 6.79-6.94 (m, 4H), 7.1-7.4 (m, 8H).  $^{13}\text{C NMR}$  (DMSO- $d_6$ , 400MHz):  $\delta$  29.48, 31.32, 129.1, 129.2, 129.4, 129.5, 132.7, 134.5, 124.67, 125.5, 126.1. EIMS  $m/z = 394(\text{M}+1)^+$ .

**5-(3,4-Dimethoxyphenyl)-7,8,13,14-tetrahydrodibenzo[a,i]phenanthridine (3d)**

Brown solid, IR (KBr,  $\nu$   $\text{cm}^{-1}$ ) 3014, 2879, 1598, 1415, 1325, 1217.  $^1\text{H NMR}$  (DMSO- $d_6$ , 400MHz):  $\delta$  2.91-3.12 (m, 8H), 3.71 (s, 6H), 6.8 (d, 1H), 7.73 (m, 10H).  $^{13}\text{C NMR}$  ( $\text{CDCl}_3$ , 100 MHz):  $\delta$  25.6, 25.8, 26.7, 27.1, 27.4, 35.2, 56.1, 56.1, 112.7, 115.7, 120.8, 129.1, 135.3, 135.7, 147.9, 150.8, 155.3, 155.7, 163.9. EIMS  $m/z = 420 (\text{M}+1)^+$ .

**5-(3,4-Methylenedioxyphenyl)-7,8,13,14-tetrahydrodibenzo[a,i]phenanthridine (3e)**

Grey solid, IR (KBr,  $\nu$   $\text{cm}^{-1}$ ): 3014, 1648, 1236, 1579, 1469.  $^1\text{H NMR}$  (DMSO- $d_6$ , 400MHz)  $\delta$ : 2.79 (s, 6H), 3.32(d, 2H), 5.82(s, 2H), 6.75(d,1H), 7.1-7.6 (m,10H).  $^{13}\text{C NMR}$  (DMSO- $d_6$ , 100MHz) $\delta$ : 25.4, 25.9, 26.7, 27.3, 27.7, 101.6, 112.3, 115.7, 120.6, 129.5, 134.1, 135.6, 147.2, 147.9, 149.2, 151.4, 155.6, 164.3. EIMS  $m/z = 403 (\text{M}+1)^+$ .

**5-(2-Methoxyphenyl)-7,8,13,14-tetrahydrodibenzo[a,i]phenanthridine (3f)**

Light brown solid, IR (KBr,  $\nu$   $\text{cm}^{-1}$ ): 3035, 1565, 1421, 1347, 1218.  $^1\text{H NMR}$  (DMSO- $d_6$ , 400 MHz)  $\delta$ : 2.72-2.78 (m, 6H), 3.11 (t, 2H), 3.73 (s, 3H), 6.7 (m, 2H), 7.34-7.77 (m,10H).  $^{13}\text{C NMR}$  (DMSO- $d_6$ , 100MHz):  $\delta$  23.8, 27.8, 29.6, 30.1, 56.4, 114.7, 121.4, 122.1, 126.5, 127.1, 127.2, 127.3, 128.3, 128.4, 128.4, 128.6, 128.7, 132.2, 132.3, 136.4, 142.2, 148.4, 151.4, 156.7, 157.5, 159.1. EIMS  $m/z = 390 (\text{M}+1)^+$ .

**5-(2-Furyl)-7,8,13,14-tetrahydrodibenzo[a,i]phenanthridine (3g)**

Viscous Black solid, IR (KBr,  $\nu$   $\text{cm}^{-1}$ ) 3047, 2872, 1618, 1575, 1345, 1335, 1205.  $^1\text{H NMR}$  (DMSO- $d_6$ , 400MHz)  $\delta$ : 2.82-2.89 (m, 8H), 6.2 (m, 2H), 7.5d (1H), 7.6-7.9 (m, 8H).  $^{13}\text{C NMR}$  (DMSO- $d_6$ , 100MHz):  $\delta$  24.4, 24.9, 25.8, 26.8, 27.1, 34.8, 105.2, 107.1, 134.3, 135.6, 142.8, 151.4, 155.3, 157.6, 164.3. EIMS  $m/z = 350 (\text{M}+1)^+$ .

**5-(3-Indolyl)-7,8,13,14-tetrahydrodibenzo[a,i]phenanthridine (3h)**

Grey-black solid, IR (KBr,  $\nu$   $\text{cm}^{-1}$ ): 3049, 1674, 1572, 2873, 3492.  $^1\text{H NMR}$  (DMSO- $d_6$ , 400MHz): 2.87 (t, 6H), 3.21 (t, 2H), 7.14-7.17 (m, 9H), 7.43 (d, 4H), 10.23(s, 1H, NH).  $^{13}\text{C NMR}$  (DMSO- $d_6$ , 100MHz)  $\delta$ : 25.4, 25.8, 26.1, 27.8, 27.9, 114.7, 114.7, 128.4, 134.2, 135.7. EIMS  $m/z = 399(\text{M}+1)^+$ .

TABLE-1 Physical and the analytical data of the compounds (3a-h)

Product 3(a-h)	Ar/Het. 2(a-h)	M.Pt ( $^{\circ}\text{C}$ )	Yield (%)	Molecular formula	Analysis %		
					Calcd./Found		
					C	H	N
3a	Phenyl	172	75	$\text{C}_{27}\text{H}_{21}\text{N}$	90.21 90.18	5.89 5.88	3.90 3.88
3b	4-Methoxyphenyl	175	85	$\text{C}_{28}\text{H}_{23}\text{NO}$	86.34 86.28	5.95 5.93	3.60 3.59
3c	4-Chlorophenyl	178	70	$\text{C}_{27}\text{H}_{20}\text{ClN}$	82.33 82.01	5.12 5.07	3.56 3.44
3d	3,4-Dimethoxyphenyl	190	80	$\text{C}_{29}\text{H}_{25}\text{NO}_2$	83.03 82.27	6.01 5.87	3.34 3.28
3e	3,4-Methylenedioxyphenyl	185	78	$\text{C}_{28}\text{H}_{21}\text{NO}_2$	83.35 83.21	5.25 5.14	3.47 2.76
3f	2-Methoxyphenyl	180	72	$\text{C}_{28}\text{H}_{23}\text{NO}$	86.34 86.23	5.95 5.87	3.60 3.52
3g	2-Furyl	167	60	$\text{C}_{25}\text{H}_{19}\text{NO}$	85.93 85.91	5.48 5.42	4.01 3.98
3h	3-Indolyl	169	65	$\text{C}_{29}\text{H}_{22}\text{N}_2$	87.41 87.20	5.56 5.43	7.03 6.67

**Procedure and Spectral data**

**Procedure for the synthesis of 5-phenyl-1,2,3,6,7,8-hexahydrodicyclopenta[b,d]pyridine (5a)** 2 mmol cyclopentanone (4) was taken in a round bottom flask and was treated with 1mmol of the concerned araldehyde/heteraldehyde(2a) and 1.5 mmol of ammonium acetate in 25ml in anhydrous methanol. The mixture was refluxed on water bath for 2 hr when colour changed to yellow. Reaction mixture was kept overnight at room temperature. The progress of reaction was monitored by TLC The solid thus settled was separated and the crude product was purified by running it over silica gel column using petroleum ether and ethyl acetate as eluent (9:1). When reaction was carried using ortho substituted aldehydes (6a,6b) under similar conditions of catalyst and solvent, the product obtained was entirely different and were characterised as azabicyclo compounds 7a,7b. The compounds 5(b-g) were obtained by a similar procedure to that described for the preparation of 5a using aldehydes 2(a-e, g-h).

TABLE- 2 Physical and analytical data of the compounds (5a-g)

Product 5(a-g)	Ar/Het. (2a-e, 2g-h)	M.Pt (°C)	Yield (%)	Molecular formula	Analysis %		
					Calcd./Found	C	H
5a	Phenyl	185	45	C <sub>17</sub> H <sub>17</sub> N	86.77	7.28	5.95
					86.74	7.13	5.92
5b	4-Methoxyphenyl	153	50	C <sub>18</sub> H <sub>19</sub> NO	81.47	7.22	5.28
					81.38	7.15	5.21
5c	4-Chlorophenyl	165	42	C <sub>17</sub> H <sub>16</sub> ClN	75.69	5.98	5.19
					75.42	5.78	5.13
5d	3,4-Dimethoxyphenyl	189	48	C <sub>19</sub> H <sub>21</sub> NO <sub>2</sub>	77.26	7.17	4.74
					77.21	7.13	4.23
5e	3,4-Methylenedioxyphenyl	174	40	C <sub>18</sub> H <sub>17</sub> NO <sub>2</sub>	77.40	6.13	5.01
					77.28	5.98	4.47
5f	2-Furyl	Oily liquid	35	C <sub>15</sub> H <sub>15</sub> NO	79.97	6.71	6.22
					79.91	6.68	6.18
5g	3-Indolyl	192	41	C <sub>19</sub> H <sub>18</sub> N <sub>2</sub>	83.18	6.61	10.21
					83.13	6.41	10.14

**Spectral data****5-Phenyl-1,2,3,6,7,8-hexahydrodicyclopenta[b,d]pyridine(5a)**

Green solid, IR (KBr,  $\nu$  cm<sup>-1</sup>): 3052, 2992, 1297, 1589, 1448. <sup>1</sup>H NMR(CDCl<sub>3</sub>, 400MHz)  $\delta$ : 1.85-2.1 (m, 4H), 2.8 (t, 8H), 7.1-7.5 (m, 5H); <sup>13</sup>C NMR (CDCl<sub>3</sub>, 100 MHz)  $\delta$ : 24.8, 25.3, 25.7, 26.7, 26.8, 35.4, 127.1, 127.8, 126.8, 128.8, 129.5, 132.7, 135.1, 135.9, 151.6, 157.3, 165.2. EIMS m/z = 236 (M+1)<sup>+</sup>.

**5-(4-Methoxyphenyl)-1,2,3,6,7,8-hexahydrodicyclopenta[b,d]pyridine (5b)**

Light yellow solid, IR (KBr,  $\nu$  cm<sup>-1</sup>) 3037, 2994, 1568, 1228, 1417. <sup>1</sup>H NMR(CDCl<sub>3</sub>, 400MHz)  $\delta$ : 1.91-2.4 (m, 4H), 2.83-2.93(t, 8H), 3.71(s, 3H), 7.45-7.81(m, 4H). <sup>13</sup>C NMR(CDCl<sub>3</sub>, 100MHz)  $\delta$ : 25.3, 25.7, 26.3, 27.1, 27.4, 35.6, 55.2, 114.5, 114.5, 128.3, 128.9, 128.5, 134.1, 134.8, 150.2, 155.2, 159.4, 164.3. EIMS m/z = 266 (M+1)<sup>+</sup>.

**5-(4-Chlorophenyl)-1,2,3,6,7,8-hexahydrodicyclopenta[b,d] pyridine (5c)**

Yellow grey solid, IR (KBr,  $\nu$  cm<sup>-1</sup>): 3043, 3018, 1572, 1278, 1242, 700. <sup>1</sup>H NMR(CDCl<sub>3</sub>, 400 MHz)  $\delta$ : 1.93-2.23 (m, 4H), 2.65-2.89 (t, 8H), 7.36-7.89 (m, 4H). <sup>13</sup>C NMR (CDCl<sub>3</sub>, 100 MHz)  $\delta$ : 24.3, 25.7, 26.7, 27.1, 27.8, 35.4, 128.5, 128.8, 129.1, 129.1, 132.2, 134.4, 134.5, 136.1, 152.5, 156.1, 164.3. EIMS m/z = 270 (M+1).

**5-(3,4-Dimethoxyphenyl)-1,2,3,6,7,8-hexahydrodicyclopenta[b,d] pyridine (5d)**

Bright Orange solid, IR (KBr,  $\nu$  cm<sup>-1</sup>): 3033, 2839, 1563, 1422, 1272. <sup>1</sup>H NMR (CDCl<sub>3</sub>, 400MHz)  $\delta$ : 1.89-2.44 (m, 4H), 2.72-2.78(t, 8H),  $\delta$ 3.65(s, 6H), 7.03-7.75(m, 3H). <sup>13</sup>C NMR (CDCl<sub>3</sub>, 100 MHz):  $\delta$  25.3, 26.3, 26.8, 27.5, 35.1, 56.2, 56.2, 112.3, 115.3, 120.5, 129.1, 134.1, 135.8, 148.2, 150.1, 150.3, 155.4, 155.8, 163.8. EIMS m/z = 296 (M+1)<sup>+</sup>.

**5-(3,4-Methylenedioxy phenyl)-1,2,3,6,7,8-hexahydrodicyclopenta[b,d] pyridine(5e)**

Yellow solid, IR (KBr,  $\nu$  cm<sup>-1</sup>): 3049, 2956, 1581, 1289, 1156, 1135. <sup>1</sup>H NMR (CDCl<sub>3</sub>, 400 MHz)  $\delta$ : 1.87-2.23(m, 4H), 2.7-2.81(t, 8H), 5.93(s, 2H), 6.7(d, 1H), 7.2-7.4(m, 2H). <sup>13</sup>C NMR (CDCl<sub>3</sub>, 100 MHz)  $\delta$ : 25.6, 25.7, 26.4, 27.1, 27.4, 35.3, 101.5, 111.2, 115.4, 120.6, 129.7, 134.2, 135.7, 147.3, 149.2, 151.2, 153.2, 164.2. EIMS m/z = 280(M+1)<sup>+</sup>.

**5-(2-Furyl)-1,2,3,6,7,8-hexahydrodicyclopenta[b,d]pyridine(5f)**

Oily liquid, IR (KBr,  $\nu$  cm<sup>-1</sup>): 3045, 2992, 1574, 1277, 1170. <sup>1</sup>H NMR (CDCl<sub>3</sub>, 400MHz)  $\delta$ : 1.97-2.01 (m, 4H), 2.4-2.9 (t, 8H), 6.23 (d, 2H), 7.34 (d, 1H). <sup>13</sup>C NMR (CDCl<sub>3</sub>, 100MHz):  $\delta$  24.2, 25.1, 26.3, 26.9, 27.8, 34.5, 104.9, 107.6, 134.8, 135.7, 142.4, 151.2, 155.9, 157.2, 164.5. EIMS m/z=226(M+1)<sup>+</sup>.

**5-(3-Indolyl)-1,2,3,6,7,8-hexahydrodicyclopenta[b,d]pyridine(5g)**

Black solid, IR (KBr,  $\nu$  cm<sup>-1</sup>): 3043, 2917, 1578, 1242, 871. <sup>1</sup>H NMR (CDCl<sub>3</sub>, 400MHz)  $\delta$ : 1.8-2.2(m, 4H), 2.46-3.05(t, 8H), 7.3-7.89(m, 5H), 10.1(s, 1H NH). <sup>13</sup>C NMR (CDCl<sub>3</sub>, 100MHz):  $\delta$  25.3, 25.8, 26.2, 27.3, 27.8, 34.4, 134.2, 135.4, 135.9, 151.2, 163.6. EIMS m/z = 275(M+1)<sup>+</sup>.

**2,4-Di(o-chlorophenyl)-3-azabicyclo[3.2.1]octan-8-one.(7a)**

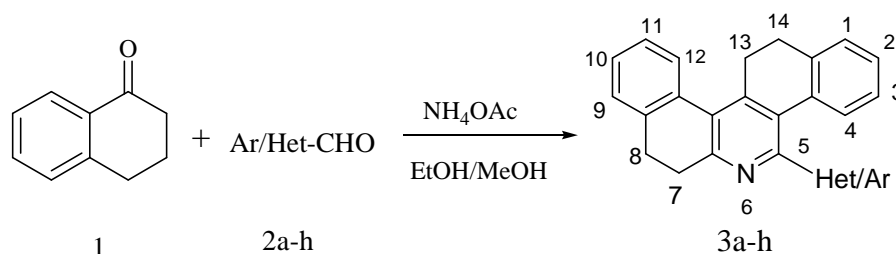
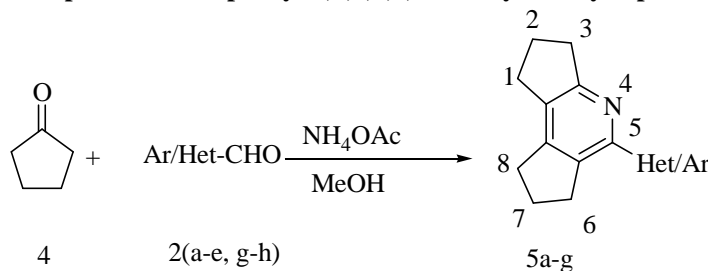
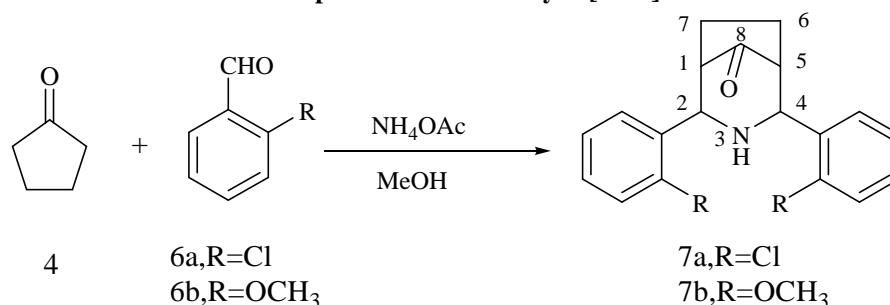
M.p: 206 °C, <sup>1</sup>H NMR (DMSO-d<sub>6</sub>, 400MHz)  $\delta$ : 1.86-2.2 (m, 4H), 2.7-2.9(m, 2H), 3.1(s, 1H, NH), 4.1-4.7 (d, 2H), 6.8-7.36 (m, 8H). <sup>13</sup>C NMR (DMSO-d<sub>6</sub>, 100 MHz)  $\delta$  12.8, 57.1, 64.3, 114.3, 120.5, 126.3, 127.2, 128.6, 129.4, 157.3, 215.5.

**2,4-Di(o-methoxyphenyl)- 3-azabicyclo[3.2.1]octan-8-one (7b)**

M.p: 214<sup>o</sup>C, <sup>1</sup>HNMR (DMSO-d<sub>6</sub>, 400MHz) δ: 1.87-2.1 (m,4H), 2.81-2.84(m,2H), 3.76(s,6H), 4.2(s,1H,NH), 4.3(s,2H), 7.1-7.4(m,8H). <sup>13</sup>CNMR (DMSO-d<sub>6</sub>, 100MHz): δ 12.7, 56.1, 57.1, 64.3, 114.2, 120.9, 127.1, 126.4, 129.2, 157.8, 216.3.

**RESULTS AND DISCUSSION**

With α-tetralone under similar conditions, all aryl and heteraldehydes including ortho and para substituted arylaldehydes in presence of ammonium acetate under anhydrous methanol media give entirely the dibenzophenanthridine (**3a-h**) products as shown in **Scheme-1**. No azabicyclo[3.3.0]nonane analogues were obtained as the alternative products even with the ortho substituted benzaldehydes. However, when reaction was tried with liquid ammonia in presence of fused sodium acetate in ethanol replacing the ammonium acetate under methanol/ethanol azabicyclononane[3.3.0] analogues were also obtained as the bi-products though in less than 5% ratio with the main dibenzophenanthridine product. Hence, we have discovered a one-step reaction with all the aryl and heteraldehydes substituted variedly in ortho and para positions with ammonium acetate and α-tetralone generating 5-aryl/hetaryl-7,8,13,14-tetrahydodibenzo[a,i]phenanthridines (**3a-h**).

**SCHEME-1: Preparation of 5-phenyl-7,8,13,14-tetrahydodibenzo[a,i]phenanthridine****SCHEME-2: Preparation of 5-phenyl-1,2,3,6,7,8-hexahydrodicyclopenta[b,d]pyridine****SCHEME-3: Preparation of 3-azabicyclo[3.2.1]octan-8-one**

In continuation to this work on α-tetralone, this one-step reaction was extended to other cyclic ketones like cyclopentanone and camphor. With cyclopentanone using ammonium acetate in methanol, the para substituted and ortho substituted aldehydes gave different products. With parasubstituted and unsubstituted aryl/ heteraldehydes, the products obtained were characterised as 1,2,3,6,7,8-hexahydrodicyclopenta[b,d]pyridines (**5a-g**) as exhibited by **Scheme-2**. However, the yield could not be obtained above 50% in all the cases. With ortho-substituted aromatic aldehydes using ammonium acetate in methanol, a new product was generated which was characterised as 3-azabicyclo[3.2.1]octan-8-one (**7**) (**Scheme-3**). The two reactions were not competing with each other and the product formation was exclusive but the yield was poorer in case of azabicyclo compounds. The same conditions when extended on camphor, it behaved like α-tetralone producing entirely a single product 6-Aryl/hetaryl-4,10-dimethyl-1,4;7,10-bisdimethylmethano-1,2,3,4,7,8,9,10-octahydrophenanthridine with ammonium acetate in

methanol. It is concluded logically that cyclic ketones like  $\alpha$ -tetralone and camphor which have only one methylene group adjoining the carbonyl functionality produce entirely single product **3** using ammonium acetate in methanol whereas cyclic ketones like cyclopentanone in which the carbonyl functionality is flanked by methylene moieties on either side produce two different products with differently substituted aryl aldehydes.

### Antimicrobial activity of the compounds

The compounds have been screened for their antifungal activity against *Asperigellus*, *Pencillium* and *Cladosporium* species. For antibacterial activity, these compounds have been screened against *Escherichia coli*, *Bacillus subtilis* and *Bacillus cereus*. Both the activities were evaluated at the same concentration of 1000 $\mu$ g and through well diffusion technique [22]. The standard antifungal agent flucanazole and antibacterial agent norfloxin were also screened under similar conditions for comparative study. The inhibition zones formed were measured in mm and are listed in **Table 3**.

**TABLE- 3: Antimicrobial activity of the compounds**

Compound. No.	Antibacterial activity	Antifungal activity				
	<i>E.coli</i>	<i>B.subtilis</i>	<i>B.cereus</i>	<i>Aniger</i>	<i>P. species</i>	<i>C.species</i>
3a	15	11	19	19	15	18
3b	13	12	17	16	13	16
3c	17	10	20	20	17	17
3d	14	11	18	16	16	18
3e	18	16	15	19	18	18
5a	20	12	17	14	16	16
5c	20	13	15	20	14	20
5d	19	14	14	17	18	19
NR	28	26	28	----	---	----
Flu	----	----	----	19	25	23

Note: 10mm, inactive; 11-15mm, weakly active; 16-28mm moderately active.  
NR: Norfloxin Flu: Flucanazole.

### CONCLUSION

In summary, we have established a one-pot efficient multicomponent synthesis of 5-aryl/heteryl-7,8,13,14-tetrahydrobenzo[a,i]phenanthridines and hitherto unknown bridged 2,4-diaryl-3-azabicyclo[3.2.1]octan-8-ones. The method has several advantages like creation of different novel heterocyclic systems, simple work up and easy handling.

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