# One-pot, pseudo four-component synthesis of spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-trione library

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### SUPPORTING INFORMATION

#### Proposed mechanism of the reaction:



#### **Experimental Section:**

**General Methods**: Melting points were measured on an Elecrtothermal 9100 apparatus and are uncorrected. <sup>1</sup>H and <sup>13</sup>C NMR spectra were recorded on a BRUKER DRX-300 AVANCE spectrometer at 300.13 and 75.47 MHz, respectively. <sup>1</sup>H and <sup>13</sup>C NMR spectra were obtained on solutions in DMSO- $d_6$  using TMS as internal standard. IR spectra were recorded using an FTIR apparatus. Elemental analyses were performed using a Heracus CHN-O-Rapid analyzer.

The chemical used in this work were obtained from Fluka and Merck and were used without purification.

Typical procedure for preparation of 5-phenyl-5*H*-spiro[diindeno[1,2-*b*:2',1'*e*]pyridine-11,3'-indoline]-2',10,12-trione (4a)



A mixture of 1,3-indandione (0.30 g, 2 mmol), aniline (0.09 g, 1 mmol), isatine (0.15 g, 1 mmol) and *p*-TSA (0.1 g) in refluxing acetonitrile (5 mL) was stirred for 30 min. After completion of the reaction confirmed by TLC (eluent: EtOAc/*n*-hexane, 1:3), the reaction mixture was cooled to room temperature. Then, the precipitated product was filtered and washed with water (10 mL) and ethanol (5 mL) to afford the pure product **4a** as a red powder (0.39 g, 82%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3437, 3132, 1703, 1624. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm H}$  (ppm) 5.46 (2H, d, <sup>3</sup>*J*<sub>HH</sub> = 6.0 Hz, H-Ar), 6.45-8.14 (15H, m, H-Ar), 10.65 (1H, s, NH). <sup>13</sup>C NMR (75 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm C}$  (ppm) 46.1, 109.5, 111.9, 121.8, 121.9, 122.7, 124.9, 125.9, 128.5, 129.0, 130.7, 132.1, 132.8, 134.8, 136.5, 138.2, 142.6, 156.2, 178.0, 190.0. Anal. Calcd for C<sub>32</sub>H<sub>18</sub>N<sub>2</sub>O<sub>3</sub>: C, 80.32; H, 3.79; N, 5.85. Found: C, 80.41; H, 3.71; N, 5.76.

5-(4-Bromophenyl)-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12-trione (4b).



Red Powder (yield 85%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3495, 1708, 1629. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_H$  (ppm) 5.56 (2H, d,  ${}^3J_{HH}$  = 6.0 Hz, H-Ar), 6.84-8.15 (14H, m, H-Ar), 10.65 (1H, s, NH). Anal. Calcd for C<sub>32</sub>H<sub>17</sub>BrN<sub>2</sub>O<sub>3</sub>: C, 68.95; H, 3.07; N, 5.03. Found: C, 68.92; H, 3.02; N, 4.96.

Due to very low solubility of the products **4b-4h**, we can not report the <sup>13</sup>C NMR data for these products.

5-(4-Nitrophenyl)-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12-trione (4c).



Red Powder (yield 80%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3337, 1713, 1613. <sup>1</sup>H NMR (300 MHz, DMSO-*d<sub>6</sub>*):  $\delta_{\rm H}$  (ppm) 5.55 (2H, d, <sup>3</sup>*J*<sub>HH</sub> = 6.0 Hz, H-Ar), 6.87-8.60 (14H, m, H-Ar), 10.66 (1H, s, NH). Anal. Calcd for C<sub>32</sub>H<sub>17</sub>N<sub>3</sub>O<sub>5</sub>: C, 73.42; H, 3.27; N, 8.03. Found: C, 73.49; H, 3.20; N, 8.11.

5-*p*-Tolyl-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12-trione (4d).



Red Powder (yield 90%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3379, 1729, 1698. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm H}$  (ppm) 2.56 (3H, s, CH<sub>3</sub>), 5.53 (2H, d, <sup>3</sup>*J*<sub>HH</sub> = 6.0 Hz, H-Ar), 6.85-7.99 (14H, m, H-Ar), 10.64 (1H, s, NH). Anal. Calcd for C<sub>33</sub>H<sub>20</sub>N<sub>2</sub>O<sub>3</sub>: C, 80.47; H, 4.09; N, 5.69. Found: C, 80.39; H, 4.01; N, 5.60.

5-(4-Methoxyphenyl)-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12-trione (4e).



Red Powder (yield 92%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3374, 1734, 1682. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_{\rm H}$  (ppm) 3.06 (3H, s, OCH<sub>3</sub>), 5.61 (2H, d, <sup>3</sup> $J_{\rm HH}$  = 6.0 Hz, H-

Ar), 6.84-8.05 (14H, m, H-Ar), 10.62 (1H, s, NH). Anal. Calcd for C<sub>33</sub>H<sub>20</sub>N<sub>2</sub>O<sub>4</sub>: C, 77.94; H, 3.96; N, 5.51. Found: C, 77.81; H, 3.89; N, 5.58.

1'-Methyl-5-phenyl-5H-spiro[diindeno[1,2-b:2',1'-e]pyridine-11,3'-indoline]-2',10,12-trione (4f).



Red Powder (yield 78%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3064, 1705, 1614. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_{\rm H}$  (ppm) 3.26 (3H, s, CH<sub>3</sub>), 5.47 (2H, d, <sup>3</sup> $J_{\rm HH}$  = 7.5 Hz, H-Ar), 6.98-8.15 (15H, m, H-Ar). Anal. Calcd for C<sub>33</sub>H<sub>20</sub>N<sub>2</sub>O<sub>3</sub>: C, 80.47; H, 4.09; N, 5.69. Found: C, 80.53; H, 4.17; N, 5.63.

5-(4-Bromophenyl)-1'-methyl-5H-spiro[diindeno[1,2-b:2',1'-e]pyridine-11,3'indoline]-2',10,12-trione (4g).



Red Powder (yield 78%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3069, 1708, 1614. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_H$  (ppm) 3.25 (3H, s, CH<sub>3</sub>), 5.58 (2H, d, <sup>3</sup> $J_{HH}$  = 7.2 Hz, H-Ar), 6.95-8.15 (14H, m, H-Ar). Anal. Calcd for C<sub>33</sub>H<sub>19</sub>BrN<sub>2</sub>O<sub>3</sub>: C, 69.36; H, 3.35; N, 4.90. Found: C, 69.29; H, 3.40; N, 4.81.

1'-Methyl-5-(4-nitrophenyl)-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'indoline]-2',10,12-trione (4h).



Red Powder (yield 75%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3059, 1703, 1614. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_H$  (ppm) 3.26 (3H, s, CH<sub>3</sub>), 5.57 (2H, d, <sup>3</sup> $J_{HH}$  = 7.8 Hz, H-Ar), 6.96-8.60 (14H, m, H-Ar). Anal. Calcd for C<sub>33</sub>H<sub>19</sub>N<sub>3</sub>O<sub>5</sub>: C, 73.74; H, 3.56; N, 7.82. Found: C, 73.85; H, 3.49; N, 7.89.

1'-Methyl-5-p-tolyl-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12trione (4i).



Red Powder (yield 84%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3059, 1702, 1614. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_{\rm H}$  (ppm) 2.57 (3H, s, CH<sub>3</sub>), 3.25 (3H, s, NCH<sub>3</sub>), 5.54 (2H, d,  ${}^{3}J_{\rm HH}$  = 7.4 Hz, H-Ar), 6.97-8.03 (14H, m, H-Ar). <sup>13</sup>C NMR (75 MHz, DMSO- $d_6$ ):  $\delta_{\rm C}$  (ppm) 21.5, 26.9, 45.6, 108.4, 111.6, 121.9, 122.0, 122.7, 124.6, 129.2, 130.1, 130.8,131.2, 132.7, 132.9, 133.9, 135.6, 136.6, 142.0, 144.0, 156.5, 176.5, 190.0. Anal. Calcd for C<sub>34</sub>H<sub>22</sub>N<sub>2</sub>O<sub>3</sub>: C, 80.62; H, 4.38; N, 5.53. Found: C, 80.53; H, 4.31; N, 5.44.

5-(4-Methoxyphenyl)-1'-methyl-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'indoline]-2',10,12-trione (4j).



Red Powder (yield 87%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3069, 1701, 1614. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_{\rm H}$  (ppm) 3.25 (3H, s, CH<sub>3</sub>), 3.96 (3H, s, OCH<sub>3</sub>), 5.62 (2H,

d,  ${}^{3}J_{HH}$  = 7.2 Hz, H-Ar), 6.95-8.06 (14H, m, H-Ar). Anal. Calcd for C<sub>34</sub>H<sub>22</sub>N<sub>2</sub>O<sub>4</sub>: C, 78.15; H, 4.24; N, 5.36. Found: C, 78.27; H, 4.29; N, 5.42.

Due to very low solubility of the product 4j, we can not report the <sup>13</sup>C NMR data for this product.

5'-Bromo-5-phenyl-5H-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12trione (4k).



Red Powder (yield 80%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3342, 3064, 1729, 1694. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_{\rm H}$  (ppm) 5.45 (2H, d,  ${}^{3}J_{\rm HH}$  = 7.2 Hz, H-Ar), 6.83-8.24 (14H, m, H-Ar), 10.80 (1H, s, NH). <sup>13</sup>C NMR (75 MHz, DMSO- $d_6$ ):  $\delta_{\rm C}$  (ppm) 46.3, 111.2, 111.4, 113.9, 121.9, 122.0, 127.8, 130.2, 130.7, 130.9, 131.7, 132.1, 132.7, 132.9, 136.5, 136.9, 138.2, 142.0, 156.6, 177.7, 190.0. Anal. Calcd for C<sub>32</sub>H<sub>17</sub>BrN<sub>2</sub>O<sub>3</sub>: C, 68.95; H, 3.07; N, 5.03. Found: C, 68.83; H, 3.13; N, 5.10.

5'-Bromo-5-(4-bromophenyl)-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'indoline]-2',10,12-trione (4l).



Red Powder (yield 80%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3379, 3080, 1698, 1629. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_{\rm H}$  (ppm) 5.56 (2H, d,  ${}^{3}J_{\rm HH}$  = 7.2 Hz, H-Ar), 6.83-8.23 (13H, m, H-Ar), 10.81 (1H, s, NH). <sup>13</sup>C NMR (75 MHz, DMSO- $d_6$ ):  $\delta_{\rm C}$  (ppm) 46.2, 111.4, 113.9, 121.9, 122.1, 125.5, 127.9, 130.9, 131.7, 132.6, 133.1, 133.9, 136.4, 136.9, 137.6, 142.0, 156.4, 177.6, 190.0. Anal. Calcd for  $C_{32}H_{16}Br_2N_2O_3$ : C, 60.40; H, 2.53; N, 4.40. Found: C, 60.30; H, 2.46; N, 4.34. 5'-Bromo-5-(4-nitrophenyl)-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12-trione (4m).



Red Powder (yield 76%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3385, 3080, 1740, 1698. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm H}$  (ppm) 5.55 (2H, d, <sup>3</sup>*J*<sub>HH</sub> = 7.5 Hz, H-Ar), 6.83-8.63 (13H, m, H-Ar), 10.81 (1H, s, NH). Anal. Calcd for C<sub>32</sub>H<sub>16</sub>BrN<sub>3</sub>O<sub>5</sub>: C, 63.80; H, 2.68; N, 6.98. Found: C, 63.88; H, 2.61; N, 6.93.

Due to very low solubility of the product 4m, we can not report the <sup>13</sup>C NMR data for this product.

5'-Bromo-5-p-tolyl-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12-trione (4n).



Red Powder (yield 85%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3232, 3064, 1703, 1619. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_{\rm H}$  (ppm) 2.57 (3H, s, CH<sub>3</sub>), 5.52 (2H, d,  ${}^{3}J_{\rm HH}$  = 7.2 Hz, H-Ar), 6.82-8.09 (13H, m, H-Ar), 10.79 (1H, s, NH). <sup>13</sup>C NMR (75 MHz, DMSO- $d_6$ ):  $\delta_{\rm C}$ (ppm) 21.5, 46.3, 111.2, 111.4, 113.9, 122.0, 127.8, 129.8, 130.3, 130.9, 131.1, 131.7, 132.8, 132.9, 135.7,136.5, 137.0, 142.0, 156.8, 177.8, 190.0. Anal. Calcd for C<sub>33</sub>H<sub>19</sub>BrN<sub>2</sub>O<sub>3</sub>: C, 69.36; H, 3.35; N, 4.90. Found: C, 69.47; H, 3.42; N, 4.81.

5'-Bromo-5-(4-methoxyphenyl)-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'indoline]-2',10,12-trione (40).



Red Powder (yield 88%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3332, 3059, 1726, 1619. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_{\rm H}$  (ppm) 3.96 (3H, s, OCH<sub>3</sub>), 5.60 (2H, d,  ${}^{3}J_{\rm HH}$  = 7.5 Hz, H-Ar), 6.83-8.13 (13H, m, H-Ar), 10.80 (1H, s, NH). <sup>13</sup>C NMR (75 MHz, DMSO- $d_6$ ):  $\delta_{\rm C}$ (ppm) 46.3, 56.3, 111.2, 111.4, 113.9, 115.5, 115.7, 121.9, 122.1, 127.8, 130.7, 130.8, 131.2, 131.7, 132.8, 133.0, 136.6, 137.0, 142.0, 157.1, 161.5, 177.8, 190.1. Anal. Calcd for C<sub>33</sub>H<sub>19</sub>BrN<sub>2</sub>O<sub>4</sub>: C, 67.47; H, 3.26; N, 4.77. Found: C, 67.54; H, 3.33; N, 4.85.

5'-Nitro-5-phenyl-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12trione (4p).



Red Powder (yield 84%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3311, 3064, 1745, 1698. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm H}$  (ppm) 5.48 (2H, d, <sup>3</sup>*J*<sub>HH</sub> = 7.2 Hz, H-Ar), 7.10-8.45 (14H, m, H-Ar), 11.41 (1H, s, NH). Anal. Calcd for C<sub>32</sub>H<sub>17</sub>N<sub>3</sub>O<sub>5</sub>: C, 73.42; H, 3.27; N, 8.03. Found: C, 73.31; H, 3.22; N, 8.92.

Due to very low solubility of the products **4p**,**q**, we can not report the <sup>13</sup>C NMR data for these products.

5-(4-Bromophenyl)-5'-nitro-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12-trione (4q).



Red Powder (yield 82%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3364, 3064, 1740, 1703. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm H}$  (ppm) 5.57 (2H, d, <sup>3</sup>*J*<sub>HH</sub> = 6.0 Hz, H-Ar), 7.09-8.46 (13H, m, H-Ar), 11.42 (1H, s, NH). Anal. Calcd for C<sub>32</sub>H<sub>16</sub>BrN<sub>3</sub>O<sub>5</sub>: C, 63.80; H, 2.68; N, 6.98. Found: C, 63.89; H, 2.60; N, 6.91.

5'-Nitro-5-(4-nitrophenyl)-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12-trione (4r).



Red Powder (yield 77%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3490, 3206, 1729, 1698. <sup>1</sup>H NMR (300 MHz, DMSO-*d<sub>6</sub>*):  $\delta_{\rm H}$  (ppm) 5.57 (2H, d, <sup>3</sup>*J*<sub>HH</sub> = 7.5 Hz, H-Ar), 7.09-8.67 (13H, m, H-Ar), 11.43 (1H, s, NH). <sup>13</sup>C NMR (75 MHz, DMSO-*d<sub>6</sub>*):  $\delta_{\rm C}$  (ppm) 46.1, 109.7, 110.8, 121.1, 122.1, 122.4, 126.0, 126.7, 131.0, 132.4, 133.4, 135.3, 136.2, 143.0, 143.4, 149.2, 149.7, 156.6, 178.6, 190.0. Anal. Calcd for C<sub>32</sub>H<sub>16</sub>N<sub>4</sub>O<sub>7</sub>: C, 67.61; H, 2.84; N, 9.86. Found: C, 67.74; H, 2.93; N, 9.77.

5'-Nitro-5-p-tolyl-5*H*-spiro[diindeno[1,2-*b*:2',1'-*e*]pyridine-11,3'-indoline]-2',10,12trione (4s).



Red Powder (yield 88%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3442, 3195, 1738, 1698. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm H}$  (ppm) 2.57 (3H, s, CH<sub>3</sub>), 5.54 (2H, d, <sup>3</sup>*J*<sub>HH</sub> = 6.9 Hz, H-Ar), 7.08-8.43 (13H, m, H-Ar), 11.40 (1H, s, NH). Anal. Calcd for C<sub>33</sub>H<sub>19</sub>N<sub>3</sub>O<sub>5</sub>: C, 73.74; H, 3.56; N, 7.82. Found: C, 73.62; H, 3.51; N, 7.74.

Due to very low solubility of the product 4s, we can not report the <sup>13</sup>C NMR data for this product.

5-(4-Methoxyphenyl)-5'-nitro-5H-spiro[diindeno[1,2-b:2',1'-e]pyridine-11,3'-

indoline]-2',10,12-trione (4t).



Red Powder (yield 91%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3301, 3064, 1740, 1698. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_{\rm H}$  (ppm) 3.96 (3H, s, OCH<sub>3</sub>), 5.61 (2H, d,  ${}^{3}J_{\rm HH}$  = 6.6 Hz, H-Ar), 7.08-8.43 (13H, m, H-Ar), 11.40 (1H, s, NH). <sup>13</sup>C NMR (75 MHz, DMSO- $d_6$ ):  $\delta_{\rm C}$ (ppm) 46.2, 56.2, 109.6, 110.5, 115.5, 115.7, 120.9, 122.0, 122.3, 126.6, 130.6, 131.0, 131.2, 131.7, 132.7, 133.1, 135.4, 136.6. 143.0, 149.2, 157.6, 161.5, 190.1. Anal. Calcd for C<sub>33</sub>H<sub>19</sub>N<sub>3</sub>O<sub>6</sub>: C, 71.61; H, 3.46; N, 7.59. Found: C, 71.70; H, 3.52; N, 7.65. **5'-Phenyl-5',5a'-dihydro-2***H***,4b'***H***-spiro[acenaphthylene-1,11'-diindeno[1,2-***b***:2',1'-**

*e*]pyridine]-2,10',12'(10a'*H*,11a'*H*)-trione (7a).



Red Powder (yield 80%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3442, 1725, 1687, 1621. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm H}$  (ppm) 5.48 (2H, d, <sup>3</sup>*J*<sub>HH</sub> = 7.2 Hz, H-Ar), 7.09-8.32 (19H, m, H-Ar). Anal. Calcd for C<sub>36</sub>H<sub>19</sub>NO<sub>3</sub>: C, 84.20; H, 3.73; N, 2.73. Found: C, 84.33; H, 3.67; N, 2.65.

Due to very low solubility of the products 7a-d we can not report the <sup>13</sup>C NMR data for these products.

5'-(4-Bromophenyl)-5',5a'-dihydro-2*H*,4b'*H*-spiro[acenaphthylene-1,11'-diindeno [1,2-*b*:2',1'-*e*]pyridine]-2,10',12'(10a'*H*,11a'*H*)-trione (7b).



Red Powder (yield 85%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3435, 1732, 1690,1619. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_{\rm H}$  (ppm) 5.60 (2H, d, <sup>3</sup> $J_{\rm HH}$  = 5.9 Hz, H-Ar), 7.17-8.31(18H, m, H-Ar). Anal. Calcd for C<sub>36</sub>H<sub>18</sub>BrNO<sub>3</sub>: C, 72.98; H, 3.06; N, 2.36. Found: C, 72.91; H, 3.11; N, 2.43.

5'-(4-Nitrophenyl)-5',5a'-dihydro-2*H*,4b'*H*-spiro[acenaphthylene-1,11'-diindeno[1,2*b*:2',1'-*e*]pyridine]-2,10',12'(10a'*H*,11a'*H*)-trione (7c).



Red Powder (yield 78%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3038, 1729, 1693, 1619. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm H}$  (ppm) 5.59 (2H, d, <sup>3</sup>*J*<sub>HH</sub> = 7.4 Hz, H-Ar), 7.11-8.62 (18H, m, H-Ar). Anal. Calcd for C<sub>36</sub>H<sub>18</sub>N<sub>2</sub>O<sub>5</sub>: C, 77.41; H, 3.25; N, 5.02. Found: C, 77.30; H, 3.18; N, 5.11.

5'-(4-Chlorophenyl)-5',5a'-dihydro-2*H*,4b'*H*-spiro[acenaphthylene-1,11'-diindeno [1,2-*b*:2',1'-*e*]pyridine]-2,10',12'(10a'*H*,11a'*H*)-trione (7d).



Red Powder (yield 83%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3737, 1734, 1687, 1614. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm H}$  (ppm) 5.60(2H, d, <sup>3</sup>*J*<sub>HH</sub> = 7.0 Hz, H-Ar), 7.15-8.34 (18H, m, H-Ar). Anal. Calcd for C<sub>36</sub>H<sub>18</sub>ClNO<sub>3</sub>: C, 78.90; H, 3.31; N, 2.56. Found: C, 78.88; H, 3.39; N, 2.64.

5'-*p*-Tolyl-5',5a'-dihydro-2*H*,4b'*H*-spiro[acenaphthylene-1,11'-diindeno[1,2-*b*:2',1'*e*]pyridine]-2,10',12'(10a'*H*,11a'*H*)-trione (7e).



Red Powder (yield 88%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3500, 1697, 1619, 1610. <sup>1</sup>H NMR (300 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm H}$  (ppm) 2.57 (3H, s, CH<sub>3</sub>), 5.56(2H, d, <sup>3</sup>*J*<sub>HH</sub> = 7.4 Hz, H-Ar), 7.14-8.33 (18H, m, H-Ar). <sup>13</sup>C NMR (75 MHz, DMSO-*d*<sub>6</sub>):  $\delta_{\rm C}$  (ppm) 21.3, 50.9, 109.7, 113.1, 113.5, 117.3, 121.1, 121.8, 122.0, 125.2, 128.9, 129.2, 129.9, 130.6, 131.0, 131.8, 132.6, 132.8, 135.7, 136.7, 141.2, 142.0, 156.6, 158.0, 158.6, 159.1, 159.6, 190.3, 204.5, 206.7. Anal. Calcd for C<sub>37</sub>H<sub>21</sub>NO<sub>3</sub>: C, 84.23; H, 4.01; N, 2.65. Found: C, 84.10; H, 3.94; N, 2.71.

5'-(4-Methoxyphenyl)-5',5a'-dihydro-2*H*,4b'*H*-spiro[acenaphthylene-1,11'-diindeno [1,2-*b*:2',1'-*e*]pyridine]-2,10',12'(10a'*H*,11a'*H*)-trione (7f).



Red Powder (yield 90%); mp >300 °C. IR (KBr) ( $v_{max}$  /cm<sup>-1</sup>): 3432, 1728, 1694, 1622. <sup>1</sup>H NMR (300 MHz, DMSO- $d_6$ ):  $\delta_H$  (ppm) 3.97 (3H, s, OCH<sub>3</sub>), 5.64 (2H, d,  ${}^3J_{HH}$  = 7.1 Hz, H-Ar), 7.13-8.31 (18H, m, H-Ar). Anal. Calcd for C<sub>37</sub>H<sub>21</sub>NO<sub>4</sub>: C, 81.76; H, 3.89; N, 2.58% Found: C, 81.87; H, 3.83; N, 2.50.

Due to very low solubility of the product 7f, we can not report the <sup>13</sup>C NMR data for this product.









































































