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# Applications of novel composite UiO-66-NH<sub>2</sub>/Melamine with phosphorous acid tags as a porous and efficient catalyst for the preparation of novel spiro-oxindoles

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#### Spectral data

### 3'-(1*H*-Indol-3-yl)-5-iodo-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (1a).

Cream soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3414, 3248, 1717, 1630,1578; <sup>1</sup>H NMR (400 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  12.36 (s, 1H), 11.22 (s, 1H), 10.51 (s, 1H), 10.18 (s, 1H), 10.01 (s, 1H), 9.32 (s, 1H), 7.38 (dd, J = 8.1, 1.8 Hz, 1H), 7.32 (d, J = 8.2 Hz, 1H), 7.27 (d, J = 1.9 Hz, 1H), 7.18 (d, J = 7.8 Hz, 1H), 7.08 (t, J = 6.3 Hz, 1H), 6.96 (t, J = 6.4 Hz, 1H), 6.35 (d, J = 8.1 Hz, 1H), 5.96 (d, J = 2.4 Hz, 1H). <sup>13</sup>C NMR (101 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  178.4, 142.33, 135.8, 135.3, 133.6, 131.4, 126.3, 124.2, 121.4, 119.2, 118.8, 111.3, 111.1, 100.0, 83.7, 47.5. MS m/z (%) found for C<sub>23</sub>H<sub>14</sub>IN<sub>7</sub>O<sub>3</sub>: 563.1

### 3'-(1*H*-Indol-3-yl)-5-iodo-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'H,8'*H*)-trione (2a).

Cream soild M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3417, 3213, 2834, 1686, 1673, 1654, 1610. <sup>1</sup>H NMR (400 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  12.46 (s, 1H), 11.23 (s, 1H), 10.25 (s, 1H), 10.00 (s, 1H), 7.37 (dd, J = 8.1, 1.9 Hz, 1H), 7.33 (d, J = 8.1 Hz, 1H), 7.29 (d, J = 2.0 Hz, 1H), 7.17 (d, J = 8.1 Hz, 1H), 7.09 (t, J = 7.6 Hz, 1H), 6.96 (t, J = 7.5 Hz, 1H), 6.33 (d, J = 8.1 Hz, 1H), 5.98 (d, J = 2.7 Hz, 1H), 3.53 (s, 3H), 3.01 (s, 3H). <sup>13</sup>C NMR (101 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  178.5, 159.8, 150.4, 147.7, 142.3, 141.1, 135.7, 135.3, 133.3, 131.4, 126.4, 124.3, 121.4, 119.2, 118.7, 111.3, 111.1, 83.7, 48.4, 30.3, 27.3. MS m/z (%) found for C<sub>25</sub>H<sub>18</sub>IN<sub>7</sub>O<sub>3</sub>: 591.1

### 5-Chloro-3'-(1*H*-indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (3a).

Yellow soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3408, 3235, 1718, 1648, 1554, <sup>1</sup>H NMR (400 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  11.09 (s, 1H), 10.72 (s, 1H), 10.34 (s, 1H), 10.21 (s, 1H), 9.41 (s, 1H), 7.75 (d, J = 7.9 Hz, 2H), 7.71 – 7.64 (m, 3H), 7.50 (t, J = 7.3 Hz, 1H), 7.30 (d, J = 8.1 Hz, 1H), 7.24 (d, J = 2.3 Hz, 1H), 7.20 (dd, J = 8.1, 2.2 Hz, 1H), 7.07 (t, J = 7.6 Hz,

1H), 6.96 (t, J = 7.6 Hz, 1H), 6.67 (d, J = 8.1 Hz, 1H), 6.09 (d, J = 2.7 Hz, 1H). <sup>13</sup>C NMR (101 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  178.5, 161.8, 149.4, 143.9, 141.73, 139.0, 137.7, 135.5, 129.8, 127.7, 127.5, 126.5, 125.3, 124.2, 123.7, 122.9, 121.3, 120.3, 119.0, 111.1, 110.1, 106.5, 99.1, 43.2. MS m/z (%) found for C<sub>29</sub>H<sub>18</sub>ClN<sub>7</sub>O<sub>3</sub>: 547.10

### 5-Chloro-3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione(4a).

Cream soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3228, 3189, 3123, 2925, 1688, 1641, 1628, 1608. <sup>1</sup>H NMR (400 MHz, DMS- $d_6$ )  $\delta_{ppm}$  12.46 (s, 1H), 11.25 (s, 1H), 10.27 (s, 1H), 10.02 (s, 1H), 7.34 (d, J = 8.2 Hz, 1H), 7.18 (d, J = 8.1 Hz, 1H), 7.11 – 7.06 (m, 3H), 6.96 (t, J = 7.5 Hz, 1H), 6.47 (d, J = 8.6 Hz, 1H), 6.04 (d, J = 2.7 Hz, 1H), 3.54 (s, 3H), 3.01 (s, 3H). <sup>13</sup>C NMR (101 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  178.9, 159.8, 150.4, 147.7, 146.1, 141.5, 140.3, 135.3, 133.3, 127.0, 126.4, 124.9, 124.3, 123.3, 121.4, 119.2, 118.7, 111.3, 109.8, 102.3, 100.0, 85.5, 48.7, 30.3, 27.3.

## 3'-(1*H*-Indol-3-yl)-6',8'-dimethyl-1',9'-dihydro-2H-spiro[acenaphthylene-1,4' pyrazolo[4',3':5,6]pyrido[2,3-d]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (5a).

Brown soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>):3433, 3330, 2936, 1727, 1698, 1653, 1567. <sup>1</sup>H NMR (400 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  12.35 (s, 1H), 10.63 (s, 1H), 10.34 (s, 1H), 7.96 (d, J = 8.2 Hz, 1H), 7.75 (d, J = 8.3 Hz, 1H), 7.54 (t, J = 7.7 Hz, 1H), 7.47 (t, J = 7.6 Hz, 1H), 7.33 (d, J = 6.9 Hz, 2H), 7.04 (d, J = 8.2 Hz, 1H), 6.93 (t, J = 7.4 Hz, 1H), 6.84 (d, J = 7.9 Hz, 1H), 6.79 (t, J = 7.1 Hz, 1H), 5.37 (s, 1H), 3.59 (s, 3H), 2.88 (s, 3H). <sup>13</sup>C NMR (101 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  204.7, 160.3, 150.5, 147.8, 147.0, 146.0, 140.0, 134.7, 133.5, 133.1, 129.9, 129.2, 128.4, 127.4, 126.0, 124.4, 123.5, 121.1, 119.8, 119.3, 119.0, 118.3, 110.8, 103.0, 101.8, 87.2, 52.8, 30.4, 27.2.

3'-(1*H*-Indol-3-yl)-1',9'-dihydro-2*H*-spiro[acenaphthylene-1,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (6a). Cream soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3433, 3330, 2936, 1722, 1698, 1653. <sup>1</sup>H NMR (400 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  12.26 (s, 1H), 10.62 (s, 1H), 10.42 (s, 1H), 10.26 (s, 1H), 9.36 (s, 1H), 7.97 (d, J = 8.1 Hz, 1H), 7.76 (d, J = 8.3 Hz, 1H), 7.56 (t, J = 7.7 Hz, 1H), 7.48 (t, J = 7.6 Hz, 1H), 7.33 (t, J = 6.6 Hz, 2H), 7.04 (d, J = 8.1 Hz, 1H), 6.93 (t, J = 7.5 Hz, 1H), 6.85 (d, J = 7.8 Hz, 1H), 6.79 (t, J = 7.5 Hz, 1H), 5.33 (s, 1H). <sup>13</sup>C NMR (101 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  204.7, 162.2, 146.8, 145.3, 140.1, 134.7, 133.5, 133.5, 130.0, 129.3, 128.4, 127.5, 125.9, 124.3, 123.5, 121.1, 119.8, 119.3, 119.0, 118.3, 110.8, 102.9, 101.8, 86.6, 52.0.

### 3'-(1*H*-Indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indeno[1,2-b]quinoxaline-11,4' pyrazolo[4',3':5,6]pyrido[2,3-d]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (7a).

Brown soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3168, 3050, 2890, 1688, 1634, 1518. <sup>1</sup>H NMR (400 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  12.37 (s, 1H), 10.87 (s, 1H), 10.42 (s, 1H), 7.89 (t, J = 9.9 Hz, 2H), 7.68 – 7.62 (m, 3H), 7.43 (d, J = 6.6 Hz, 2H), 7.30 (t, J = 8.1 Hz, 1H), 7.06 (d, J = 8.1 Hz, 1H), 6.79 (t, J = 7.7 Hz, 1H), 6.59 (d, J = 8.1 Hz, 1H), 6.44 (t, J = 7.5 Hz, 1H), 5.46 (s, 1H), 3.63 (s, 3H), 2.80 (s, 3H). <sup>13</sup>C NMR (101 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  <sup>13</sup>C NMR (101 MHz, DMSO- $d_6$ )  $\delta_{167.8}$ , 159.6, 154.8, 154.6, 150.5, 148.0, 146.14, 141.0, 140.8, 136.0, 134.7, 131.6, 128.5, 128.5, 128.4, 128.1, 127.5, 125.9, 124.6, 123.7, 121.1, 120.4, 118.5, 118.0, 110.9, 102.8, 87.0, 48.7, 30.5, 27.2.

# 3'-(1*H*-Indol-3-yl)-1',9'-dihydrospiro[indeno[1,2-b]quinoxaline-11,4'-pyrazolo[4',3':5,6]pyrido[2,3-d]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (8a).

Brown soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3190, 2890, 2808, 1722, 1642, 1604. <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ<sub>ppm</sub> 12.26 (s, 1H), 10.86 (s, 1H), 10.29 (s, 2H), 9.55 (s, 1H), 7.91 – 7.86 (m, 2H), 7.68 – 7.62 (m, 3H), 7.45 – 7.40 (m, 2H), 7.30 (t, *J* = 7.3 Hz, 1H), 7.05 (d, *J* = 8.2 Hz, 1H), 6.81 (t, *J* = 7.4 Hz, 1H), 6.63 (d, *J* = 8.1 Hz, 1H), 6.46 (t, *J* = 7.5 Hz, 1H), 5.45 (d, *J* = 2.6 Hz, 1H). <sup>13</sup>C NMR (101 MHz, DMSO-*d*<sub>6</sub>) δ<sub>ppm</sub> 167.5, 162.3, 161.7, 154.7,

154.6, 141.0, 140.8, 136.0, 134.7, 133.70, 131.65, 128.6, 128.5, 128.4, 128.2, 127.5, 125.8, 124.6, 123.6, 121.1, 120.53, 118.6, 118.1, 110.9, 102.6, 102.3, 86.4, 48.0.

### 3'-(1*H*-Indol-3-yl)-7-nitro-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (9a).

Brown soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3409, 1717, 1639, 1580; <sup>1</sup>H NMR (400 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  12.35 (s, 1H), 10.88 (s, 1H), 10.52 – 10.37 (m, 2H), 9.67 (s, 1H), 8.73 (d, J = 2.7 Hz, 1H), 8.35 (dd, J = 9.1, 2.6 Hz, 1H), 8.00 – 7.95 (m, 2H), 7.71 (d, J = 7.5 Hz, 1H), 7.56 (t, J = 7.5 Hz, 1H), 7.38 (t, J = 7.4 Hz, 1H), 7.03 (d, J = 8.2 Hz, 1H), 6.72 (t, J = 8.1 Hz, 1H), 6.47 (d, J = 7.9 Hz, 1H), 6.32 (t, J = 7.4 Hz, 1H), 5.67 (d, J = 2.7 Hz, 1H). <sup>13</sup>C NMR (101 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  169.7, 162.3, 161.8, 157.2, 155.2, 146.0, 144.2, 139.6, 134.8, 134.7, 133.6, 133.1, 129.8, 127.9, 125.9, 124.79, 124.5, 124.1, 122.1, 121.2, 121.0, 118.4, 117.8, 110.8, 103.0, 85.9, 47.9.

## 3'-(1*H*-Indol-3-yl)-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (10a).

Yellow soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3419, 3377, 3212, 3047, 2935, 2856,1722.1652. <sup>1</sup>H NMR (250 MHz, DMSO-*d*<sub>6</sub>) δ<sub>ppm</sub> 12.25 (s, 1H), 11.14 (s, 1H), 10.43 (s, 1H), 10.10 (s, 1H), 9.86 (s, 1H), 9.23 (s, 1H), 7.26 (dd, *J* = 13.1, 8.1 Hz, 2H), 7.10-6.91 (m, 5H), 6.55 (d, *J* = 7.6 Hz, 1H), 5.73 (s, 1H). <sup>13</sup>C NMR (63 MHz, DMSO-*d*<sub>6</sub>) δ<sub>ppm</sub> 179.3, 162.2, 150.2, 147.8, 145.9, 143.1, 138.8, 135.9, 134.2, 127.7, 126.7, 124.8, 123.8, 121.8, 121.5, 119.7, 119.4, 111.7, 109.2, 103.1, 85.9, 48.0.

#### 3'-(1*H*-Indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (11a).

Cream soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3274, 3088, 288, 1679, 1639, 1607. <sup>1</sup>H NMR (250 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  12.36 (s, 1H), 11.15 (s, 1H), 10.14 (s, 1H), 9.87 (s, 1H), 7.27 (dd, J = 15.3, 7.9 Hz, 2H), 7.10 – 6.94 (m, 4H), 6.85 (t, J = 7.5 Hz, 1H), 6.55 (d, J = 7.6 Hz, 1H), 5.75 (s, 1H), 3.49 (s, 3H), 2.98 (s, 3H). <sup>13</sup>C NMR (63 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  179.6,

160.2, 150.9, 148.0, 146.6, 143.1, 139.0, 135.9, 133.9, 127.7, 126.8, 124.9, 123.7, 121.9, 121.6, 119.7, 119.3, 111.8, 109.2, 103.0, 100.7, 86.6, 48.9, 30.7, 27.8.

### 3'-(1H-Indol-3-yl)-6',8'-dimethyl-1'-phenyl-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'-pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (12a).

Brown soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2) FT-IR (KBr, cm<sup>-1</sup>):3469, 3212, 1689, 1637, 1578. <sup>1</sup>H NMR (400 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  10.87 (s, 1H), 10.42 (s, 1H), 7.90 – 7.85 (m, 3H), 7.68 – 7.61 (m, 5H), 7.44 – 7.40 (m, 3H), 7.32 – 7.26 (m, 2H), 7.06 (d, J = 8.2 Hz, 1H), 6.79 (t, J = 7.0 Hz, 1H), 6.59 (d, J = 8.1 Hz, 1H), 6.44 (t, J = 7.4 Hz, 1H), 5.46 (d, J = 2.7 Hz, 1H), 3.63 (s, 3H), 2.80 (s, 3H).<sup>13</sup>C NMR (101 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  167.8, 159.6, 154.8, 154.6, 150.5, 148.0, 146.1, 141.0, 140.8, 136.0, 134.7, 133.3, 131.6, 128.5, 128.5, 128.4, 128.2, 127.5, 125.9, 124.6, 123.7, 121.1, 120.4, 118.5, 118.0, 110.9, 102.8, 102.1, 86.9, 48.7, 30.5, 27.2.

### 5-Chloro-3'-(1*H*-indol-3-yl)-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (13a).

Cream soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3301, 3195, 3100, 2994, 1720, 1688, 1623, 1574. <sup>1</sup>H NMR (250 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  12.35 (s, 1H), 11.16 (s, 1H), 10.50 (s, 1H), 10.24 (s, 1H), 9.98 (s, 1H), 9.35 (s, 1H), 7.30 (d, J = 8.3 Hz, 1H), 7.16 (d, J = 7.9 Hz, 1H), 7.06 – 7.02 (m, 3H), 6.92 (t, J = 7.6 Hz, 1H), 6.47 (d, J = 8.2 Hz, 1H), 6.02 (s, 1H). <sup>13</sup>C NMR (63 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  179.3, 162.3, 150.3, 148.0, 145.8, 141.8, 140.4, 135.8, 134.3, 127.5, 126.76, 125.6, 124.8, 123.8, 122.0, 119.7, 119.2, 111.8, 110.4, 102.8, 100.4, 85.4, 48.3.

### 3'-(1*H*-Indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (14a).

Brown soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>): 3408, 3248, 3053, 2819, 1698, 1639. <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ<sub>ppm</sub> 11.68 (m, 2H), 10.78 (s, 2H), 8.42 (m, 3H), 8.12 (m, 4H), 7.52 (m, 5H), 7.33 (m, 6H).

3'-(1*H*-Indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (15a). Yellow soild, M.p: >300 °C; (Ethyl acetate: Methanol 8:2), FT-IR (KBr, cm<sup>-1</sup>); 3422, 3225, 2800, 1715, 1640. <sup>1</sup>H NMR (400 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  11.05 (s, 1H), 10.69 (s, 1H), 10.21 (s, 2H), 9.36 (s, 1H), 7.82 – 7.62 (m, 5H), 7.50 (s, 1H), 7.27 (d, 1H), 7.24 – 7.14 (m, 2H), 7.06 (s, 1H), 6.95 (d, J = 6.4 Hz, 2H), 6.72 (d, 1H), 5.88 (s, 1H). <sup>13</sup>C NMR (101 MHz, DMSO- $d_6$ )  $\delta_{ppm}$  178.6, 161.7, 149.3, 145.9, 144.1, 142.9, 137.7, 129.8, 127.9, 126.5, 124.1, 123.9, 122.8, 121.3, 120.4, 119.0, 111.0, 87.9, 46.4.



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-5-iodo-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (1a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-5-iodo-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (1a)





Mass spectrum of 3'-(1*H*-indol-3-yl)-5-iodo-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (1a)



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-5-iodo-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (2a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-5-iodo-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (2a)



<sup>13</sup>C-NMR spectrum of 3'-(1*H*-indol-3-yl)-5-iodo-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (2a)



Mass spectrum of 3'-(1*H*-indol-3-yl)-5-iodo-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (2a)



FT-IR spectrum of 5-chloro-3'-(1*H*-indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (3a)



<sup>1</sup>H-NMR spectrum of 5-chloro-3'-(1*H*-indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (3a)



<sup>13</sup>C-NMR spectrum of 5-chloro-3'-(1*H*-indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (3a)



Mass spectrum of 5-chloro-3'-(1*H*-indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (3a)



FT-IR spectrum of 5-chloro-3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (4a)



<sup>1</sup>H-NMR spectrum of 5-chloro-3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (4a)



<sup>13</sup>C-NMR spectrum of 5-chloro-3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (4a)



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydro-2*H*-spiro[acenaphthylene-1,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (5a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydro-2*H*-spiro[acenaphthylene-1,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (5a)



<sup>13</sup>C-NMR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydro-2*H*-spiro[acenaphthylene-1,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (5a)



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-1',9'-dihydro-2*H*-spiro[acenaphthylene-1,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (6a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-1',9'-dihydro-2*H*-spiro[acenaphthylene-1,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (6a)



<sup>13</sup>C-NMR spectrum of 3'-(1*H*-indol-3-yl)-1',9'-dihydro-2H-spiro[acenaphthylene-1,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (6a)



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (7a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (7a)



<sup>13</sup>C-NMR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (7a)



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (8a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (8a)



<sup>13</sup>C-NMR spectrum of 3'-(1*H*-indol-3-yl)-1',9'-dihydrospiro[indeno[1,2-b]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (8a)



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-7-nitro-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (9a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-7-nitro-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (9a)



<sup>13</sup>C-NMR spectrum of 3'-(1*H*-indol-3-yl)-7-nitro-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (9a)



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (10a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (10a)



<sup>13</sup>C-NMR spectrum of 3'-(1*H*-indol-3-yl)-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (10a)



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (11a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (11a)



<sup>13</sup>C NMR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1',9'-dihydrospiro[indoline-3,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione(11a)



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1'-phenyl-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione(12a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1'-phenyl-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (12a)



<sup>13</sup>C-NMR spectrum of 3'-(1*H*-indol-3-yl)-6',8'-dimethyl-1'-phenyl-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'-pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione(12a)



FT-IR spectrum of 5-chloro-3'-(1*H*-indol-3-yl)-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3d]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (13a)



<sup>1</sup>H-NMR spectrum of 5-chloro-3'-(1*H*-indol-3-yl)-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (13a).



<sup>13</sup>C- NMR spectrum of 5-chloro-3'-(1*H*-indol-3-yl)-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (13a)



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indeno[1,2-b]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (14a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indeno[1,2-*b*]quinoxaline-11,4'pyrazolo[4',3':5,6]pyrido[2,3-*d*]pyrimidine]-5',7'(6'*H*,8'*H*)-dione (14a)



FT-IR spectrum of 3'-(1*H*-indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (15a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (15a)



<sup>1</sup>H-NMR spectrum of 3'-(1*H*-indol-3-yl)-1'-phenyl-1',9'-dihydrospiro[indoline-3,4'-pyrazolo[4',3':5,6]pyrido[2,3*d*]pyrimidine]-2,5',7'(6'*H*,8'*H*)-trione (15a)