Supporting Information for

TEMPO-Catalyzed Oxidative Homocoupling Route
to 3,2'-Biindolin-2-ones via an Indolin-3-one Intermediate

Bo Yin, Panpan Huang, Yingbing Lu,* Liangxian Liu*

Key Laboratory of Organo-Pharmaceutical Chemistry of Jiangxi Province, Gannan Normal University, Ganzhou 341000, PR China
E-mail: lxliu@xmu.edu.cn

CONTENTS
1. General Methods..................................................................................................................................2
2. General Procedure and Spectroscopic Data of the Products 9 and 13................................................2
3. Copies of $^1$H, $^{13}$C Spectra.............................................................................................................8
4. Copies of HRESIMS Spectra.............................................................................................................25
5. X-ray Data of Compound 9a .............................................................................................................30
1. General Methods

Melting points were determined on a digital melting point apparatus and temperatures were uncorrected. Infrared spectra were measured with a Nicolet Avatar 360 FT-IR spectrometer using film KBr pellet techniques. $^1$H and $^{13}$C NMR spectra were recorded on a Bruker spectrometers at 400 and 100 MHz, respectively. Chemical shifts were reported in ppm relative to TMS for $^1$H and $^{13}$C NMR spectra. DMSO-$d_6$ was used as the NMR solvent. Mass spectra were recorded with Bruker Dalton Esquire 3000 plus LC-MS apparatus. Elemental analysis was carried out on a Perkin-Elmer 240B instrument. HRFABMS spectra were recorded on a FTMS apparatus. Silica gel (300-400 mesh) was used for flash column chromatography, eluting (unless otherwise stated) with an ethyl acetate/petroleum ether (PE) (60-90 °C) mixture.

2. General Procedure for the Preparation of 9.

To a solution of indole (0.3 mmol), Ag$_2$CO$_3$ (0.105 mmol), and NaOAc (0.03 mmol) in DMF (1 mL) was added TEMPO (0.045 mmol) under an air atmosphere and the mixture was stirred at 80 °C for 12 h. The reaction mixture was concentrated under reduced pressure. The residue was purified by flash chromatography on silica gel (eluent: EtOAc/PE = 1:4) to yield the corresponding product 9.

Spectroscopic Data of the Products 9.

2,3′-Bi(3H-indol)-3-one (9a)

Red solid, mp: 214-215 °C (from EtOAc/PE = 1:4). IR (KBr) $\nu_{\text{max}}$: 3187, 1719, 1604, 1559, 1447, 1368, 1240, 1135 cm$^{-1}$. $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 12.14 (s, 1H, NH), 8.50 (d, $J = 3.0$ Hz, 1H, Ar-H), 8.40 (dd, $J = 7.0$, 2.5 Hz, 1H, Ar-H), 7.59-7.50 (m, 3H, Ar-H), 7.34 (d, $J = 7.5$ Hz, 1H, Ar-H), 7.28-7.24 (m, 2H, Ar-H), 7.18 (dt, $J = 0.5$, 7.5 Hz, 1H, Ar-H).

$^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 195.7, 163.4, 158.5, 137.8, 137.3, 133.7, 126.8, 126.4, 124.9, 123.9, 123.0, 122.9, 122.2, 121.1, 113.0, 106.9. HRESIMS calcd for [C$_{16}$H$_{10}$N$_2$O + H]$^+$ 247.08714, found 247.08625.

5,5′-Difluoro-2,3′-bi(3H-indol)-3-one (9b)

Red solid, mp: 193-195 °C (from EtOAc/PE = 1:4). IR (KBr) $\nu_{\text{max}}$: 3460, 1723, 1626, 1572, 1469, 1358, 1263, 1125, cm$^{-1}$. $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 12.23 (s, 1H, NH), 8.49 (s, 1H, Ar-H), 8.02 (d, $J = 8.7$ Hz, 1H, Ar-H), 7.53 (q, $J = 4.3$ Hz, 1H, Ar-H), 7.38 (d, $J = 4.3$ Hz, 2H, Ar-H), 7.37 (s, 1H, Ar-H), 7.11 (t, $J = 8.7$ Hz, 1H, Ar-H). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 194.2, 161.2 (d, $J = 237.05$ Hz), 159.0 (d, $J = 2.3$ Hz), 158.9 (d, $J = 227.8$ Hz), 158.8 (d, $J = 4.0$ Hz), 134.9, 133.8, 126.9 (d, $J = 11.0$ Hz), 124.2 (d, $J = 7.9$ Hz), 123.0 (d, $J = 23.7$ Hz), 122.2 (d, $J = 7.6$ Hz), 114.3 (d, $J = 9.8$ Hz), 112.5 (d, $J = 25.3$ Hz), 112.0 (d, $J = 25.9$ Hz), 107.6 (d, $J = 24.7$ Hz), 106.8 (d, $J = 4.3$ Hz). HRESIMS calcd for [C$_{16}$H$_{8}$F$_2$N$_2$O + H]$^+$ 283.06714, found 283.06625.

5,5′-Dibromo-2,3′-bi(3H-indol)-3-one (9c)
Red solid, mp: 157-159 °C (from EtOAc/PE = 1:4). IR (KBr) νmax: 3409, 1728, 1560, 1440, 1351, 1286, 1104, 1019 cm⁻¹. ¹H NMR (400 MHz, DMSO-d₆): δ 12.32 (s, 1H, NH), 8.46 (s, 2H, Ar-H), 7.70 (dd, J = 8.0, 1.6 Hz, 1H, Ar-H), 7.59 (s, 1H, Ar-H), 7.49 (d, J = 8.5 Hz, 1H, Ar-H), 7.38 (dd, J = 8.5, 1.6 Hz, 1H, Ar-H), 7.32 (d, J = 8.0 Hz, 1H, Ar-H).

¹³C NMR (100 MHz, DMSO-d₆): δ 193.3, 161.4, 157.7, 139.0, 135.6, 134.5, 127.6, 126.9, 126.0, 124.4, 124.3, 122.5, 118.4, 114.6, 114.5, 105.8. HRESIMS calcd for [C₁₆H₈Br₂N₂O + H]⁺ 402.90816 (51%), 404.90612 (100%), found 402.90646 (51%), 404.90418 (100%).

5,5'-Dimethyl-2,3'-bi(3H-indol)-3-one (9d)

Red amorphous solid. IR (KBr) νmax: 3415, 1719, 1611, 1559, 1473, 1354, 1207, 1105 cm⁻¹. ¹H NMR (400 MHz, DMSO-d₆): δ 11.99 (s, 1H, NH), 8.41 (s, 1H, Ar-H), 8.17 (s, 1H, Ar-H), 7.39 (d, J = 8.2 Hz, 1H, Ar-H), 7.31 (d, J = 7.5 Hz, 1H, Ar-H), 7.28 (s, 1H, Ar-H), 7.21 (d, J = 7.5 Hz, 1H, Ar-H), 7.07 (d, J = 8.2 Hz, 1H, Ar-H), 2.43 (s, 3H, CH₃), 2.27 (s, 3H, CH₃).

¹³C NMR (100 MHz, DMSO-d₆): δ 196.0, 161.3, 158.0, 137.7, 136.2, 135.5, 133.3, 131.0, 125.7, 125.4, 125.3, 123.0, 122.6, 120.7, 112.6, 106.6, 21.8, 20.9. HRESIMS calcd for [C₁₈H₁₄N₂O + H]⁺ 275.11844, found 275.11810.

5,5'-Dimethoxy-2,3'-bi(3H-indol)-3-one (9e)

Red solid, mp: 198-199 °C (from EtOAc/PE = 1:4). IR (KBr) νmax: 3429, 1724, 1619, 1557, 1477, 1360, 1216, 1135, 1029 cm⁻¹. ¹H NMR (400 MHz, DMSO-d₆): δ 11.93 (s, 1H, NH), 8.37 (d, J = 3.0 Hz, 1H, Ar-H), 7.87 (d, J = 2.5 Hz, 1H, Ar-H), 7.41 (d, J = 8.8 Hz, 1H, Ar-H), 7.24 (d, J = 7.9 Hz, 1H, Ar-H), 7.06 (s, 1H, Ar-H), 7.05 (dd, J = 3.0, 7.9 Hz, 1H, Ar-H), 6.89 (dd, J = 8.8, 2.5 Hz, 1H, Ar-H), 3.81 (s, 3H, OCH₃), 3.78 (s, 3H, OCH₃).

¹³C NMR (100 MHz, DMSO-d₆): δ 195.9, 158.6, 157.7, 156.3, 155.8, 133.0, 132.0, 127.1, 123.9, 121.7, 121.4, 113.6, 113.2, 111.2, 106.8, 105.2, 56.3, 55.9. HRESIMS calcd for [C₁₃H₁₄N₂O₃ + H]⁺ 307.10827, found 307.10690.

5,5'-Bis(benzyloxy)-2,3'-bi(3H-indol)-3-one (9f)
Red solid, mp: 167-169 °C (from EtOAc/PE = 1:3). IR (KBr) \( v_{\text{max}} \): 3419, 1719, 1617, 1562, 1470, 1382, 1270, 1126, 1018 cm\(^{-1}\). \(^1\)H NMR (400 MHz, DMSO-\(d_6\)): \( \delta \) 11.96 (s, 1H, NH), 8.37 (d, \( J = 2.6 \) Hz, 1H, Ar-H), 7.98 (d, \( J = 2.3 \) Hz, 1H, Ar-H), 7.51 (d, \( J = 7.4 \) Hz, 2H, Ar-H), 7.46-7.38 (m, 7H, Ar-H), 7.35-7.30 (m, 2H, Ar-H), 7.26 (d, \( J = 8.9 \) Hz, 1H, Ar-H), 7.16 (s, 1H, Ar-H), 7.14 (d, \( J = 8.9 \) Hz, 1H, Ar-H), 6.98 (dd, \( J = 8.9, 2.3 \) Hz, 1H, Ar-H), 5.14 (s, 4H, OCH\(_2\)). \(^{13}\)C NMR (100 MHz, DMSO-\(d_6\)): \( \delta \) 195.8, 157.8, 157.6, 156.6, 154.8, 137.9, 137.3, 133.1, 132.2, 128.9, 128.8, 128.4, 128.3, 128.2, 128.1, 127.1, 123.8, 122.6, 121.7, 113.6, 113.5, 112.0, 107.0, 106.8, 70.5, 70.4. HRESIMS calcd for [C\(_{30}\)H\(_{22}\)N\(_2\)O\(_3\) + H]\(^+\) 459.17087, found 459.17087.

6,6’-Difluoro-2,3’-bi(3\(H\)-indol)-3-one (9g)

Red solid, mp: 203-205 °C (from EtOAc/PE = 1:4). IR (KBr) \( v_{\text{max}} \): 3381, 1717, 1615, 1563, 1422, 1366, 1232, 1122, 1084 cm\(^{-1}\). \(^1\)H NMR (400 MHz, DMSO-\(d_6\)): \( \delta \) 12.24 (s, 1H, NH), 8.50 (s, 1H, Ar-H), 8.31 (t, \( J = 6.2 \) Hz, 1H, Ar-H), 7.55 (t, \( J = 6.2 \) Hz, 1H, Ar-H), 7.32 (d, \( J = 8.9 \) Hz, 1H, Ar-H), 7.18 (d, \( J = 8.9 \) Hz, 1H, Ar-H), 7.10 (t, \( J = 8.4 \) Hz, 1H, Ar-H), 6.95 (t, \( J = 8.4 \) Hz, 1H, Ar-H). \(^{13}\)C NMR (100 MHz, DMSO-\(d_6\)): \( \delta \) 193.3, 168.4 (d, \( J = 254.3 \) Hz), 166.2 (d, \( J = 13.6 \) Hz), 160.2 (d, \( J = 237.9 \) Hz), 159.8, 137.5 (d, \( J = 12.8 \) Hz), 135.2, 126.9 (d, \( J = 11.5 \) Hz), 124.0 (d, \( J = 10.0 \) Hz), 123.0, 119.5 (d, \( J = 2.5 \) Hz), 112.8 (d, \( J = 23.6 \) Hz), 110.6 (d, \( J = 24.0 \) Hz), 109.5 (d, \( J = 25.1 \) Hz), 106.8, 99.5 (d, \( J = 26.0 \) Hz). HRESIMS calcd for [C\(_{16}\)H\(_8\)F\(_2\)N\(_2\)O\(_2\) + H]\(^+\) 283.06829, found 283.06872.

6,6’-Dichloro-2,3’-bi(3\(H\)-indol)-3-one (9h)

Red solid, mp: 210-212 °C (from EtOAc/PE = 1:4). IR (KBr) \( v_{\text{max}} \): 3397, 1717, 1559, 1406, 1356, 1256, 1128, 1057 cm\(^{-1}\). \(^1\)H NMR (400 MHz, DMSO-\(d_6\)): \( \delta \) 12.25 (s, 1H, NH), 8.49 (s, 1H, Ar-H), 8.29 (d, \( J = 8.4 \) Hz, 1H, Ar-H), 7.55 (s, 1H, Ar-H), 7.47 (d, \( J = 7.6 \) Hz, 1H, Ar-H), 7.36 (s, 1H, Ar-H), 7.26 (d, \( J = 8.4 \) Hz, 1H, Ar-H), 7.19 (d, \( J = 7.6 \) Hz, 1H, Ar-H). \(^{13}\)C NMR (100 MHz, DMSO-\(d_6\)): \( \delta \) 193.6, 164.6, 159.5, 142.0, 137.8, 135.3, 128.6, 126.5, 125.9, 125.1, 124.1, 122.6, 121.7, 121.4, 112.9, 106.9. HRESIMS calcd for [C\(_{16}\)H\(_8\)Cl\(_2\)N\(_2\)O + H]\(^+\) 315.00919 (100%), 317.00624 (64%), found 315.00980 (100%), 317.00684 (64%).

6,6’-Dimethyl-2,3’-bi(3\(H\)-indol)-3-one (9i)
Red amorphous solid. IR (KBr) $\nu_{\text{max}}$: 3421, 1721, 1611, 1415, 1367, 1275, 1122 cm$^{-1}$. $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 12.03 (s, 1H, NH), 8.43 (s, 1H, Ar-H), 8.23 (d, $J = 8.0$ Hz, 1H, Ar-H), 7.38 (d, $J = 7.3$ Hz, 1H, Ar-H), 7.31 (s, 1H, Ar-H), 7.15 (s, 1H, Ar-H), 7.08 (d, $J = 8.0$ Hz, 1H, Ar-H), 6.96 (d, $J = 7.3$ Hz, 1H, Ar-H), 2.45 (s, 3H, CH$_3$), 2.42 (s, 3H, CH$_3$). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 195.0, 163.9, 158.9, 149.1, 137.7, 133.6, 133.4, 126.8, 124.8, 124.2, 123.9, 122.6, 121.9, 120.6, 112.9, 107.1, 22.4, 21.8. HRESIMS calcd for [C$_{18}$H$_{14}$N$_2$O$^+$ + H]$^+$ 275.11844, found 275.11914.

7,7'-Dichloro-2,3'-bi(3H-indol)-3-one (9j)

Red solid, mp: 222-224 °C (from EtOAc/PE = 1:4). IR (KBr) $\nu_{\text{max}}$: 3417, 1719, 1566, 1412, 1369, 1263, 1125, 1013 cm$^{-1}$. $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 12.61 (s, 1H, NH), 8.42 (s, 1H, Ar-H), 8.35 (d, $J = 7.8$ Hz, 1H, Ar-H), 7.58 (d, $J = 7.8$ Hz, 1H, Ar-H), 7.44 (d, $J = 7.2$ Hz, 1H, Ar-H), 7.35 (d, $J = 7.6$ Hz, 1H, Ar-H), 7.27 (t, $J = 7.8$ Hz, 1H, Ar-H), 7.17 (t, $J = 7.2$ Hz, 1H, Ar-H). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 194.0, 158.7, 137.5, 134.6, 134.2, 128.4, 128.3, 128.2, 125.5, 125.1, 123.7, 123.6. 123.5, 121.9, 117.4, 108.0. HRESIMS calcd for [C$_{16}$H$_{8}$Cl$_2$N$_2$O$^+$ + H]$^+$ 315.00919 (100%), 317.00624 (64%), found 315.01022 (100%), 317.00720 (64%).

7,7'-Dibromo-2,3'-bi(3H-indol)-3-one (9k)

Red solid, mp: 224-226 °C (from EtOAc/PE = 1:4). IR (KBr) $\nu_{\text{max}}$: 3418, 1717, 1563, 1430, 1368, 1261, 1114, 1048 cm$^{-1}$. $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 12.44 (s, 1H, NH), 8.42 (s, 1H, Ar-H), 8.40 (d, $J = 7.0$ Hz, 1H, Ar-H), 7.71 (d, $J = 7.8$ Hz, 1H, Ar-H), 7.49 (d, $J = 7.6$ Hz, 1H, Ar-H), 7.46 (d, $J = 7.0$ Hz, 1H, Ar-H), 7.21 (t, $J = 7.8$ Hz, 1H, Ar-H), 7.09 (t, $J = 7.6$ Hz, 1H, Ar-H). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 194.3, 160.4, 158.8, 140.2, 135.8, 134.6, 128.7, 128.0, 126.8, 125.2, 123.9, 123.8, 122.4, 115.3, 108.0, 105.6. HRESIMS calcd for [C$_{16}$H$_{8}$Br$_2$N$_2$O$^+$ + H]$^+$ 402.90816 (51%), 404.90612 (100%), found 402.90848 (51%), 404.90631 (100%).

7,7'-Dimethyl-2,3'-bi(3H-indol)-3-one (9l)

Red solid, mp: 208-209 °C (from EtOAc/PE = 1:4). IR (KBr) $\nu_{\text{max}}$: 3378, 1707, 1623, 1567, 1441, 1373, 1241, 1129 cm$^{-1}$. $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 12.12 (s, 1H, NH), 8.41 (d, $J = 3.0$ Hz, 1H, Ar-H), 8.39 (d, $J = 7.8$ Hz, 1H, Ar-H), 7.71 (d, $J = 7.8$ Hz, 1H, Ar-H), 7.49 (d, $J = 7.6$ Hz, 1H, Ar-H), 7.46 (d, $J = 7.0$ Hz, 1H, Ar-H), 7.21 (t, $J = 7.8$ Hz, 1H, Ar-H), 7.09 (t, $J = 7.6$ Hz, 1H, Ar-H). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 194.3, 160.4, 158.8, 140.2, 135.8, 134.6, 128.7, 128.0, 126.8, 125.2, 123.9, 123.8, 122.4, 115.3, 108.0, 105.6. HRESIMS calcd for [C$_{16}$H$_{8}$Br$_2$N$_2$O$^+$ + H]$^+$ 402.90816 (51%), 404.90612 (100%), found 402.90848 (51%), 404.90631 (100%).
1H, Ar-H), 8.28 (d, J = 7.6 Hz, 1H, Ar-H), 7.37 (d, J = 7.6 Hz, 1H, Ar-H), 7.30 (d, J = 6.9 Hz, 1H, Ar-H), 7.16 (t, J = 7.6 Hz, 1H, Ar-H), 7.06 (d, J = 7.6 Hz, 1H, Ar-H), 7.04 (t, J = 6.9 Hz, 1H, Ar-H), 2.51 (s, 3H, CH$_3$), 2.42 (s, 3H, CH$_3$).

$^{13}$C NMR (100 MHz, DMSO-$d_6$): δ 196.2, 161.1, 157.6, 139.1, 136.7, 132.8, 130.3, 126.4, 126.3, 124.5, 122.6, 122.4, 122.3, 122.2, 120.7, 107.6, 17.1, 15.3. HRESIMS calcd for [C$_{18}$H$_{14}$N$_2$O$+\text{H}$]$^+$ 275.11844, found 275.11698.

7,7'-Dimethoxy-2,3'-bi(3H-indol)-3-one (9m)

Red solid, mp: 179-181 °C (from EtOAc/PE = 1:4). IR (KBr) $\nu_{\text{max}}$: 3348, 1726, 1622, 1561, 1442, 1371, 1263, 1129, 1069 cm$^{-1}$. $^1$H NMR (400 MHz, DMSO-$d_6$): δ 12.29 (s, 1H, NH), 8.29 (s, 1H, Ar-H), 7.95 (d, J = 7.9 Hz, 1H, Ar-H), 7.25 (dt, J = 3.5, 2.3 Hz, 1H, Ar-H), 7.18 (t, J = 7.9 Hz, 1H, Ar-H), 7.12 (d, J = 2.3 Hz, 1H, Ar-H), 7.11 (t, J = 3.5 Hz, Ar-H), 6.84 (d, J = 7.9 Hz, 1H, Ar-H), 4.02 (s, 3H, OCH$_3$), 3.95 (s, 3H, OCH$_3$).

$^{13}$C NMR (100 MHz, DMSO-$d_6$): δ 196.5, 156.6, 150.9, 149.6, 146.9, 132.0, 128.2, 127.9, 127.1, 124.5, 124.0, 123.0 117.4, 115.5, 107.7, 104.8, 57.5, 55.8. HRESIMS calcd for [C$_{18}$H$_{14}$N$_2$O$+\text{H}$]$^+$ 307.10827, found 307.10628.

7,7'-Bis(benzyloxy)-2,3'-bi(3H-indol)-3-one (9n)

Red solid, mp: 183-185 °C (from EtOAc/PE = 1:4). IR (KBr) $\nu_{\text{max}}$: 3438, 1709, 1569, 1429, 1376, 1249, 1116, 1089 cm$^{-1}$. $^1$H NMR (400 MHz, DMSO-$d_6$): δ 12.31 (s, 1H, NH), 8.32 (s, 1H, Ar-H), 7.94 (d, J = 7.7 Hz, 1H, Ar-H), 7.57 (d, J = 6.9 Hz, 2H, Ar-H), 7.52 (d, J = 6.9 Hz, 2H, Ar-H), 7.44-7.30 (m, 6H, Ar-H), 7.27 (d, J = 7.7 Hz, 1H, Ar-H), 7.14 (d, J = 6.0 Hz, 2H, Ar-H), 7.09 (d, J = 7.4 Hz, 1H, Ar-H), 6.94 (d, J = 7.4 Hz, 1H, Ar-H), 5.51 (s, 2H, OCH$_2$), 5.29 (s, 2H, OCH$_2$).

$^{13}$C NMR (100 MHz, DMSO-$d_6$): δ 195.3, 156.6, 149.9, 149.8, 145.9, 137.8, 137.5, 132.3, 129.0, 128.9, 128.5, 128.4, 128.3, 128.2, 128.1, 128.0, 127.3, 126.6, 124.9, 123.0, 118.2, 115.6, 107.7, 106.1, 72.0, 69.9. HRESIMS calcd for [C$_{30}$H$_{22}$N$_2$O$_3$+\text{H}$]$^+$ 459.17087, found 459.17139.

3-Oxo-2,3'-bi(3H-indole)-4,4'-dicarbonitrile (9o)

Brown amorphous solid. IR (KBr) $\nu_{\text{max}}$: 3413, 1729, 1610, 1561, 1421, 1356, 1251, 1129, 1038 cm$^{-1}$. $^1$H NMR (400 MHz, DMSO-$d_6$): δ 12.73 (s, 1H, NH), 8.61 (s, 1H, Ar-H), 7.89 (dd, J = 7.9, 1.0 Hz, 1H, Ar-H), 7.76 (t, J = 7.7 Hz, 1H, Ar-H), 7.73 (d, J = 7.3 Hz, 1H, Ar-H), 7.63 (d, J = 7.3 Hz, 1H, Ar-H), 7.60 (d, J = 7.7 Hz, 1H, Ar-H), 7.43 (t, J = 7.9 Hz, 1H, Ar-H).

$^{13}$C NMR (100 MHz, DMSO-$d_6$): δ 191.0, 162.6, 158.0, 138.2, 138.0, 136.7, 130.6, 130.4, 125.8, 124.6, 124.0,
123.8, 119.6, 118.5, 115.6, 106.8, 106.4, 104.8. MS (ESI): 297 (M+H+, 100), 319 (M+Na+, 15).
Anal calcd for C_{18}H_{8}N_{4}O: C, 72.97; H, 2.72; N, 18.91. Found C, 72.75; H, 3.02; N, 18.69.
Dimethyl 3-oxo-2,3’-bi(3H-indole)-6,6’-dicarboxylate (9p)

Red solid, mp: 177-179 °C (from EtOAc/PE = 1:1). IR (KBr) ν_{max}: 3306, 1728, 1561, 1428, 1355, 1279, 1139, 1022 cm^{-1}. ¹H NMR (400 MHz, DMSO-\(d_6\)): δ 12.43 (s, 1H, NH), 8.35 (d, J = 3.0 Hz, 1H, Ar-H), 7.72 (d, J = 8.0 Hz, 1H, Ar-H), 7.68 (d, J = 7.7 Hz, 1H, Ar-H), 7.50 (d, J = 7.4 Hz, 1H, Ar-H), 7.45 (d, J = 7.4 Hz, 2H, Ar-H), 7.33 (t, J = 7.7 Hz, 1H, Ar-H), 3.87 (s, 3H, OCH₃), 2.72 (s, 3H, OCH₃).
¹³C NMR (100 MHz, DMSO-\(d_6\)): δ 191.5, 169.3, 166.0, 162.9, 159.6, 138.1, 137.6, 134.6, 130.0, 126.9, 126.6, 124.1, 123.0, 122.8, 122.7, 120.4, 116.2, 106.9, 52.9, 52.3. HRESIMS calcd for [C_{20}H_{14}N_{2}O_{5} - H]⁺ 361.08245, found 361.08298.

7-Methyl-2-(7-methyl-1H-indol-3-yl)indolin-3-one (13)

White amorphous solid. IR (KBr) ν_{max}: 3375, 1716, 1620, 1569, 1236, 1125 cm^{-1}. ¹H NMR (400 MHz, DMSO-\(d_6\)): δ 12.13 (s, 1H, NH), 7.72 (s, 1H, Ar-H), 7.60 (s, 1H, Ar-H), 7.21 (s, 1H, Ar-H), 7.12 (d, J = 6.3 Hz, 1H, Ar-H), 6.99 (s, 1H, Ar-H), 6.81 (s, 1H, Ar-H), 6.60 (d, J = 6.3 Hz, 1H, Ar-H), 5.93 (s, 2H), 3.94 (s, 3H, CH₃), 3.83 (s, 3H, CH₃). ¹³C NMR (100 MHz, DMSO-\(d_6\)): δ 192.1, 147.4, 146.7, 139.6, 133.9, 128.4, 127.0, 123.7, 122.7, 121.4, 117.2, 114.6, 114.5, 114.4, 113.0, 104.0, 56.2, 55.8. MS (ESI): 277 (M+H+, 100). Anal calcd for C_{18}H_{16}N_{2}O_{1}: C, 78.24; H, 5.84; N, 10.14. Found C, 77.89; H, 6.07; N, 9.83.

3. Copies of ¹H, ¹³C Spectra
$^1$H and $^{13}$C NMR Spectra for 9a

$^1$H and $^{13}$C NMR Spectra for 9b
$^1$H and $^{13}$C NMR Spectra for 9c
$^1$H and $^{13}$C NMR Spectra for 9d
$^{1}$H and $^{13}$C NMR Spectra for 9e
$^1$H and $^{13}$C NMR Spectra for 9f
$^1$H and $^{13}$C NMR Spectra for 9g
$^1$H and $^{13}$C NMR Spectra for 9h
$^1$H and $^{13}$C NMR Spectra for 9i
\(^1\text{H} \text{ and } ^{13}\text{C} \text{ NMR Spectra for 9j}\)
$^{1}$H and $^{13}$C NMR Spectra for 9k
$^1$H and $^{13}$C NMR Spectra for 91
$^1$H and $^{13}$C NMR Spectra for 9m
$^{1}H$ and $^{13}C$ NMR Spectra for 9n
$^1$H and $^{13}$C NMR Spectra for 9o
$^1$H and $^{13}$C NMR Spectra for 9p
$^1$H and $^{13}$C NMR Spectra for 13
4. Copies of HRESIMS Spectra
2,3'-Bi(3H-indol)-3-one (9a)

5,5'-Difluoro-2,3'-bi(3H-indol)-3-one (9b)

5,5'-Dibromo-2,3'-bi(3H-indol)-3-one (9c)

5,5'-Dimethyl-2,3'-bi(3H-indol)-3-one (9d)
5,5'-Dimethoxy-2,3'-bi(3H-indol)-3-one (9e)

5,5'-Bis(benzyloxy)-2,3'-bi(3H-indol)-3-one (9f)

6,6'-Difluoro-2,3'-bi(3H-indol)-3-one (9g)
6,6'-Dichloro-2,3'-bi(3H-indol)-3-one (9h)

6,6'-Dimethyl-2,3'-bi(3H-indol)-3-one (9i)

7,7'-Dichloro-2,3'-bi(3H-indol)-3-one (9j)
7,7'-Dibromo-2,3'-bi(3H-indol)-3-one (9k)

7,7'-Dimethyl-2,3'-bi(3H-indol)-3-one (9l)

7,7'-Dimethoxy-2,3'-bi(3H-indol)-3-one (9m)
5. X-ray Data of Compound 9a
Figure 1. ORTEP representation of the molecular structure of 9a.
The data have been assigned the following deposition numbers, CCDC 1453369.