Ultrasound assisted synthesis of diversely functionalized tetrahydro-1H-indol-4(5H)-one using Brønsted base silica sodium carbonate (SSC) as a catalyst under solvent-free condition.

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Preparation of SSC

Step-1

To a 250mL round bottom flask equipped with a reflux condenser, in which the hose interface was connected to a pot of water, 10 g of silica gel 60 (63–200 μm) that was previously dried at 120 °C for 6 h was added. Then 40 mL of thionyl chloride was added dropwise to this flask which was perched in an ice bath (caution). After the addition of thionyl chloride, the reaction mixture was removed from the ice bath and stirred for 0.5 h in room temperature and 48 h under reflux conditions. Afterwards, the reaction mixture was filtrated to obtain silica chloride.

Step-2

To a stirred 250 mL round bottom flask containing 10 g of sodium bicarbonate and 25 mL of n-hexane under reflux conditions, 10 g of silica chloride (after drying at 120 °C for 6 h) was added. After 24 h, the reaction mixture was filtrated to separate the catalyst and the solid product was washed with 50 mL of distilled water ten times, using 5 mL each time until filtrate became quite neutral, in order to remove the remaining sodium bicarbonate. Finally, dryid at 100 °C for 12 h, to afford SSC

General Procedure for the Synthesis of tetrahydro-1H-indol-4(5H)-one at Ultrasound (4a-4ac)

To a 10 mL vessel was added nitroolefins (**1**, 1 mmol), cyclic 1,3-dicarbonyl (**2**, 1mmol), amine (**3**, 1 mmol) and SSC (4 mol%), the reaction was carried out under ultrasound (20 KHz). The resulting solution was syndicated with an US probe. The reactions were followed by thin layer chromatography (TLC) using hexane/ethyl acetate as an eluent. After completion of the reaction, the mixture was washed with ethyl acetate and filtered to recover the catalyst. The filtrate was evaporated, and the crude product was purified by flash column chromatography on silica gel (200–300mesh) with ethyl acetate and hexane as eluent to afford the product **4**. The SSC catalyst was reused by the way of addition of ethyl acetate to the reaction mixture and filtration followed by drying in a vacuum oven every time.

Spectral data of the compounds

3-(4-fluorophenyl)-6,7-dihydro-1-(3-methoxyphenyl)-6,6-dimethyl-1H-indol-4(5H)-one (**4a**)

Yield 89 %; off-white solid; Mp: 130-132; ^1H NMR (400 MHz, CDCl_3): δ = 1.10 (s, 6H, 2 CH_3), 2.42 (s, 2H, CH_2), 2.67 (s, 2H, CH_2), 3.86 (s, 3H, OCH_3), 6.84 (s, 1H, ArH), 6.87 (s, 1H, ArH), 6.92 (d, J =7.7 Hz, 1H, ArH), 6.97 (d, J =8.4 Hz, 1H, ArH), 7.03 (t, J =8.6 Hz, 2H, ArH), 7.41 (t, J =8.1 Hz, 1H, ArH), 7.63-7.66 (m, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 28.53, 35.44, 37.53, 53.23, 55.70, 111.50, 113.51, 114.72, 114.93, 117.18, 117.55, 121.79, 124.31, 130.37, 130.45, 139.58, 143.51, 160.84, 163.31 (C-F, d, J = 246 Hz), 193.84; HRMS (ESI, m/z): calcd for $\text{C}_{23}\text{H}_{22}\text{FNO}_2$ ($\text{M}+\text{H}^+$) 363.1634, found: 363.1633.

3-(4-fluorophenyl)-6,7-dihydro-6,6-dimethyl-1-*p*-tolyl-1H-indol-4(5H)-one (**4b**)

Yield 88 %; yellow solid; Mp: 168-170; ^1H NMR (400 MHz, CDCl_3): δ = 1.09 (s, 6H, 2 CH_3), 2.41 (s, 2H, CH_2), 2.43 (s, 3H, PhCH_3), 2.63 (s, 2H, CH_2), 6.81 (s, 1H, ArH), 7.02 (t, J =8.8 Hz, 2H, ArH), 7.22 (d, J =8.4 Hz, 2H, ArH), 7.30 (d, J =8.1 Hz, 2H, ArH), 7.62-7.67 (m, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 21.21, 28.51, 35.40, 37.43, 53.22, 114.67, 114.88, 116.96, 121.91, 124.14, 125.19, 130.24, 130.34, 130.42, 135.98, 138.31, 143.63, 160.83, 163.26, (C-F, d, J = 243 Hz) 193.82; HRMS (ESI, m/z): calcd for $\text{C}_{23}\text{H}_{22}\text{FNO}$ ($\text{M}+\text{H}^+$) 347.1685, found: 347.1686

3-(4-fluorophenyl)-6,7-dihydro-6,6-dimethyl-1-m-tolyl-1H-indol-4(5H)-one (4c)

4c

Yield 88 %; yellow solid; Mp: 138-140; ^1H NMR (400 MHz, CDCl_3): δ = 1.09 (s, 6H, 2 CH_3), 2.41 (s, 2H, CH_2), 2.42 (s, 3H, PhCH_3), 2.65 (s, 2H, CH_2), 6.82 (s, 1H, ArH), 7.02 (t, J =8.8 Hz, 2H, ArH), 7.13 (d, J =8.1 Hz, 1H, ArH), 7.15 (s, 1H, ArH), 7.24 (d, J =7.7 Hz, 1H, ArH), 7.38 (t, J =7.7 Hz, 1H, ArH), 7.63-7.67 (m, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 21.48, 28.48, 35.40, 37.44, 53.21, 114.64, 114.85, 117.02, 121.87, 122.36, 124.15, 125.94, 128.96, 129.41, 130.13, 130.33, 130.41, 138.41, 139.89, 143.52, 160.80, 163.23, (C-F, d, J = 243 Hz) 193.82; HRMS (ESI, m/z): calcd for $\text{C}_{23}\text{H}_{22}\text{FNO}$ ($\text{M}+\text{H}^+$) 347.1685, found: 347.1685.

3-(4-fluorophenyl)-6,7-dihydro-6,6-dimethyl-1-o-tolyl-1H-indol-4(5H)-one (4d)

4d

Yield 87 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.09 (s, 6H, 2 CH_3), 2.11 (s, 3H, PhCH_3), 2.24 (d, J =16.5 Hz, 1H, CH_2), 2.41 (d, J =9.2 Hz, 2H, CH_2), 2.47 (d, J =16.5 Hz, 1H, CH_2), 6.69 (s, 1H, ArH), 7.02 (t, J =8.8 Hz, 2H, ArH), 7.21 (d, J =7.3 Hz, 1H, ArH), 7.32 (t, J =7.7 Hz, 1H, ArH), 7.36-7.40 (m, 2H, ArH), 7.66-7.70 (m, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 17.48, 27.92, 29.00, 35.31, 36.61, 53.31, 114.66, 114.86, 121.81, 123.93, 127.06, 127.74, 129.43, 130.27, 130.35, 131.28, 135.53, 137.38, 144.64, 160.76, 163.20, (C-F, d, J = 244 Hz) 193.66; HRMS (ESI, m/z): calcd for $\text{C}_{23}\text{H}_{22}\text{FNO}$ ($\text{M}+\text{H}^+$) 347.1685, found: 347.1687.

6,7-dihydro-3-(3,4-dimethoxyphenyl)-6,6-dimethyl-1-o-tolyl-1H-indol-4(5H)-one (4e)

4e

Yield 88 %; yellow solid; Mp: 138-140; ^1H NMR (400 MHz, CDCl_3): δ = 1.09 (s, 6H, 2 CH_3), 2.12 (s, 3H, PhCH_3), 2.25 (d, J =16.5 Hz, 1H, CH_2), 2.40 (d, J =16.1 Hz, 1H, CH_2), 2.46 (d, J =18.7 Hz, 2H, CH_2), 3.89 (s, 3H, OCH_3), 3.96 (s, 3H, OCH_3), 6.75 (s, 1H, ArH), 6.86 (d, J =8.4 Hz, 1H, ArH), 7.23 (d, J =8.4 Hz, 2H, ArH), 7.31-7.40 (m, 3H, ArH), 7.55 (s, 1H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 17.52, 27.96, 29.01, 35.23, 36.73, 53.50, 56.03, 110.99, 112.84, 116.26, 120.46, 121.48, 124.76, 127.04, 127.80, 129.38, 131.26, 135.62, 137.53, 144.59, 148.01, 148.44, 193.71; HRMS (ESI, m/z): calcd for $\text{C}_{25}\text{H}_{27}\text{NO}_3$ ($\text{M}+\text{H}^+$) 389.1990, found: 389.1991.

6,7-dihydro-3-(3,4-dimethoxyphenyl)-6,6-dimethyl-1-m-tolyl-1H-indol-4(5H)-one (4f)

4f

Yield 88 %; yellow solid; Mp: 160-162; ^1H NMR (400 MHz, CDCl_3): δ = 1.10 (s, 6H, 2 CH_3), 2.43 (s, 2H, CH_2), 2.44 (s, 3H, PhCH_3), 2.65 (s, 2H, CH_2), 3.89 (s, 3H, OCH_3), 3.95 (s, 3H, OCH_3), 6.85-6.87 (m, 2H, ArH), 7.14 (d, J =8.1Hz, 1H, ArH), 7.16 (s, 1H, ArH), 7.20-7.26 (m, 2H, ArH), 7.38 (m, J =7.7 Hz, 1H, ArH), 7.48 (s, 1H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 21.50, 28.51, 35.33, 37.54, 53.37, 56.02, 110.97, 112.86, 117.08, 120.62, 121.58, 122.45, 124.99, 126.02, 126.96, 128.91, 129.39, 138.55, 139.86, 143.50, 148.06, 148.41, 193.88; HRMS (ESI, m/z): calcd for $\text{C}_{25}\text{H}_{27}\text{NO}_3$ ($\text{M}+\text{H}^+$) 389.1990, found: 389.1989.

6,7-dihydro-6,6-dimethyl-3-phenyl-1-p-tolyl-1H-indol-4(5H)-one (4g)

Yield 92 %; brown sticky liquid; lit^[38] ; ¹H NMR (400 MHz, CDCl₃): δ = 1.09 (s, 6H, 2CH₃), 2.41 (s, 2H, CH₂), 2.42 (s, 3H, PhCH₃), 2.63 (s, 2H, CH₂), 6.85 (s, 1H, ArH), 7.21-7.24 (m, 3H, ArH), 7.28 (s, 1H, ArH), 7.30-7.36 (m, 3H, ArH), 7.68 (d, *J*=8.4 Hz, 2H, ArH), ¹³C NMR (100 MHz, CDCl₃): δ = 21.20, 28.52, 35.37, 37.48, 53.28, 117.11, 122.08, 125.20, 126.72, 127.99, 128.80, 130.20, 134.12, 136.10, 138.19, 143.53, 193.63; HRMS (ESI, *m/z*): calcd for C₂₃H₂₃NO (M+H⁺) 329.1779, found: 329.1780.

6,7-dihydro-1-(3-methoxyphenyl)-6,6-dimethyl-3-phenyl-1H-indol-4(5H)-one (4h)

Yield 90 %; brown sticky liquid; ¹H NMR (400 MHz, CDCl₃): δ = 1.09 (s, 6H, 2CH₃), 2.41 (s, 2H, CH₂), 2.66 (s, 2H, CH₂), 3.83 (s, 3H, OCH₃), 6.86 (s, 2H, ArH), 6.90-6.96 (m, 2H, ArH), 7.23 (s, 1H, ArH), 7.32-7.40 (m, 3H, ArH), 7.68 (d, *J*=8.4 Hz, 2H, ArH), ¹³C NMR (100 MHz, CDCl₃): δ = 28.45, 35.33, 37.49, 53.21, 55.61, 111.41, 113.42, 117.48, 121.91, 125.22, 126.71, 127.95, 128.76, 130.35, 134.00, 139.59, 143.38, 160.45, 193.60; HRMS (ESI, *m/z*): calcd for C₂₃H₂₃NO₂ (M+H⁺) 345.1728, found: 345.1729.

6,7-dihydro-6,6-dimethyl-1-(3,4-dimethylphenyl)-3-phenyl-1H-indol-4(5H)-one (4i)

4i

Yield 87 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.08 (s, 6H, 2 CH_3), 2.32 (s, 3H, PhCH_3), 2.32 (s, 3H, PhCH_3), 2.41 (s, 2H, CH_2), 2.63 (s, 2H, CH_2), 6.84 (s, 1H, ArH), 7.05 (dd, J =2.20 Hz, J =6.0 Hz, 1H, ArH), 7.11 (s, 1H, ArH), 7.23-7.25 (m, 2H, ArH), 7.33 (d, J =7.7 Hz, 2H, ArH), 7.68 (d, J =8.4 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 19.53, 20.00, 28.49, 35.36, 37.42, 53.24, 116.93, 122.11, 122.60, 124.96, 126.36, 126.64, 127.95, 128.76, 130.53, 134.14, 136.27, 136.82, 138.21, 143.54, 193.68; HRMS (ESI, m/z): calcd for $\text{C}_{24}\text{H}_{25}\text{NO}$ ($\text{M}+\text{H}^+$) 343.1936, found: 343.1937.

6,7-dihydro-6,6-dimethyl-1-(2,5-dimethylphenyl)-3-phenyl-1H-indol-4(5H)-one (4j)

4j

Yield 85 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.08 (s, 3H, CH_3), 1.09 (s, 3H, CH_3), 2.05 (s, 3H, CH_3), 2.25 (d, J =16.1 Hz, 1H, CH_2), 2.37 (s, 3H, CH_3), 2.42 (d, J =9.2 Hz, 2H, CH_2), 2.48 (d, J =16.1, 1H, CH_2), 6.71 (s, 1H, ArH), 7.04 (s, 1H, ArH), 7.18-7.25 (m, 3H, ArH), 7.32-7.36 (m, 2H, ArH), 7.72 (d, J =8.1 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 16.99, 20.86, 27.86, 29.03, 35.27, 36.16, 53.34, 116.15, 122.01, 124.74, 126.55, 127.95, 128.17, 128.66, 130.05, 130.94, 132.17, 134.15, 136.89, 137.24, 144.52, 193.50; HRMS (ESI, m/z): calcd for $\text{C}_{24}\text{H}_{25}\text{NO}$ ($\text{M}+\text{H}^+$) 343.1936, found: 343.1935.

6,7-dihydro-6,6-dimethyl-1-(3,5-dimethylphenyl)-3-phenyl-1H-indol-4(5H)-one (4k)

4k

Yield 87 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.09 (s, 6H, 2 CH_3), 2.39 (s, 6H, Ph_2CH_3), 2.42 (s, 2H, CH_2), 2.64 (s, 2H, CH_2), 6.84 (s, 1H, ArH), 6.94 (s, 2H, ArH), 7.05 (s, 1H, ArH), 7.16 (s, 1H, ArH), 7.22-7.26 (m, 1H, ArH), 7.34 (t, J =7.3 Hz, 1H, ArH), 7.68 (d, J =7.32 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 21.39, 28.51, 35.41, 37.48, 53.27, 117.03, 122.10, 123.03, 125.02, 126.67, 127.97, 128.78, 129.75, 134.15, 138.47, 139.50, 143.46, 193.72; HRMS (ESI, m/z): calcd for $\text{C}_{24}\text{H}_{25}\text{NO}$ ($\text{M}+\text{H}^+$) 343.1936, found: 343.1938.

6,7-dihydro-3-phenyl-1-o-tolyl-1H-indol-4(5H)-one (4l)

4l

Yield 92 %; brown sticky liquid; lit^[39]; ^1H NMR (400 MHz, CDCl_3): δ = 2.06-2.21 (m, 5H, PhCH_3 , CH_2), 2.39-2.46 (m, 1H, CH_2), 2.51-2.61 (m, 3H, CH_2), 6.71 (s, 1H, ArH), 7.23-7.26 (m, 2H, ArH), 7.29-7.38 (m, 5H, ArH), 7.69 (d, J =8.1 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 17.47, 22.74, 23.69, 39.22, 117.45, 121.91, 125.06, 126.65, 126.99, 127.66, 127.96, 128.79, 129.30, 131.25, 134.22, 135.54, 137.46, 145.55, 194.08; HRMS (ESI, m/z): calcd for $\text{C}_{21}\text{H}_{19}\text{NO}$ ($\text{M}+\text{H}^+$) 301.1466, found: 301.1468.

6,7-dihydro-6,6-dimethyl-3-phenyl-1-m-tolyl-1H-indol-4(5H)-one (4m)

4m

Yield 87 %; brown sticky liquid; lit; ^1H NMR (400 MHz, CDCl_3): δ = 1.08 (s, 6H, 2 CH_3), 2.41 (s, 2H, CH_2), 2.42 (s, 3H, PhCH_3), 2.64 (s, 2H, CH_2), 6.85 (s, 1H, ArH), 7.12 (d, J =7.7 Hz, 1H, ArH), 7.14 (s, 1H, ArH), 7.22 (d, J =7.3 Hz, 2H, ArH), 7.31-7.36 (m, 3H, ArH), 7.69 (d, J =7.7 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 21.43, 28.44, 35.34, 37.42, 53.20, 117.06, 121.98, 122.30, 125.09, 125.87, 126.63, 127.92, 128.73, 128.82, 129.33, 134.04, 138.43, 139.77, 143.41, 193.65; HRMS (ESI, m/z): calcd for $\text{C}_{23}\text{H}_{23}\text{NO}$ ($\text{M}+\text{H}^+$) 329.1779, found: 329.1780.

6,7-dihydro-6,6-dimethyl-3-phenyl-1-o-tolyl-1H-indol-4(5H)-one (4n)

4n

Yield 89 %; brown sticky liquid^[39]; ^1H NMR (400 MHz, CDCl_3): δ = 1.08 (s, 6H, 2 CH_3), 2.10 (s, 3H, PhCH_3), 2.24 (d, J =16.5 Hz, 1H, CH_2), 2.41 (d, J =9.5 Hz, 2H, CH_2), 2.47 (d, J =16.5 Hz, 1H, CH_2), 6.72 (s, 1H, ArH), 7.18-7.24 (m, 2H, ArH), 7.28-7.38 (m, 5H, ArH), 7.72 (d, J =7.3 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 17.40, 27.83, 28.95, 35.21, 36.56, 53.29, 116.22, 121.94, 124.81, 126.54, 126.96, 127.67, 127.91, 128.63, 129.29, 131.18, 134.06, 135.46, 137.37, 144.49, 193.43; HRMS (ESI, m/z): calcd for $\text{C}_{23}\text{H}_{23}\text{NO}$ ($\text{M}+\text{H}^+$) 329.1779, found: 329.1779.

6,7-dihydro-3-phenyl-1-m-tolyl-1H-indol-4(5H)-one (4o)

4o

Yield 87 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 2.06-2.12 (m, 2H, CH_2), 2.41 (s, 3H, PhCH_2), 2.52 (t, J =5.9 Hz, 2H, CH_2), 2.77 (t, J =6.2 Hz, 2H, CH_2), 6.84 (s, 1H, ArH), 7.13 (d, J =8.4 Hz, 1H, ArH), 7.15 (s, 1H, ArH), 7.21 (d, J =7.0 Hz, 1H, ArH), 7.25 (d, J =7.3 Hz, 1H, ArH), 7.33 (d, J =7.3 Hz, 3H, ArH), 7.66 (d, J =7.7 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 21.43, 23.64, 23.79, 39.10, 118.14, 121.75, 122.22, 125.33, 125.82, 126.68, 127.91, 128.81, 129.34, 134.14, 138.44, 139.75, 144.58, 194.22; HRMS (ESI, m/z): calcd for $\text{C}_{21}\text{H}_{19}\text{NO}$ ($\text{M}+\text{H}^+$) 301.1466, found: 301.1467

6,7-dihydro-3-phenyl-1-p-tolyl-1H-indol-4(5H)-one (4p)

4p

Yield 86 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 2.09-2.14 (m, 2H, CH_2), 2.42 (s, 3H, PhCH_2), 2.53 (t, J =6.2 Hz, 2H, CH_2), 2.77 (t, J =6.2 Hz, 2H, CH_2), 6.83 (s, 1H, ArH), 7.21-7.25 (m, 3H, ArH), 7.28 (d, J =8.1 Hz, 2H, ArH), 7.34 (t, J =7.3 Hz, 2H, ArH), 7.66 (d, J =7.3 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 21.20, 23.63, 23.83, 39.17, 118.15, 121.85, 125.09, 125.35, 126.75, 127.98, 128.86, 130.19, 134.20, 136.07, 138.16, 144.65, 194.18; HRMS (ESI, m/z): calcd for $\text{C}_{21}\text{H}_{19}\text{NO}$ ($\text{M}+\text{H}^+$) 301.1466, found: 301.1468.

6,7-dihydro-1-(3,4-dimethylphenyl)-3-phenyl-1H-indol-4(5H)-one (4q)

4q

Yield 84 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 2.08-2.11 (m, 2H, CH_2), 2.31 (s, 6H, Ph_2CH_3), 2.52 (t, $J=5.9$ Hz, 2H, CH_2), 2.76 (t, $J=5.9$ Hz, 2H, CH_2), 6.82 (s, 1H, ArH), 7.06 (d, $J=8.1$ Hz, 1H, ArH), 7.11 (s, 1H, ArH), 7.21-7.26 (m, 2H, ArH), 7.33 (t, $J=7.7$ Hz, 2H, ArH), 7.66 (d, $J=7.7$ Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 19.50, 19.96, 23.60, 23.80, 39.50, 118.00, 121.87, 122.49, 125.19, 126.27, 126.65, 127.92, 128.83, 130.52, 134.23, 136.27, 136.77, 138.15, 144.65, 194.18; HRMS (ESI, m/z): calcd for $\text{C}_{22}\text{H}_{21}\text{NO}$ ($\text{M}+\text{H}^+$) 315.1623, found: 315.1622.

6,7-dihydro-1-(2,5-dimethylphenyl)-3-phenyl-1H-indol-4(5H)-one (4r)

4r

Yield 85 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 2.06 (s, 3H, PhCH_3), 2.09-2.13 (m, 2H, CH_2), 2.37 (s, 3H, PhCH_3), 2.41-2.47 (m, 1H, CH_2), 2.51-2.61 (m, 3H, CH_2), 6.70 (s, 1H, ArH), 7.06 (s, 1H, ArH), 7.18 (d, $J=8.1$ Hz, 1H, ArH), 7.23 (d, $J=8.8$ Hz, 2H, ArH), 7.34 (t, $J=7.3$ Hz, 2H, ArH), 7.69 (d, $J=8.1$ Hz, 2H, ArH) ^{13}C NMR (100 MHz, CDCl_3): δ = 17.01, 20.86, 22.78, 23.71, 39.25, 117.38, 121.94, 124.98, 126.62, 127.95, 128.14, 128.79, 130.02, 131.00, 132.19, 134.29, 136.91, 137.98, 145.56, 194.08; HRMS (ESI, m/z): calcd for $\text{C}_{22}\text{H}_{21}\text{NO}$ ($\text{M}+\text{H}^+$) 315.1623, found: 315.1624.

6,7-dihydro-1,3-bis(4-methoxyphenyl)-6,6-dimethyl-1H-indol-4(5H)-one (4s)

Yield 84 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.09 (s, 6H, 2 CH_3), 2.41 (s, 2H, CH_2), 2.59 (s, 2H, CH_2), 3.81 (s, 3H, OCH_3), 3.86 (s, 3H, OCH_3), 6.78 (s, 1H, ArH), 6.90 (d, J =8.8 Hz, 2H, ArH), 6.99 (d, J =8.8 Hz, 2H, ArH), 7.25 (d, J =8.8 Hz, 2H, ArH), 7.63 (d, J =8.8 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 28.54, 35.31, 37.35, 53.26, 55.37, 55.69, 113.49, 114.70, 116.80, 121.61, 124.63, 126.67, 129.90, 131.55, 143.59, 158.60, 159.28, 193.69; HRMS (ESI, m/z): calcd for $\text{C}_{24}\text{H}_{25}\text{NO}_3$ ($\text{M}+\text{H}^+$) 375.1834, found: 375.1834.

6,7-dihydro-3-(4-methoxyphenyl)-6,6-dimethyl-1-p-tolyl-1H-indol-4(5H)-one (4t)

Yield 89 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.08 (s, 6H, CH_3), 2.40 (s, 2H, CH_2), 2.42 (s, 3H, PhCH_3), 2.62 (s, 2H, CH_2), 3.80 (s, 3H, OCH_3), 6.79 (s, 1H, ArH), 6.89 (d, J =8.8 Hz, 2H, ArH), 7.21 (d, J =8.1 Hz, 2H, ArH), 7.28 (d, J =8.1 Hz, 2H, ArH), 7.63 (d, J =8.8 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 21.14, 28.45, 35.28, 37.40, 53.19, 55.28, 113.41, 116.94, 121.36, 124.72, 125.08, 126.59, 129.87, 130.13, 136.04, 138.03, 143.32, 158.54, 193.69; HRMS (ESI, m/z): calcd for $\text{C}_{24}\text{H}_{25}\text{NO}_2$ ($\text{M}+\text{H}^+$) 359.1885, found: 359.1886.

6,7-dihydro-1-(3-methoxyphenyl)-3-(4-methoxyphenyl)-6,6-dimethyl-1H-indol-4(5H)-one (4u)

4u

Yield 87 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.09 (s, 6H, 2 CH_3), 2.41 (s, 2H, CH_2), 2.66 (s, 2H, CH_2), 3.80 (s, 3H, OCH_3), 3.84 (s, 3H, OCH_3), 6.82 (s, 1H, ArH), 6.87 (t, J =8.1 Hz, 1H, ArH), 6.89 (d, J =8.8 Hz, 2H, ArH), 6.91-6.96 (m, 2H, ArH), 7.39 (t, J =8.4 Hz, 1H, ArH), 7.62 (d, J =8.8 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 28.41, 35.27, 37.45, 53.16, 55.25, 55.56, 111.31, 113.28, 113.39, 117.11, 117.40, 121.21, 124.83, 126.46, 129.85, 130.29, 139.58, 143.16, 158.56, 160.36, 193.66; HRMS (ESI, m/z): calcd for $\text{C}_{24}\text{H}_{25}\text{NO}_3$ ($\text{M}+\text{H}^+$) 375.1834, found: 375.1833.

6,7-dihydro-3-(4-methoxyphenyl)-6,6-dimethyl-1-o-tolyl-1H-indol-4(5H)-one (4v)

4v

Yield 87 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.08 (s, 6H, 2 CH_3), 2.10 (s, 3H, PhCH_3), 2.23 (d, J =16.5 Hz, 1H, CH_2), 2.41 (d, J =9.9 Hz, 2H, CH_2), 2.46 (d, J =16.1 Hz, 1H, CH_2), 3.80 (s, 3H, OCH_3), 6.67 (s, 1H, ArH), 6.89 (d, J =8.8 Hz, 2H, ArH), 7.21 (d, J =7.7 Hz, 1H, ArH), 7.29-7.41 (m, 3H, ArH), 7.66 (d, J =8.8 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 17.45, 27.87, 35.23, 36.60, 53.32, 55.30, 113.43, 116.18, 121.30, 124.53, 126.65, 126.96, 127.72, 129.27, 129.82, 131.19, 135.53, 137.50, 144.33, 158.52, 193.52; HRMS (ESI, m/z): calcd for $\text{C}_{24}\text{H}_{25}\text{NO}_2$ ($\text{M}+\text{H}^+$) 359.1885, found: 359.1886.

6,7-dihydro-3-(4-methoxyphenyl)-6,6-dimethyl-1-m-tolyl-1H-indol-4(5H)-one (4w)

4w

Yield 87 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.09 (s, 6H, 2 CH_3), 2.42 (s, 2H, CH_2), 2.44 (s, 3H, PhCH_3), 2.65 (s, 2H, CH_2), 3.82 (s, 3H, OCH_3), 6.82 (s, 1H, ArH), 6.90 (d, J =9.2 Hz, 2H, ArH), 7.14 (d, J =7.7 Hz, 1H, ArH), 7.16 (s, 1H, ArH), 7.23 (d, J =7.7 Hz, 1H, ArH), 7.39 (t, J =7.7 Hz, 1H, ArH), 7.62 (d, J =8.8 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 21.52, 28.54, 35.41, 37.54, 53.27, 55.39, 113.50, 117.13, 121.42, 122.40, 124.86, 125.98, 126.63, 128.83, 129.38, 129.96, 138.62, 139.84, 143.30, 158.65, 193.82; HRMS (ESI, m/z): calcd for $\text{C}_{24}\text{H}_{25}\text{NO}_2$ ($\text{M}+\text{H}^+$) 359.1885, found: 359.1887.

6,7-dihydro-1-(2,3-dihydro-1H-inden-6-yl)-6,6-dimethyl-3-phenyl-1H-indol-4(5H)-one (4x)

4x

Yield 88 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.09 (s, 6H, 2 CH_3), 2.12-2.19 (m, 2H, CH_2), 2.42 (s, 2H, CH_2), 2.64 (s, 2H, CH_2), 2.95-3.00 (m, 4H, $\text{CH}_2(\text{CH}_2)\text{CH}_2$), 6.84 (s, 1H, ArH), 7.08 (d, J =8.1 Hz, 1H, ArH), 7.18 (s, 1H, ArH), 7.23 (s, 1H, ArH), 7.30-7.36 (m, 3H, ArH), 7.69 (d, J =7.7 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 25.71, 28.49, 32.60, 32.99, 35.34, 37.45, 53.28, 116.93, 121.45, 122.25, 123.30, 124.94, 125.08, 126.61, 127.94, 128.76, 134.18, 136.75, 143.60, 144.44, 146.01, 193.61; HRMS (ESI, m/z): calcd for $\text{C}_{25}\text{H}_{25}\text{NO}$ ($\text{M}+\text{H}^+$) 355.1936, found: 355.1937.

1-cyclohexyl-3-(4-fluorophenyl)-6,7-dihydro-6,6-dimethyl-1H-indol-4(5H)-one (4y)

4y

Yield 79 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.15 (s, 6H, 2 CH_3), 1.28-1.30 (m, 1H, CH_2), 1.38-1.48 (m, 2H, CH_2), 1.60-1.70 (m, 2H, CH_2), 1.77 (d, J =13.2 Hz, 1H, CH_2), 1.93 (d, J =13.2 Hz, 2H, CH_2), 2.00 (d, J =12.5 Hz, 2H, CH_2), 2.37 (s, 2H, CH_2), 2.64 (s, 2H, CH_2), 3.77-3.85 (m, 1H, CH_2), 6.72 (s, 1H, ArH), 6.97-7.02 (m, 2H, ArH), 7.58-7.62 (m, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 25.39, 25.85, 28.78, 34.02, 35.04, 36.47, 53.29, 55.91, 114.57, 114.79, 117.32, 123.46, 130.19, 130.27, 142.70, 193.47; HRMS (ESI, m/z): calcd for $\text{C}_{22}\text{H}_{26}\text{FNO}$ ($\text{M}+\text{H}^+$) 339.1998, found: 339.1999.

1-cyclohexyl-6,7-dihydro-3-(4-methoxyphenyl)-6,6-dimethyl-1H-indol-4(5H)-one (4z)

4z

Yield 82 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.14 (s, 6H, 2 CH_3), 1.25-1.29 (m, 1H, CH_2), 1.37-1.47 (m, 2H, CH_2), 1.60-1.70 (m, 2H, CH_2), 1.77 (d, J =12.5 Hz, 1H, CH_2), 1.92 (d, J =13.2 Hz, 2H, CH_2), 2.00 (d, J =12.1 Hz, 2H, CH_2), 2.37 (s, 2H, CH_2), 2.63 (s, 2H, CH_2), 3.75-3.82 (m, 4H, PhOCH_3 , CH), 6.70 (s, 1H, ArH), 6.87 (d, J =8.8 Hz, 2H, ArH), 7.58 (d, J =8.8 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 25.38, 25.85, 28.78, 33.98, 34.98, 36.47, 53.33, 55.37, 55.82, 113.41, 115.76, 116.80, 124.06, 127.30, 129.77, 142.40, 158.40, 193.32; HRMS (ESI, m/z): calcd for $\text{C}_{23}\text{H}_{29}\text{NO}_2$ ($\text{M}+\text{H}^+$) 351.2198, found: 351.2199.

6,7-dihydro-1-(3-methoxypropyl)-6,6-dimethyl-3-phenyl-1H-indol-4(5H)-one (4ab)

Yield 76 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.14 (s, 6H, 2 CH_3), 1.93-1.99 (m, 2H, CH_2), 2.37 (s, 2H, CH_2), 2.63 (s, 2H, CH_2), 3.31-3.34 (m, 5H, $\text{CH}_2(\text{CH}_2)\text{CH}_2$), 3.92 (t, J =7.0 Hz, 2H, OCH_3), 6.66 (s, 1H, ArH), 7.21 (t, J =7.4 Hz, 1H, ArH), 7.30 (d, J =8.1 Hz, 1H, ArH), 7.64 (d, J =7.3 Hz, 2H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 28.68, 30.85, 35.05, 36.11, 43.35, 53.14, 58.75, 68.50, 116.00, 121.08, 124.57, 126.45, 127.90, 128.62, 134.38, 143.55, 193.38; HRMS (ESI, m/z): calcd for $\text{C}_{20}\text{H}_{25}\text{NO}_2$ ($\text{M}+\text{H}^+$) 311.1885, found: 311.1886.

6,7-dihydro-6,6-dimethyl-1,3-diphenyl-1H-indol-4(5H)-one (**4ac**)

Yield 92 %; brown sticky liquid; ^1H NMR (400 MHz, CDCl_3): δ = 1.12 (s, 6 H, 2 CH_3), 2.46 (s, 2 H, CH_2), 2.68 (s, 2 H, CH_2), 6.91 (s, 1 H, CH), 7.39–7.35 (m, 4 H, ArH), 7.47–7.43 (m, 2 H, ArH), 7.55–7.52 (m, 2 H, ArH), 7.71–7.68 (m, 2 H, ArH), ^{13}C NMR (100 MHz, CDCl_3): δ = 28.41, 35.33, 37.42, 53.27, 117.22, 121.92, 125.34, 126.67, 126.72, 127.93, 128.04, 128.72, 128.95, 129.61, 133.91, 138.52, 143.42, 193.67; HRMS (ESI, m/z): calcd for $\text{C}_{22}\text{H}_{21}\text{NO}$ ($\text{M}+\text{H}^+$) 315.1623, found: 315.1621.

^1H NMR and ^{13}C NMR spectra of the compounds

4i

4j

X : parts per Million : 1H

X : parts per Million : 13C

4m

4t

X : parts per Million : 1H

X : parts per Million : 13C