Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.

Supporting Information

Hydride Transfer Enabled Switchable Dearomatization of Indoles in the Carbocyclic Ring and the Pyrrole Ring

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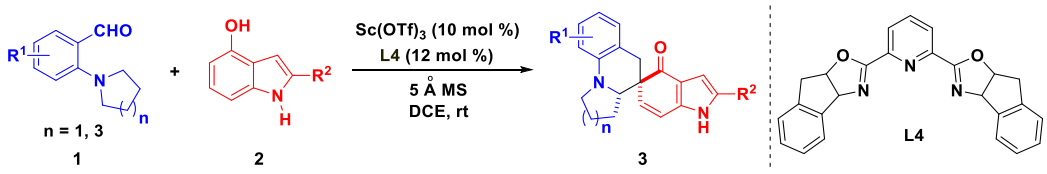
1. General Information

Unless otherwise noted, all reagents and solvents were purchased from the commercial sources and used as received. Thin layer chromatography (TLC) was used to monitor the reaction on Merck 60 F254 precoated silica gel plate (0.2 mm thickness). TLC spots were visualized by UV-light irradiation on Spectroline Model ENF-24061/F 254 nm. The products were purified by flash column chromatography (200-300 mesh silica gel) eluted with the gradient of petroleum ether and ethyl acetate. Proton nuclear magnetic resonance spectra (^{1}H NMR) were recorded on a Bruker 500 MHz NMR spectrometer ($CDCl_3$ or DMSO-d_6 solvent). The chemical shifts were reported in parts per million (ppm), downfield from $SiMe_4$ (δ 0.0) and relative to the signal of chloroform-d (δ 7.26, singlet) or dimethyl sulfoxide-d_6 (δ 2.54, singlet). Multiplicities were afforded as: s (singlet); d (doublet); t (triplet); q (quartet); dd (doublets of doublet) or m (multiplets). The number of protons for a given resonance is indicated by nH. Coupling constants were reported as a *J* value in Hz. Carbon nuclear magnetic resonance spectra (^{13}C NMR) was referenced to the appropriate residual solvent peak. High resolution mass spectral analysis (HRMS) was performed on Waters XEVO G2 Q-TOF. All substituted 2-fluorobenzaldehydes, 4-hydroxyindole, and 4-hydroxycarbazole were purchased from adamas-beta. All *o*-aminobenzaldehydes and 2-ary-4-hydroxyindoles were prepared according to literature.[1,2]

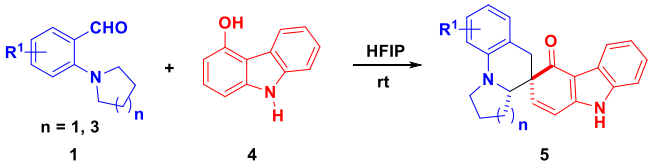
2. General Procedure

2.1 General Procedure for the Dearomatization of 4-Hydroxyindoles in the Carbocyclic Ring



A reaction tube was charged with *o*-aminobenzaldehyde **1** (0.1 mmol), 4-hydroxyindole **2** (0.12 mmol), $Sc(OTf)_3$ (10 mol %), **L4** (12 mol %), 5Å MS (60 mg) and DCE (1.0 mL). The mixture was stirred at room temperature under an air atmosphere. Upon completion of the reaction as indicated by TLC analysis, the reaction mixture was concentrated in vacuum and the residue was directly purified by flash column chromatography on silica gel (eluent: ethyl acetate/petroleum ether, 1:3) to afford the desired dearomatized indole **3**.

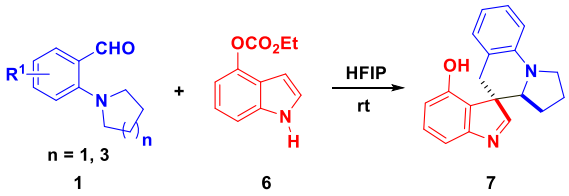
2.2 General Procedure for the Dearomatization of 4-hydroxycarbazoles in the Carbocyclic Ring



A reaction tube was charged with *o*-aminobenzaldehyde **1** (0.1 mmol), 4-hydroxycarbazole **4** (0.1 mmol), and HFIP (1.0 mL). The mixture was stirred at room temperature under an air atmosphere. Upon completion of the reaction as indicated by TLC analysis, the mixture was concentrated in vacuum and the residue was directly purified by flash column chromatography on silica gel (eluent:

ethyl acetate/petroleum ether, 1:3) to afford the desired dearomatized carbazole **5**.

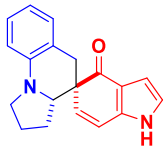
2.3 General Procedure for the Dearomatization of the Pyrrole Ring of OH protected 4-Hydroxyindoles



A reaction tube was charged with *o*-aminobenzaldehyde **1** (0.1 mmol), 4-hydroxycarbazole **6** (0.1 mmol), and HFIP (2.0 mL). The mixture was stirred at room temperature under an air atmosphere. Upon completion of the reaction as indicated by TLC analysis, the mixture was concentrated in vacuum and the residue was directly purified by flash column chromatography on silica gel (eluent: ethyl acetate/petroleum ether, 1:3) to afford the desired spiroindolenine **7**.

3. Characterization of Products

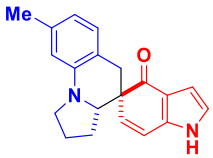
1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3a)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (27.6 mg, 95% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 11.62 (s, 1H), 7.08 (t, *J* = 7.6 Hz, 1H), 7.01 (d, *J* = 7.4 Hz, 1H), 6.93 (s, 1H), 6.74 (d, *J* = 9.9 Hz, 1H), 6.61 – 6.49 (m, 2H), 6.43 (s, 1H), 5.61 (d, *J* = 9.9 Hz, 1H), 3.69 (dd, *J* = 9.8, 5.8 Hz, 1H), 3.48 (t, *J* = 7.8 Hz, 1H), 3.21 (d, *J* = 15.6 Hz, 1H), 3.09 (dd, *J* = 16.6, 9.0 Hz, 1H), 2.54 (s, 1H), 1.89 (ddd, *J* = 15.9, 8.7, 5.4 Hz, 2H), 1.68 (dt, *J* = 11.8, 5.9 Hz, 1H), 1.18 (dd, *J* = 19.6, 10.2 Hz, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 195.6, 143.3, 140.3, 134.1, 129.3, 127.8, 121.8, 119.808, 119.2, 117.8, 115.4, 110.9, 105.5, 64.7, 48.1, 47.5, 39.1, 27.6, 23.5. **HRMS (ESI):** calcd. for $C_{19}H_{19}N_2O$ $[M+H]^+$: 291.1492, found: 291.1495.

8'-methyl-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3b)

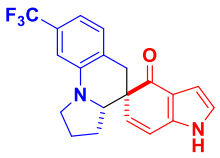


Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (28.3 mg, 93% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.93 (s, 1H), 6.92 (d, *J* = 7.5 Hz, 1H), 6.82 – 6.75 (m, 1H), 6.64 (s, 1H), 6.59 (d, *J* = 10.0 Hz, 1H), 6.43 (d, *J* = 7.5 Hz, 1H), 6.35 (s, 1H), 5.86 (d, *J* = 10.0 Hz, 1H), 3.87 (dd, *J* = 9.9, 5.5 Hz, 1H), 3.48 (td, *J* = 8.7, 2.6 Hz, 1H), 3.42 (d, *J* = 15.5 Hz, 1H), 3.17 (dt, *J* = 16.4, 8.2 Hz, 1H), 2.61 (d, *J* = 15.6 Hz, 1H), 2.31 (s, 3H), 1.98 – 1.80 (m, 3H), 1.29 (dd, *J* = 12.6, 5.1 Hz, 1H); **^{13}C NMR** (126 MHz, $CDCl_3$) δ 197.2, 142.9, 140.5, 137.1, 136.1, 128.9, 120.3, 119.5, 116.9, 116.2, 116.0, 111.0,

106.2, 64.7, 49.3, 47.1, 39.0, 27.5, 23.5, 21.6. **HRMS (ESI):** calcd. for $C_{20}H_{21}N_2O$ $[M+H]^+$: 305.1648, found: 305.1640.

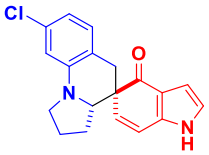
8'-(trifluoromethyl)-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3c)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (16.1 mg, 45% yield, dr >20:1) as a white solid.

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.54 (s, 1H), 7.11 (d, *J* = 7.7 Hz, 1H), 6.82 (d, *J* = 8.3 Hz, 2H), 6.69 (s, 2H), 6.63 (d, *J* = 10.0 Hz, 1H), 5.78 (d, *J* = 9.9 Hz, 1H), 3.90 (dd, *J* = 10.0, 5.5 Hz, 1H), 3.53 (t, *J* = 8.0 Hz, 1H), 3.47 (d, *J* = 15.9 Hz, 1H), 3.20 (dd, *J* = 16.7, 8.5 Hz, 1H), 2.67 (d, *J* = 16.0 Hz, 1H), 1.96 (dd, *J* = 17.2, 9.8 Hz, 2H), 1.91 – 1.80 (m, 1H), 1.38 – 1.21 (m, 1H); **^{13}C NMR** (126 MHz, $CDCl_3$) δ 196.3, 143.0, 140.1, 135.2, 129.8, 129.3, 126.8 (q, *J* = 273.4 Hz), 123.5, 120.4, 119.6, 116.6, 111.5 (q, *J* = 3.8 Hz), 106.6, 106.4 (q, *J* = 5.0 Hz), 64.8, 48.4, 47.2, 39.1, 27.5, 23.5. **HRMS (ESI):** calcd. for $C_{20}H_{18}F_3NO_3$ $[M+H]^+$: 359.1366, found: 359.1370.

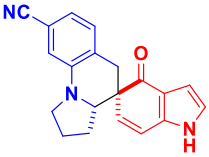
8'-chloro-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3d)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (18.9 mg, 58% yield, dr >20:1) as a white solid.

^{1}H NMR (500 MHz, DMSO) δ 11.64 (s, 1H), 7.01 (d, *J* = 7.7 Hz, 1H), 6.94 (s, 1H), 6.75 (d, *J* = 9.9 Hz, 1H), 6.53 (d, *J* = 7.4 Hz, 2H), 6.44 (s, 1H), 5.58 (d, *J* = 9.9 Hz, 1H), 3.70 (dd, *J* = 9.7, 5.8 Hz, 1H), 3.49 (t, *J* = 8.6 Hz, 1H), 3.14 (d, *J* = 15.8 Hz, 1H), 3.09 (d, *J* = 8.3 Hz, 1H), 2.56 (s, 1H), 1.88 (dd, *J* = 17.2, 7.8 Hz, 2H), 1.74 – 1.64 (m, 1H), 1.22 – 1.10 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 195.3, 144.4, 140.3, 133.6, 132.2, 130.5, 121.9, 119.1, 118.8, 118.1, 114.6, 109.9, 105.6, 64.5, 47.7, 47.5, 38.6, 27.5, 23.5. **HRMS (ESI):** calcd. for $C_{19}H_{18}ClN_2O$ $[M+H]^+$: 325.1102, found: 325.1110.

4-oxo-1,1',2',3',3a',4-hexahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinoline]-8'-carbonitrile (3e)

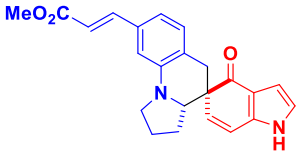


Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (29.0 mg, 92% yield, dr >20:1) as a reddish brown solid.

^{1}H NMR (500 MHz, DMSO) δ 7.20 (d, *J* = 7.4 Hz, 1H), 6.95 (s, 1H), 6.92 (d, *J* = 7.5 Hz, 1H), 6.88 (s, 1H), 6.76 (d, *J* = 9.9 Hz, 1H), 6.44 (s, 1H), 5.53 (d, *J* = 9.9 Hz, 1H), 3.77 – 3.66 (m, 1H), 3.54 (t, *J* = 8.8 Hz, 1H), 3.21 (d, *J* = 16.2 Hz, 1H), 3.15 – 3.05 (m, 1H), 2.63 (d, *J* = 16.2 Hz, 1H), 1.89 (dd, *J* = 18.2,

8.2 Hz, 2H), 1.70 (dd, *J* = 11.7, 5.7 Hz, 1H), 1.27 – 1.08 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 199.8, 148.3, 145.1, 137.9, 134.9, 130.5, 126.8, 126.7, 124.9, 123.8, 123.3, 123.1, 123.1, 117.7, 117.6, 115.1, 110.4, 69.3, 52.3, 51.9, 43.8, 32.3, 28.2. **HRMS (ESI):** calcd. for $C_{20}H_{18}N_3O$ $[M+H]^+$: 316.1444, found: 316.1440.

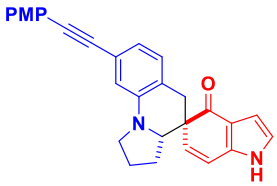
methyl(*E*)-3-(4-oxo-1,1',2',3',3a',4-hexahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-8'-yl)acrylate (3f)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (31.8 mg, 85% yield, dr >20:1) as a white solid.

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.87 (s, 1H), 7.66 (d, *J* = 15.9 Hz, 1H), 7.04 (d, *J* = 7.3 Hz, 1H), 6.83 – 6.72 (m, 2H), 6.64 (t, *J* = 12.5 Hz, 3H), 6.41 (d, *J* = 15.9 Hz, 1H), 5.80 (d, *J* = 9.8 Hz, 1H), 3.88 (s, 1H), 3.81 (s, 3H), 3.52 (t, *J* = 8.3 Hz, 1H), 3.46 (d, *J* = 16.1 Hz, 1H), 3.19 (d, *J* = 8.4 Hz, 1H), 2.65 (d, *J* = 16.0 Hz, 1H), 2.01 – 1.88 (m, 2H), 1.87 – 1.78 (m, 1H), 1.29 (d, *J* = 10.3 Hz, 1H); **^{13}C NMR** (126 MHz, $CDCl_3$) δ 200.9, 172.8, 143.3, 129.3, 127.8, 119.0, 115.3, 110.3, 99.3, 98.8, 74.4, 62.3, 46.7, 42.1, 37.3, 27.9, 25.9, 23.4. **HRMS (ESI):** calcd. for $C_{23}H_{23}N_2O_3$ $[M+H]^+$: 375.1703, found: 375.1705.

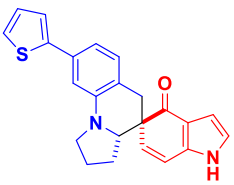
8'-((4-methoxyphenyl)ethynyl)-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3g)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (21.0 mg, 50% yield, dr >20:1) as a white solid.

^{1}H NMR (500 MHz, DMSO) δ 11.63 (s, 1H), 7.48 (d, *J* = 8.6 Hz, 2H), 7.04 (d, *J* = 7.6 Hz, 1H), 6.98 (d, *J* = 8.6 Hz, 2H), 6.93 (s, 1H), 6.75 (d, *J* = 9.9 Hz, 1H), 6.69 (d, *J* = 7.6 Hz, 1H), 6.63 (s, 1H), 6.43 (s, 1H), 5.59 (d, *J* = 9.9 Hz, 1H), 3.79 (s, 3H), 3.70 (dd, *J* = 9.8, 5.7 Hz, 1H), 3.52 (t, *J* = 8.2 Hz, 1H), 3.19 (d, *J* = 15.9 Hz, 1H), 3.12 (dd, *J* = 16.7, 9.0 Hz, 1H), 2.56 (d, *J* = 16.0 Hz, 1H), 1.96 – 1.81 (m, 2H), 1.68 (dt, *J* = 11.5, 5.7 Hz, 1H), 1.18 (dd, *J* = 18.7, 11.2 Hz, 1H). **^{13}C NMR** (126 MHz, DMSO) δ 195.3, 159.8, 143.2, 140.4, 133.8, 133.3, 129.5, 121.8, 121.8, 120.8, 119.1, 118.4, 118.0, 114.8, 112.8, 105.5, 89.6, 88.4, 64.6, 55.7, 47.8, 47.4, 39.1, 27.5, 23.5. **HRMS (ESI):** calcd. for $C_{28}H_{25}N_2O_2$ $[M+H]^+$: 421.1911, found: 421.1915.

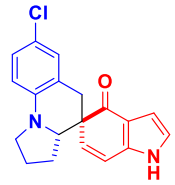
8'-(thiophen-2-yl)-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3h)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (18.9 mg, 51% yield, dr >20:1) as a brown solid.

^{1}H NMR (500 MHz, DMSO) δ 11.64 (s, 1H), 7.59 – 7.47 (m, 1H), 7.45 (s, 1H), 7.12 (s, 1H), 7.05 (d, *J* = 7.5 Hz, 1H), 6.95 (s, 1H), 6.82 (d, *J* = 7.5 Hz, 1H), 6.76 (s, 2H), 6.45 (s, 1H), 5.65 (d, *J* = 9.9 Hz, 1H), 3.80 – 3.68 (m, 1H), 3.58 (t, *J* = 8.4 Hz, 1H), 3.26 – 3.11 (m, 2H), 2.56 (d, *J* = 15.8 Hz, 1H), 1.90 (d, *J* = 22.7 Hz, 2H), 1.74 – 1.65 (m, 1H), 1.21 (dd, *J* = 21.2, 10.7 Hz, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 195.5, 144.9, 143.6, 140.3, 134.0, 133.4, 129.9, 128.7, 125.2, 123.4, 121.8, 119.8, 119.2, 117.9, 113.0, 107.6, 105.6, 64.7, 48.2, 47.5, 38.9, 27.6, 23.5. **HRMS (ESI):** calcd. for $C_{23}H_{21}N_2OS$ $[M+H]^+$: 373.1369, found: 373.1372.

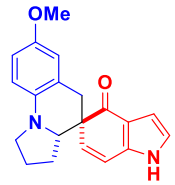
7'-chloro-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3i)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (26.6 mg, 82% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 11.65 (s, 1H), 7.10 (d, *J* = 8.2 Hz, 2H), 6.94 (s, 1H), 6.76 (d, *J* = 9.8 Hz, 1H), 6.53 (d, *J* = 8.3 Hz, 1H), 6.44 (s, 1H), 5.57 (d, *J* = 9.8 Hz, 1H), 3.76 – 3.62 (m, 1H), 3.48 (t, *J* = 8.3 Hz, 1H), 3.17 (d, *J* = 15.8 Hz, 1H), 3.12 – 3.00 (m, 1H), 2.56 (d, *J* = 15.9 Hz, 1H), 1.87 (d, *J* = 21.8 Hz, 2H), 1.69 (d, *J* = 5.5 Hz, 1H), 1.23 – 1.09 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 200.0, 146.9, 145.1, 138.4, 133.5, 133.4, 132.1, 132.1, 126.7, 126.6, 123.9, 123.5, 122.9, 122.8, 116.8, 110.3, 69.4, 52.5, 52.4, 43.5, 32.3, 28.3. **HRMS (ESI):** calcd. for $C_{19}H_{18}ClN_2O$ $[M+H]^+$: 325.1102, found: 325.1108.

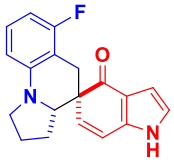
7'-methoxy-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3j)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (17.6 mg, 55% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 11.59 (s, 1H), 6.92 (s, 1H), 6.71 (dd, *J* = 17.0, 7.6 Hz, 3H), 6.49 (d, *J* = 8.5 Hz, 1H), 6.41 (s, 1H), 5.62 (d, *J* = 9.9 Hz, 1H), 3.64 (d, *J* = 7.6 Hz, 3H), 3.63 – 3.54 (m, 1H), 3.42 (t, *J* = 8.0 Hz, 1H), 3.20 (d, *J* = 15.8 Hz, 1H), 3.10 – 2.99 (m, 1H), 2.47 (s, 1H), 1.92 – 1.75 (m, 2H), 1.66 (dd, *J* = 11.9, 5.9 Hz, 1H), 1.19 – 1.08 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 195.6, 150.5, 140.3, 137.9, 134.3, 121.7, 121.0, 119.2, 117.8, 115.4, 113.5, 111.9, 105.5, 64.9, 55.8, 48.7, 48.1, 39.1, 27.5, 23.6. **HRMS (ESI):** calcd. for $C_{20}H_{21}N_2O_2$ $[M+H]^+$: 321.1598, found: 321.1602.

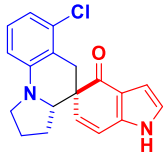
6'-fluoro-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3k)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (27.7 mg, 90% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 11.66 (s, 1H), 7.09 (dd, *J* = 15.1, 7.9 Hz, 1H), 6.95 (d, *J* = 1.7 Hz, 1H), 6.77 (d, *J* = 9.9 Hz, 1H), 6.44 (d, *J* = 2.5 Hz, 1H), 6.42 – 6.34 (m, 2H), 5.59 (d, *J* = 9.9 Hz, 1H), 3.64 (dd, *J* = 9.8, 5.9 Hz, 1H), 3.54 – 3.46 (m, 1H), 3.16 – 3.08 (m, 1H), 2.96 (d, *J* = 16.1 Hz, 1H), 2.65 (d, *J* = 15.3 Hz, 1H), 1.95 – 1.82 (m, 2H), 1.70 (ddd, *J* = 12.4, 7.3, 1.7 Hz, 1H), 1.22 – 1.12 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 195.2, 161.3 (d, *J* = 239.4 Hz), 144.8 (d, *J* = 7.6 Hz), 140.3, 133.4, 128.4 (d, *J* = 10.1 Hz), 121.9, 119.1, 118.2, 107.0, 106.4 (d, *J* = 20.2 Hz), 105.6, 101.8 (d, *J* = 22.7 Hz), 64.1, 47.8, 47.2, 31.5, 27.5, 23.4. **HRMS (ESI):** calcd. for $C_{19}H_{18}FN_2O$ $[M+H]^+$: 309.1398, found: 309.1402.

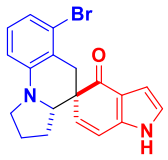
6'-chloro-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3l)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (27.5 mg, 85% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 11.67 (s, 1H), 7.09 (t, *J* = 8.1 Hz, 1H), 6.98 – 6.94 (m, 1H), 6.77 (d, *J* = 9.9 Hz, 1H), 6.69 – 6.63 (m, 1H), 6.54 (d, *J* = 8.0 Hz, 1H), 6.48 – 6.43 (m, 1H), 5.60 (d, *J* = 9.9 Hz, 1H), 3.65 (dd, *J* = 9.7, 5.9 Hz, 1H), 3.49 (td, *J* = 9.1, 2.3 Hz, 1H), 3.14 (dd, *J* = 16.6, 9.2 Hz, 1H), 3.01 (d, *J* = 16.4 Hz, 1H), 2.75 (d, *J* = 16.4 Hz, 1H), 1.97 – 1.85 (m, 2H), 1.71 (ddd, *J* = 12.2, 8.6, 3.9 Hz, 1H), 1.21 – 1.14 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 195.1, 144.6, 140.3, 133.8, 133.5, 128.5, 122.0, 119.1, 118.2, 117.1, 115.7, 109.9, 105.6, 64.0, 47.9, 47.7, 36.6, 27.5, 23.5. **HRMS (ESI):** calcd. for $C_{19}H_{18}ClN_2O$ $[M+H]^+$: 325.1102, found: 325.1104.

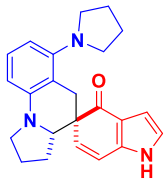
6'-bromo-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3m)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (32.4 mg, 88% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 11.67 (s, 1H), 7.01 (t, *J* = 8.0 Hz, 1H), 6.97 – 6.93 (m, 1H), 6.81 (d, *J* = 7.9 Hz, 1H), 6.78 (d, *J* = 9.9 Hz, 1H), 6.56 (d, *J* = 8.2 Hz, 1H), 6.45 (s, 1H), 5.60 (d, *J* = 9.9 Hz, 1H), 3.65 (dd, *J* = 9.7, 5.9 Hz, 1H), 3.48 (t, *J* = 8.7 Hz, 1H), 3.13 (dd, *J* = 16.8, 8.9 Hz, 1H), 3.02 (d, *J* = 16.3 Hz, 1H), 2.72 (d, *J* = 16.3 Hz, 1H), 1.97 – 1.82 (m, 2H), 1.75 – 1.65 (m, 1H), 1.18 (dd, *J* = 14.9, 6.4 Hz, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 195.0, 144.7, 140.3, 133.5, 129.0, 125.1, 122.0, 119.1, 118.9, 118.6, 118.2, 110.4, 105.6, 64.1, 48.2, 47.6, 39.6, 27.4, 23.5. **HRMS (ESI):** calcd. for $C_{19}H_{18}BrN_2O$ $[M+H]^+$: 369.0597, found: 369.0595.

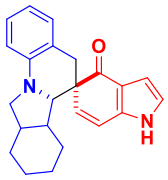
6'-(pyrrolidin-1-yl)-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3n)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (19.0 mg, 53% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.95 (s, 1H), 7.08 (t, *J* = 8.1 Hz, 1H), 6.70 – 6.66 (m, 1H), 6.63 (t, *J* = 2.3 Hz, 1H), 6.44 (d, *J* = 9.9 Hz, 1H), 6.37 (d, *J* = 7.8 Hz, 1H), 6.26 (d, *J* = 8.0 Hz, 1H), 5.64 (d, *J* = 10.0 Hz, 1H), 3.91 (dd, *J* = 9.7, 5.3 Hz, 1H), 3.57 – 3.51 (m, 1H), 3.35 (dd, *J* = 15.9, 8.2 Hz, 2H), 3.19 (d, *J* = 15.6 Hz, 1H), 3.15 – 3.10 (m, 1H), 2.87 – 2.80 (m, 2H), 2.76 (d, *J* = 15.6 Hz, 1H), 1.92 – 1.83 (m, 5H), 1.80 – 1.73 (m, 2H), 1.28 – 1.25 (m, 1H); **^{13}C NMR** (126 MHz, $CDCl_3$) δ 197.8, 150.1, 143.9, 140.5, 135.9, 127.0, 120.4, 119.6, 116.4, 110.9, 106.1, 104.5, 104.0, 64.2, 51.3, 49.7, 48.1, 36.9, 27.7, 24.6, 23.7. **HRMS (ESI):** calcd. for $C_{23}H_{26}N_3O$ $[M+H]^+$: 360.2070, found: 360.2075.

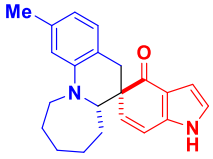
6a',6b',7',8',9',10',10a',11'-octahydro-5'*H*-spiro[indole-5,6'-isoindolo[2,1-*a*]quinolin]-4(1*H*)-one (3o)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (19.3 mg, 56% yield, dr >20:1) as a white solid.

^{1}H NMR (500 MHz, DMSO) δ 11.43 (s, 1H), 6.91 (t, *J* = 7.6 Hz, 1H), 6.82 (d, *J* = 7.1 Hz, 1H), 6.77 (s, 1H), 6.54 (d, *J* = 9.9 Hz, 1H), 6.36 (dd, *J* = 14.6, 7.6 Hz, 2H), 6.29 (s, 1H), 5.41 (d, *J* = 9.9 Hz, 1H), 3.82 (d, *J* = 9.7 Hz, 1H), 3.08 (d, *J* = 9.2 Hz, 1H), 3.02 (d, *J* = 15.2 Hz, 2H), 2.33 (s, 1H), 1.92 (d, *J* = 5.4 Hz, 1H), 1.60 (s, 1H), 1.48 (t, *J* = 14.9 Hz, 2H), 1.13 (d, *J* = 20.9 Hz, 6H); **^{13}C NMR** (126 MHz, DMSO) δ 195.9, 143.8, 140.1, 134.6, 128.9, 127.8, 121.8, 119.6, 119.3, 117.4, 115.1, 110.5, 105.5, 64.3, 55.4, 54.1, 47.5, 37.0, 28.6, 26.1, 25.2, 21.6. **HRMS (ESI):** calcd. for $C_{23}H_{25}N_2O$ $[M+H]^+$: 345.1961, found: 345.1960.

2-methyl-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,5'-indol]-4'(1'H)-one (3p)

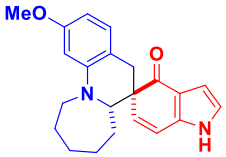


Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (23.2 mg, 70% yield, dr 4:1) as a white solid.

^{1}H NMR (500 MHz, DMSO) δ 11.58 (s, 1H), 6.90 (d, *J* = 6.7 Hz, 1H), 6.83 (d, *J* = 7.5 Hz, 1H), 6.72 (d, *J* = 9.9 Hz, 1H), 6.58 (s, 1H), 6.44 – 6.39 (m, 2H), 5.88 (d, *J* = 9.9 Hz, 1H), 3.49 (dd, *J* = 8.8, 4.7 Hz,

1H), 3.46 – 3.40 (m, 1H), 3.36 (d, *J* = 8.5 Hz, 1H), 3.22 (d, *J* = 15.8 Hz, 1H), 2.36 (d, *J* = 15.9 Hz, 1H), 2.23 (s, 3H), 1.76 (d, *J* = 6.2 Hz, 1H), 1.71 – 1.63 (m, 1H), 1.62 – 1.49 (m, 4H), 1.31 (dd, *J* = 21.6, 15.0 Hz, 2H), 1.24 (d, *J* = 9.6 Hz, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 196.1, 147.0, 140.3, 136.1, 135.3, 129.2, 121.6, 119.5, 118.3, 118.0, 116.6, 114.9, 105.3, 66.0, 51.7, 50.8, 38.9, 30.6, 29.3, 29.1, 26.4, 21.6. **HRMS (ESI):** calcd. for $C_{22}H_{25}N_2O$ $[M+H]^+$: 333.1961, found: 333.1960.

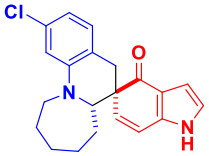
2-methoxy-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,5'-indol]-4'(1'H)-one (3q)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (20.9 mg, 60% yield, dr 5:1) as a white solid.

^{1}H NMR (500 MHz, DMSO) δ 11.58 (s, 1H), 6.91 (s, 1H), 6.84 (d, *J* = 8.2 Hz, 1H), 6.72 (d, *J* = 9.9 Hz, 1H), 6.40 (s, 1H), 6.25 (s, 1H), 6.21 (d, *J* = 8.2 Hz, 1H), 5.89 (d, *J* = 9.9 Hz, 1H), 3.71 (s, 3H), 3.55 (dd, *J* = 7.6, 5.1 Hz, 1H), 3.43 (d, *J* = 7.8 Hz, 1H), 3.37 (d, *J* = 7.9 Hz, 1H), 3.17 (d, *J* = 15.6 Hz, 1H), 2.35 (d, *J* = 15.6 Hz, 1H), 1.75 (s, 1H), 1.65 (s, 1H), 1.59 – 1.48 (m, 3H), 1.40 – 1.30 (m, 2H), 1.26 – 1.21 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 196.1, 159.1, 147.8, 140.2, 135.5, 129.8, 121.6, 119.4, 116.6, 113.8, 105.4, 102.7, 99.6, 65.6, 55.2, 51.9, 50.6, 38.7, 30.2, 29.3, 26.4. **HRMS (ESI):** calcd. for $C_{22}H_{25}N_2O_2$ $[M+H]^+$: 349.1911, found: 349.1916.

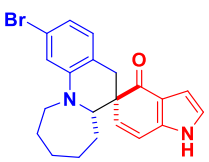
2-chloro-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,5'-indol]-4'(1'H)-one (3r)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (23.9 mg, 68% yield, dr 8:1) as a white solid.

^{1}H NMR (500 MHz, DMSO) δ 11.62 (s, 1H), 6.99 – 6.90 (m, 2H), 6.73 (d, *J* = 10.7 Hz, 2H), 6.58 (d, *J* = 7.7 Hz, 1H), 6.41 (s, 1H), 5.86 (d, *J* = 9.8 Hz, 1H), 3.60 (d, *J* = 6.6 Hz, 1H), 3.48 – 3.36 (m, 2H), 3.16 (d, *J* = 15.9 Hz, 1H), 2.41 (d, *J* = 16.0 Hz, 1H), 1.73 (s, 1H), 1.62 (d, *J* = 7.3 Hz, 2H), 1.51 (d, *J* = 4.4 Hz, 2H), 1.42 – 1.25 (m, 3H); **^{13}C NMR** (126 MHz, DMSO) δ 195.7, 147.8, 140.2, 135.1, 131.8, 130.6, 121.7, 120.1, 119.2, 116.9, 116.0, 112.6, 105.5, 65.1, 51.3, 50.3, 38.6, 29.7, 29.3, 29.2, 26.3. **HRMS (ESI):** calcd. for $C_{21}H_{22}ClN_2O$ $[M+H]^+$: 353.1415, found: 353.1413.

2-bromo-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,5'-indol]-4'(1'H)-one (3s)

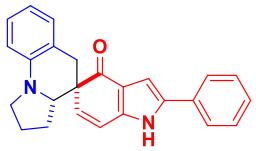


Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product

(30.9 mg, 78% yield, dr 4:1) as a white solid.

^{1}H NMR (500 MHz, DMSO) δ 11.64 (s, 1H), 6.97 – 6.89 (m, 2H), 6.87 (s, 1H), 6.74 (t, *J* = 10.0 Hz, 2H), 6.43 (s, 1H), 5.88 (d, *J* = 8.5 Hz, 1H), 3.60 (s, 1H), 3.44 (dd, *J* = 31.0, 11.2 Hz, 2H), 3.16 (d, *J* = 16.2 Hz, 1H), 2.42 (d, *J* = 16.0 Hz, 1H), 1.75 (s, 1H), 1.70 – 1.49 (m, 4H), 1.41 – 1.24 (m, 3H); **^{13}C NMR** (126 MHz, DMSO) δ 195.7, 148.1, 140.2, 135.0, 131.0, 121.8, 120.6, 120.3, 119.3, 118.9, 116.9, 115.4, 105.5, 65.1, 51.3, 50.4, 38.6, 29.7, 29.3, 29.2, 26.4. **HRMS (ESI):** calcd. for $C_{21}H_{22}BrN_2O$ $[M+H]^+$: 397.0910, found: 397.0911.

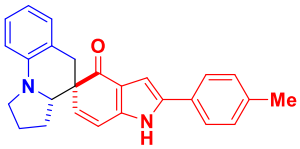
2-phenyl-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3t)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (26.1 mg, 71% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.11 (s, 1H), 7.75 (d, *J* = 7.5 Hz, 2H), 7.44 (t, *J* = 7.3 Hz, 2H), 7.29 (t, *J* = 7.1 Hz, 1H), 7.10 (t, *J* = 7.5 Hz, 1H), 7.04 (d, *J* = 7.1 Hz, 1H), 6.93 (s, 1H), 6.82 (d, *J* = 9.8 Hz, 1H), 6.55 (t, *J* = 6.8 Hz, 2H), 5.70 (d, *J* = 9.8 Hz, 1H), 3.80 – 3.68 (m, 1H), 3.50 (t, *J* = 8.4 Hz, 1H), 3.25 (d, *J* = 15.6 Hz, 1H), 3.12 (d, *J* = 8.4 Hz, 1H), 2.59 (d, *J* = 15.6 Hz, 1H), 1.92 (s, 2H), 1.80 – 1.68 (m, 1H), 1.26 – 1.19 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 200.3, 148.0, 146.3, 139.7, 139.4, 136.6, 134.1, 132.6, 132.2, 129.4, 125.1, 124.5, 122.4, 120.2, 115.6, 107.5, 107.5, 69.5, 53.0, 52.3, 43.9, 32.4, 28.3. **HRMS (ESI):** calcd. for $C_{25}H_{23}N_2O$ $[M+H]^+$: 367.1805, found: 367.1808.

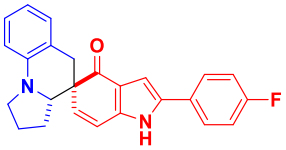
2-(*p*-tolyl)-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3u)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (19.0 mg, 50% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.04 (s, 1H), 7.64 (d, *J* = 5.9 Hz, 2H), 7.25 (d, *J* = 5.5 Hz, 2H), 7.06 (d, *J* = 33.2 Hz, 2H), 6.86 (s, 1H), 6.80 (d, *J* = 9.4 Hz, 1H), 6.55 (s, 2H), 5.68 (d, *J* = 9.3 Hz, 1H), 3.72 (s, 1H), 3.50 (s, 1H), 3.25 (d, *J* = 15.4 Hz, 1H), 3.11 (d, *J* = 7.2 Hz, 1H), 2.58 (d, *J* = 15.7 Hz, 1H), 2.34 (s, 3H), 1.91 (s, 2H), 1.72 (s, 1H), 1.32 – 1.11 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 200.3, 148.0, 146.1, 141.5, 139.6, 139.5, 134.7, 134.1, 133.9, 132.6, 129.4, 125.1, 124.5, 122.4, 120.2, 115.7, 106.9, 106.8, 69.5, 52.9, 52.3, 32.4, 28.3, 26.0. **HRMS (ESI):** calcd. for $C_{26}H_{24}N_2O$ $[M+H]^+$: 381.1961, found: 381.1966.

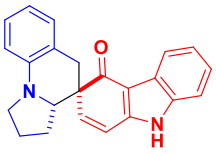
2-(4-fluorophenyl)-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3v)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (25.0 mg, 65% yield, dr 5:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.08 (s, 1H), 7.77 (dd, *J* = 8.6, 5.4 Hz, 2H), 7.27 (t, *J* = 8.8 Hz, 2H), 7.08 (t, *J* = 7.5 Hz, 1H), 7.02 (d, *J* = 7.4 Hz, 1H), 6.89 (s, 1H), 6.78 (d, *J* = 9.9 Hz, 1H), 6.53 (t, *J* = 7.5 Hz, 2H), 5.68 (d, *J* = 9.9 Hz, 1H), 3.71 (dd, *J* = 9.7, 5.8 Hz, 1H), 3.49 (t, *J* = 8.1 Hz, 1H), 3.22 (d, *J* = 15.5 Hz, 1H), 3.10 (d, *J* = 7.4 Hz, 1H), 2.56 (d, *J* = 15.7 Hz, 1H), 1.88 (d, *J* = 22.9 Hz, 2H), 1.76 – 1.67 (m, 1H), 1.23 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 195.5, 161.7 (d, *J* = 244.4 Hz), 143.2, 141.5, 135.0, 133.7, 129.3, 127.8, 126.7 (t, *J* = 7.6 Hz), 120.3, 119.7, 117.6, 116.3, 116.2, 115.4, 110.9, 102.7, 64.7, 48.2, 47.5, 39.1, 27.6, 23.5. **HRMS (ESI):** calcd. for $C_{25}H_{22}FN_2O$ $[M+H]^+$: 385.1711, found: 385.1710.

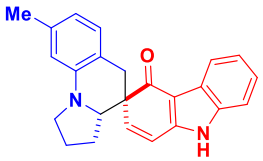
1',2',3',3a'-tetrahydro-5'*H*-spiro[carbazole-3,4'-pyrrolo[1,2-*a*]quinolin]-4(9*H*)-one (5a)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:6) afforded the product (24.8 mg, 73% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.13 (s, 1H), 8.04 (d, *J* = 7.0 Hz, 1H), 7.49 (d, *J* = 7.3 Hz, 1H), 7.32 – 7.17 (m, 2H), 7.09 (t, *J* = 7.5 Hz, 1H), 7.03 (d, *J* = 7.0 Hz, 1H), 6.93 (d, *J* = 9.9 Hz, 1H), 6.55 (t, *J* = 8.8 Hz, 2H), 5.99 (d, *J* = 9.9 Hz, 1H), 3.79 (dd, *J* = 9.2, 6.0 Hz, 1H), 3.49 (t, *J* = 7.7 Hz, 1H), 3.31 (s, 1H), 3.12 (d, *J* = 8.2 Hz, 1H), 2.61 (d, *J* = 15.8 Hz, 1H), 1.97 – 1.78 (m, 2H), 1.78 – 1.63 (m, 1H), 1.19 (d, *J* = 10.2 Hz, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 194.9, 146.9, 143.2, 140.5, 137.3, 129.3, 127.9, 124.5, 124.0, 123.0, 120.9, 119.6, 118.1, 115.5, 112.7, 111.0, 110.7, 64.7, 48.7, 47.5, 40.5, 40.4, 40.3, 40.3, 40.2, 40.1, 39.9, 39.8, 39.7, 39.5, 39.1, 27.7, 23.5. **HRMS (ESI):** calcd. for $C_{23}H_{21}N_2O$ $[M+H]^+$: 341.1648, found: 341.1653.

8'-methyl-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-4(9H)-one (5b)

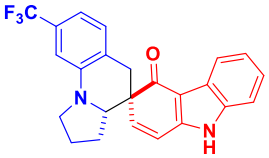


Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (21.3 mg, 60% yield, dr 3:1) as a light yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.12 (s, 1H), 8.04 (d, *J* = 6.8 Hz, 1H), 7.50 (d, *J* = 8.2 Hz, 1H), 7.30 – 7.19 (m, 2H), 6.96 – 6.90 (m, 2H), 6.38 (d, *J* = 9.5 Hz, 2H), 6.00 (d, *J* = 9.9 Hz, 1H), 3.78 (dd, *J* = 9.6, 5.9 Hz, 1H), 3.49 (t, *J* = 7.7 Hz, 1H), 3.28 (d, *J* = 15.6 Hz, 1H), 3.12 (dd, *J* = 16.7, 8.8 Hz, 1H), 2.58 (d, *J* = 15.6 Hz, 1H), 2.25 (s, 3H), 1.87 (dd, *J* = 19.3, 8.3 Hz, 2H), 1.77 – 1.68 (m, 1H), 1.21 (dd, *J* = 21.2, 11.8 Hz, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 195.0, 146.9, 143.1, 140.7, 137.3, 136.7, 129.2, 124.5, 124.0, 122.9, 120.9, 118.0, 116.8, 116.4, 112.7, 111.6, 110.7, 64.7, 49.0, 47.5, 38.8, 27.7, 23.5, 21.8. **HRMS (ESI):** calcd. for $C_{24}H_{23}N_2O$ $[M+H]^+$: 355.1805, found: 355.1808.

8'-(trifluoromethyl)-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-

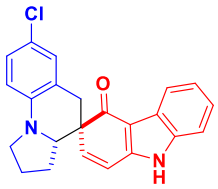
4(9H)-one (5c)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (33.5 mg, 82% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.17 (s, 1H), 8.10 – 7.97 (m, 1H), 7.51 (d, *J* = 7.2 Hz, 1H), 7.32 – 7.19 (m, 3H), 6.95 (d, *J* = 9.9 Hz, 1H), 6.84 (d, *J* = 7.7 Hz, 1H), 6.76 (s, 1H), 5.96 (d, *J* = 9.9 Hz, 1H), 3.84 (dd, *J* = 9.9, 5.8 Hz, 1H), 3.58 (t, *J* = 7.9 Hz, 1H), 3.33 (s, 1H), 3.17 (dd, *J* = 17.0, 8.9 Hz, 1H), 2.73 (d, *J* = 16.2 Hz, 1H), 1.97 – 1.86 (m, 2H), 1.75 (dt, *J* = 10.0, 5.6 Hz, 1H), 1.27 – 1.17 (m, 1H).; **^{13}C NMR** (126 MHz, DMSO) δ 194.4, 147.0, 143.5, 139.8, 137.3, 130.0, 127.4 (d, *J* = 302.4 Hz), 124.5, 124.2, 124.1, 123.1, 120.9, 118.5, 112.8, 111.3 (q, *J* = 3.8 Hz), 110.7, 106.4 (d, *J* = 3.8 Hz), 64.7, 47.9, 47.5, 38.8, 27.6, 23.5. **HRMS (ESI):** calcd. for $C_{24}H_{20}F_3N_2O$ $[M+H]^+$: 409.1522, found: 409.1526.

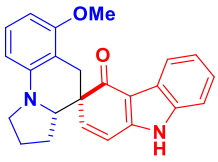
7'-chloro-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-4(9H)-one (5d)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (18.7 mg, 50% yield, dr 1.4:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.16 (s, 1H), 8.04 (d, *J* = 7.4 Hz, 1H), 7.50 (d, *J* = 7.7 Hz, 1H), 7.31 – 7.21 (m, 2H), 7.12 (s, 2H), 6.95 (d, *J* = 9.9 Hz, 1H), 6.60 – 6.53 (m, 1H), 5.96 (d, *J* = 9.9 Hz, 1H), 3.79 (dd, *J* = 9.6, 5.9 Hz, 1H), 3.50 (t, *J* = 8.1 Hz, 1H), 3.29 (d, *J* = 16.0 Hz, 1H), 3.12 (dd, *J* = 16.9, 8.7 Hz, 1H), 2.66 (d, *J* = 16.0 Hz, 1H), 1.90 (dd, *J* = 16.7, 9.0 Hz, 2H), 1.74 (dd, *J* = 11.7, 5.8 Hz, 1H), 1.23 – 1.16 (m, 1H).; **^{13}C NMR** (126 MHz, DMSO) δ 194.5, 146.9, 142.1, 140.0, 137.3, 128.8, 127.4, 124.5, 124.1, 123.1, 121.7, 120.9, 118.8, 118.4, 112.8, 112.2, 110.7, 64.7, 48.2, 47.7, 38.7, 27.6, 23.5. **HRMS (ESI):** calcd. for $C_{23}H_{20}ClN_2O$ $[M+H]^+$: 375.1259, found: 375.1265.

6'-methoxy-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-4(9H)-one (5e)

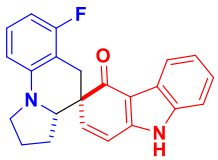


Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (20.7 mg, 56% yield, dr 14:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.09 (s, 1H), 8.11 (d, *J* = 7.6 Hz, 1H), 7.52 (d, *J* = 7.9 Hz, 1H), 7.25 (dt, *J* = 23.9, 7.2 Hz, 2H), 7.11 (t, *J* = 8.1 Hz, 1H), 6.40 (d, *J* = 10.3 Hz, 1H), 6.38 – 6.26 (m, 3H), 4.13 – 4.05 (m, 1H), 3.70 (d, *J* = 13.0 Hz, 3H), 3.50 (t, *J* = 7.5 Hz, 1H), 3.33 – 3.21 (m, 2H), 2.93 (d, *J* = 16.4 Hz, 1H), 1.85 (dd, *J* = 19.2, 8.6 Hz, 2H), 1.58 – 1.49 (m, 1H), 1.22 (dd, *J* = 20.0, 9.8 Hz, 1H); **^{13}C NMR**

(126 MHz, DMSO) δ 182.0, 157.7, 153.7, 144.3, 143.5, 137.1, 132.1, 128.4, 124.3, 123.6, 122.1, 121.0, 112.3, 112.1, 106.3, 105.2, 99.3, 63.5, 55.7, 48.0, 39.0, 34.1, 27.8, 23.3. **HRMS (ESI):** calcd. for $C_{24}H_{23}N_2O_2$ $[M+H]^+$: 371.1754, found: 371.1750.

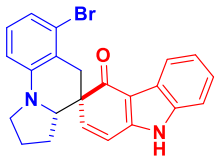
6'-fluoro-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-4(9H)-one (5f)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (24.0 mg, 67% yield, dr 4:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.18 (s, 1H), 8.05 (d, *J* = 6.9 Hz, 1H), 7.51 (d, *J* = 7.4 Hz, 1H), 7.31 – 7.23 (m, 2H), 7.12 (dd, *J* = 15.3, 7.9 Hz, 1H), 6.97 (d, *J* = 9.9 Hz, 1H), 6.41 (dd, *J* = 15.8, 8.2 Hz, 2H), 5.98 (d, *J* = 9.9 Hz, 1H), 3.76 (dd, *J* = 9.6, 5.9 Hz, 1H), 3.52 (t, *J* = 7.6 Hz, 1H), 3.16 (dd, *J* = 16.8, 8.8 Hz, 1H), 3.08 (d, *J* = 16.2 Hz, 1H), 2.75 (d, *J* = 16.0 Hz, 1H), 1.94 – 1.83 (m, 2H), 1.75 (dd, *J* = 11.1, 5.5 Hz, 1H), 1.23 (dd, *J* = 19.8, 10.1 Hz, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 194.5, 161.3 (d, *J* = 239.4 Hz), 147.0, 144.8 (d, *J* =5.0 Hz), 139.8, 137.3, 128.5 (d, *J* = 11.3 Hz), 124.5, 124.1, 123.1, 121.0, 118.5, 112.8, 110.7, 107.2, 106.2 (d, *J* = 20.1 Hz), 102.0 (d, *J* = 22.4 Hz), 64.2, 47.8, 47. 8, 31.4, 27.6, 23.4. **HRMS (ESI):** calcd. for $C_{23}H_{20}FN_2O$ $[M+H]^+$: 359.1554, found: 359.1558.

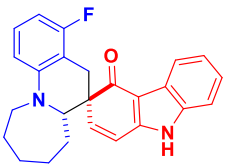
6'-bromo-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-4(9H)-one (5g)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (27.2 mg, 65% yield, dr 1:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.20 (s, 1H), 8.05 (d, *J* = 6.8 Hz, 1H), 7.51 (d, *J* = 7.2 Hz, 1H), 7.35 – 7.21 (m, 2H), 7.05 (t, *J* = 7.9 Hz, 1H), 6.98 (d, *J* = 9.8 Hz, 1H), 6.85 (d, *J* = 7.7 Hz, 1H), 6.61 (d, *J* = 8.0 Hz, 1H), 6.00 (d, *J* = 9.8 Hz, 1H), 3.81 – 3.73 (m, 1H), 3.50 (d, *J* = 7.2 Hz, 1H), 3.17 (t, *J* = 13.5 Hz, 2H), 2.81 (d, *J* = 16.4 Hz, 1H), 1.99 – 1.84 (m, 2H), 1.80 – 1.71 (m, 1H), 1.29 – 1.18 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 194.2, 147.0, 144.8, 139.9, 137.3, 129.1, 125.1, 124.5, 124.1, 123.1, 120.9, 119.1, 118.6, 112.8, 110.7, 110.6, 64.2, 55.4, 48.8, 47.7, 27.6, 23.6. **HRMS (ESI):** calcd. for $C_{23}H_{20}BrN_2O$ $[M+H]^+$: 419.0754, found: 419.0759.

4-fluoro-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,3'-carbazol]-4'(9'H)-one (5h)

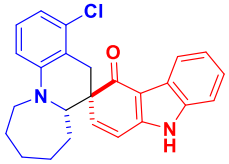


Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product

(32.8 mg, 85% yield, dr 3.3:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.16 (s, 1H), 8.10 – 8.02 (m, 1H), 7.51 (d, *J* = 7.4 Hz, 1H), 7.31 – 7.21 (m, 2H), 7.15 – 7.07 (m, 1H), 6.97 (d, *J* = 9.9 Hz, 1H), 6.67 (d, *J* = 8.4 Hz, 1H), 6.49 (t, *J* = 8.8 Hz, 1H), 6.24 (d, *J* = 9.9 Hz, 1H), 3.53 (t, *J* = 10.3 Hz, 2H), 3.39 (dd, *J* = 16.7, 9.1 Hz, 1H), 3.22 (d, *J* = 16.7 Hz, 1H), 2.60 (d, *J* = 16.4 Hz, 1H), 1.85 – 1.76 (m, 1H), 1.70 (d, *J* = 5.4 Hz, 1H), 1.63 – 1.50 (m, 3H), 1.31 (dd, *J* = 14.3, 7.4 Hz, 3H); **^{13}C NMR** (126 MHz, DMSO) δ 195.2, 161.3 (d, *J* = 240.6 Hz), 148.9 (d, *J* = 7.2 Hz), 147.0, 140.8, 137.3, 127.8 (d, *J* = 10.1 Hz), 124.4, 124.0, 123.0, 120.9, 117.4, 112.8, 111.2, 110.5, 108.4 (d, *J* = 19.5 Hz), 103.3 (d, *J* = 22.0 Hz), 66.2, 50.9, 50.7, 31.7, 30.5, 29.0, 28.5, 26.7. **HRMS (ESI):** calcd. for $C_{25}H_{24}FN_2O$ $[M+H]^+$: 387.1867, found: 387.1871.

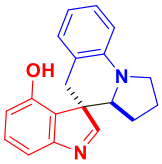
4-chloro-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,3'-carbazol]-4'(9'H)-one (5i)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (31.4 mg, 78% yield, dr 3.3:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 12.18 (s, 1H), 8.14 – 8.00 (m, 1H), 7.51 (d, *J* = 7.2 Hz, 1H), 7.30 – 7.22 (m, 2H), 7.12 (t, *J* = 8.1 Hz, 1H), 6.98 (d, *J* = 9.9 Hz, 1H), 6.83 (d, *J* = 8.4 Hz, 1H), 6.79 (d, *J* = 7.8 Hz, 1H), 6.24 (d, *J* = 9.9 Hz, 1H), 3.53 (dd, *J* = 15.3, 6.4 Hz, 1H), 3.47 (dd, *J* = 9.4, 4.7 Hz, 1H), 3.39 (dd, *J* = 20.1, 10.8 Hz, 2H), 3.26 (d, *J* = 17.0 Hz, 1H), 2.66 (d, *J* = 17.0 Hz, 1H), 1.85 – 1.78 (m, 1H), 1.74 – 1.67 (m, 1H), 1.64 – 1.57 (m, 2H), 1.57 – 1.50 (m, 1H), 1.34 – 1.25 (m, 3H); **^{13}C NMR** (126 MHz, DMSO) δ 195.0, 146.9, 140.8, 137.3, 128.0, 124.0, 123.0, 120.9, 117.7, 117.4, 113.7, 112.7, 66.0, 51.9, 50.8, 36.7, 30.5, 28.9, 28.4, 26.6. **HRMS (ESI):** calcd. for $C_{25}H_{24}ClN_2O$ $[M+H]^+$: 403.1572, found: 403.1571.

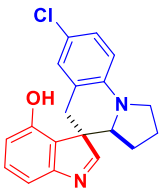
1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7a)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (18.6 mg, 64% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 9.72 (s, 1H), 7.77 (s, 1H), 7.19 (t, *J* = 7.6 Hz, 1H), 7.14 – 7.04 (m, 2H), 7.02 (d, *J* = 7.0 Hz, 1H), 6.75 (d, *J* = 8.0 Hz, 1H), 6.57 (d, *J* = 6.8 Hz, 2H), 4.45 – 4.31 (m, 1H), 3.86 (d, *J* = 16.2 Hz, 1H), 3.42 (s, 1H), 3.27 (q, *J* = 7.9 Hz, 1H), 2.45 (d, *J* = 16.2 Hz, 1H), 1.86 (s, 2H), 1.54 (d, *J* = 5.7 Hz, 1H), 0.92 – 0.80 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 175.4, 157.2, 154.1, 144.2, 129.5, 128.7, 127.9, 125.1, 119.7, 115.8, 114.4, 112.7, 111.4, 59.4, 58.0, 47.5, 32.4, 27.7, 23.2. **HRMS (ESI):** calcd. for $C_{19}H_{19}N_2O$ $[M+H]^+$: 291.1492, found: 291.1491.

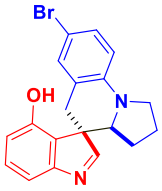
7'-chloro-1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7b)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (26.2 mg, 81% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 9.75 (s, 1H), 7.76 (s, 1H), 7.19 (t, *J* = 7.8 Hz, 1H), 7.16 – 7.02 (m, 3H), 6.75 (d, *J* = 8.1 Hz, 1H), 6.55 (d, *J* = 8.6 Hz, 1H), 4.37 (dd, *J* = 9.6, 6.1 Hz, 1H), 3.81 (d, *J* = 16.4 Hz, 1H), 3.42 (d, *J* = 10.4 Hz, 1H), 3.24 (q, *J* = 8.3 Hz, 1H), 2.48 (d, *J* = 16.6 Hz, 1H), 1.86 (d, *J* = 5.1 Hz, 2H), 1.58 – 1.49 (m, 1H), 0.91 – 0.80 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 175.0, 157.3, 154.1, 143.1, 129.7, 128.2, 127.5, 124.8, 121.7, 119.1, 114.5, 112.7, 112.6, 59.4, 57.6, 47.7, 32.1, 27.8, 23.3. **HRMS (ESI):** calcd. for $C_{19}H_{18}ClN_2O$ $[M+H]^+$: 325.1102, found: 325.1113.

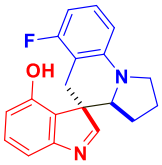
7'-bromo-1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7c)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (22.8 mg, 62% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 9.75 (s, 1H), 7.76 (s, 1H), 7.26 – 7.17 (m, 3H), 7.06 (d, *J* = 7.5 Hz, 1H), 6.75 (d, *J* = 8.1 Hz, 1H), 6.51 (d, *J* = 8.6 Hz, 1H), 4.36 (dd, *J* = 9.8, 6.0 Hz, 1H), 3.81 (d, *J* = 16.4 Hz, 1H), 3.42 (td, *J* = 9.0, 3.5 Hz, 1H), 3.23 (q, *J* = 8.5 Hz, 1H), 2.48 (d, *J* = 16.8 Hz, 1H), 1.91 – 1.82 (m, 2H), 1.57 – 1.47 (m, 1H), 0.91 – 0.81 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 174.5, 156.8, 153.6, 142.9, 130.4, 129.9, 129.2, 124.3, 121.7, 114.0, 112.6, 112.3, 106.1, 58.9, 57.0, 47.2, 31.6, 27.3, 22. 8. **HRMS (ESI):** calcd. for $C_{19}H_{18}BrN_2O$ $[M+H]^+$: 369.0597, found: 369.0605.

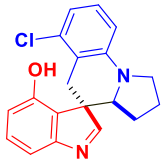
6'-fluoro-1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7d)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (23.4 mg, 76% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 9.78 (s, 1H), 7.80 (s, 1H), 7.21 (t, *J* = 7.8 Hz, 1H), 7.16 – 7.09 (m, 1H), 7.07 (d, *J* = 7.5 Hz, 1H), 6.76 (d, *J* = 8.1 Hz, 1H), 6.42 (t, *J* = 8.5 Hz, 2H), 4.33 (dd, *J* = 9.3, 6.3 Hz, 1H), 3.61 (d, *J* = 16.7 Hz, 1H), 3.43 (d, *J* = 8.0 Hz, 1H), 3.32 – 3.25 (m, 1H), 2.55 (s, 1H), 1.86 (s, 2H), 1.59 – 1.50 (m, 1H), 0.87 (t, *J* = 9.5 Hz, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 174.9, 160.9 (d, *J* = 240.7 Hz), 159.9, 157.3, 154.2, 145.9, 145.8 (d, *J* = 8.8 Hz), 129.8, 128.7 (d, *J* = 11.3 Hz), 124.7, 114.5, 112.8, 107.5, 106.3 (d, *J* = 20.3 Hz), 102.3 (d, *J* = 22.2 Hz), 58.9, 57.2, 47.8, 27.7, 25.1, 23.2. **HRMS (ESI):** calcd. for $C_{19}H_{18}FN_2O$ $[M+H]^+$: 309.1403, found: 309.1408.

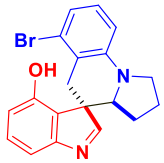
6'-chloro-1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7e)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (24.6 mg, 76% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 9.81 (s, 1H), 7.82 (s, 1H), 7.21 (t, *J* = 7.9 Hz, 1H), 7.10 (dd, *J* = 17.9, 7.9 Hz, 2H), 6.78 (d, *J* = 8.1 Hz, 1H), 6.68 (d, *J* = 7.8 Hz, 1H), 6.55 (d, *J* = 8.2 Hz, 1H), 4.33 (dd, *J* = 9.6, 6.2 Hz, 1H), 3.67 (d, *J* = 17.0 Hz, 1H), 3.40 (dt, *J* = 10.5, 7.2 Hz, 1H), 3.31 (dd, *J* = 16.8, 8.2 Hz, 1H), 2.60 (d, *J* = 17.0 Hz, 1H), 1.93 – 1.76 (m, 2H), 1.62 – 1.49 (m, 1H), 0.93 – 0.79 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 175.0, 157.3, 154.2, 145.7, 133.4, 129.9, 128.8, 124.6, 117.1, 116.2, 114.6, 112.8, 110.4, 58.8, 57.9, 47.7, 30.2, 27.7, 23.3. **HRMS (ESI):** calcd. for $C_{19}H_{18}ClN_2O$ $[M+H]^+$: 325.1108, found: 325.1110.

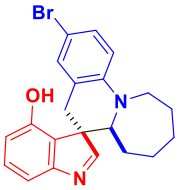
6'-bromo-1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7f)



Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (28.7 mg, 78% yield, dr >20:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 9.81 (s, 1H), 7.83 (s, 1H), 7.22 (t, *J* = 7.9 Hz, 1H), 7.11 – 7.01 (m, 2H), 6.85 (d, *J* = 7.8 Hz, 1H), 6.77 (d, *J* = 8.1 Hz, 1H), 6.60 (d, *J* = 8.2 Hz, 1H), 4.33 (dd, *J* = 9.5, 6.3 Hz, 1H), 3.65 (d, *J* = 16.9 Hz, 1H), 3.42 (dd, *J* = 11.9, 8.7 Hz, 1H), 3.34 – 3.28 (m, 1H), 2.55 (d, *J* = 16.9 Hz, 1H), 1.92 – 1.83 (m, 2H), 1.57 (dd, *J* = 10.6, 5.2 Hz, 1H), 0.92 – 0.82 (m, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 174.8, 157.1, 154.0, 145.7, 129.7, 129.1, 124.4, 124.3, 119.1, 118.4, 114.4, 112.6, 110.8, 58.7, 58.0, 47.5, 33.0, 27.5, 23.2. **HRMS (ESI):** calcd. for $C_{19}H_{18}BrN_2O$ $[M+H]^+$: 369.0602, found: 369.0600.

3-bromo-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,3'-indol]-4'-ol (7g)



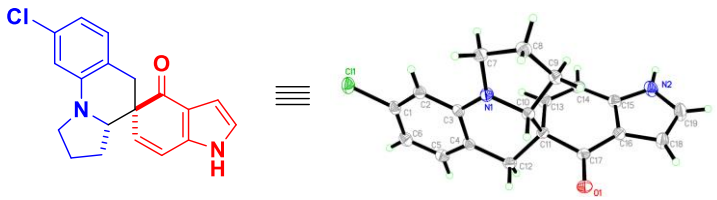
Flash column chromatography on a silica gel (ethyl acetate: petroleum ether, 1:3) afforded the product (30.0 mg, 76% yield, dr 1.5:1) as a yellow solid.

^{1}H NMR (500 MHz, DMSO) δ 9.75 (s, 1H), 7.92 (s, 1H), 7.24 – 7.15 (m, 2H), 7.11 (d, *J* = 2.2 Hz, 1H), 7.05 (d, *J* = 7.4 Hz, 1H), 6.71 (dd, *J* = 14.1, 8.5 Hz, 2H), 4.37 – 4.28 (m, 2H), 3.76 (d, *J* = 16.3 Hz, 1H), 3.40 (t, *J* = 4.8 Hz, 1H), 2.35 (d, *J* = 16.4 Hz, 1H), 1.72 (dd, *J* = 12.1, 6.1 Hz, 1H), 1.69 – 1.61 (m, 1H), 1.41 (ddd, *J* = 24.7, 13.6, 5.4 Hz, 4H), 1.31 (d, *J* = 8.6 Hz, 1H), 0.94 (dd, *J* = 9.2, 4.6 Hz, 1H); **^{13}C NMR** (126 MHz, DMSO) δ 174.9, 156.7, 153.3, 145.9, 130.3, 129.8, 129.2, 125.2, 122.8, 114.3, 113.9, 112.2, 106.8, 60.5, 59.6, 49.3, 31.6, 29.2, 29.1, 28.8, 25.7. **HRMS (ESI):** calcd. for $C_{21}H_{22}BrN_2O$ $[M+H]^+$: 396.0835, found: 396.0837.

Reference

1. Jurberg, I. D.; Peng, B.; Wçstefeld, E.; Wasserloos, M.; Maulide, N. *Angew. Chem. Int. Ed.* **2012**, *51*, 1950.
2. Huang, J.-R.; Qin, L.; Zhu, Y.-Q.; Song, Q.; Dong, L. *Chem. Commun.* **2015**, *51*, 2844.

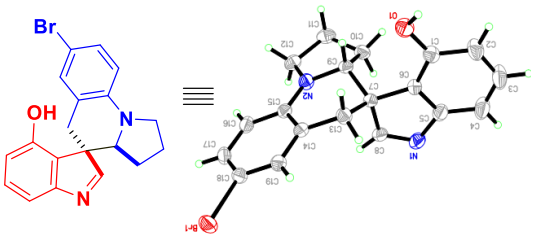
4. Crystal Structure and Data



3d (CCDC 1828278)

Table 1. Crystal data and structure refinement for **3d**.

Identification code	**3d**
Empirical formula	$C_{19}H_{17}ClN_2O$
Formula weight	324.80
Temperature	293(2) K
Wavelength	1.54184 A
Crystal system, space group	Monoclinic, P21/c
Unit cell dimensions	a = 7.5569(2) A alpha = 90 deg.
	b = 17.7897(4) A beta = 95.909(3) deg.
	c = 11.2378(3) A gamma = 90 deg.
Volume	1502.73(7) A^3
Z, Calculated density	4, 1.436 Mg/m^3
Absorption coefficient	2.292 mm^-1
F(000)	680
Crystal size	0.09 x 0.08 x 0.08 mm
Theta range for data collection	4.67 to 67.25 deg.
Limiting indices	-8<=h<=9, -13<=k<=21, -13<=l<=13
Reflections collected / unique	9001 / 2685 [R(int) = 0.0314]
Completeness to theta = 67.25	100.0 %
Max. and min. transmission	0.8379 and 0.8203
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	2685 / 0 / 208
Goodness-of-fit on F^2	1.139
Final R indices [I>2sigma(I)]	R1 = 0.0419, wR2 = 0.1237
R indices (all data)	R1 = 0.0518, wR2 = 0.1589
Largest diff. peak and hole	0.319 and -0.382 e.A^-3



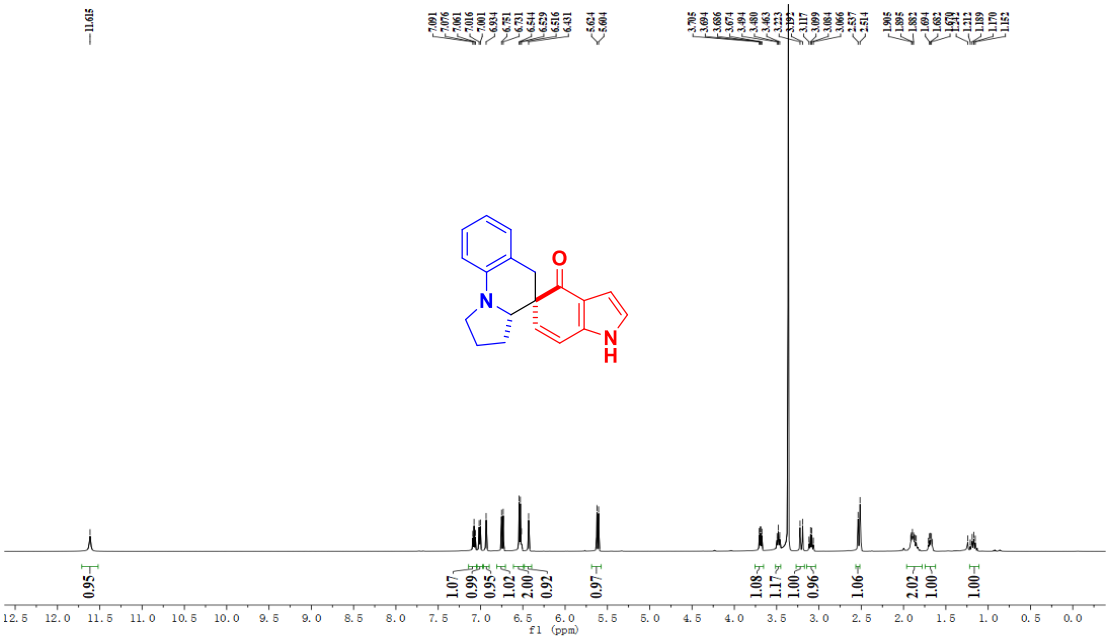
7c (CCDC1922516)

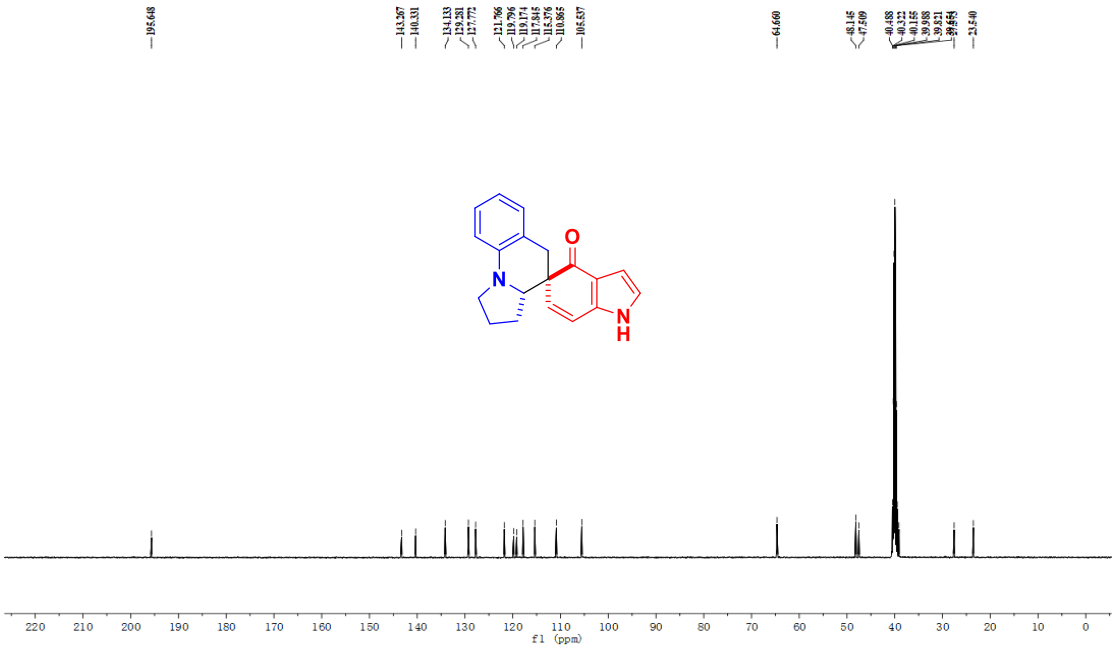
Table 1. Crystal data and structure refinement for **7c**.

Identification code	**7c**
Empirical formula	$C_{20}H_{21}BrN_2O_2$
Formula weight	401.30
Temperature	293(2) K
Wavelength	1.54178 A
Crystal system, space group	orthorhombic, Pna21
Unit cell dimensions	a = 20.4146(3) A alpha = 90 deg.
	b = 6.41670(10) A beta = 90 deg.
	c = 14.0402(2) A gamma = 90 deg.
Volume	1839.19(5) A^3
Z, Calculated density	4, 1.449 Mg/m^3
Absorption coefficient	3.168 mm^-1
F(000)	824
Crystal size	0.13 x 0.12 x 0.11 mm
Theta range for data collection	4.33 to 67.21 deg.
Limiting indices	-24<=h<=24, -7<=k<=6, -16<=l<=10
Reflections collected / unique	11838 / 2786 [R(int) = 0.0230]
Completeness to theta = 67.21	100.0 %
Max. and min. transmission	0.7220 and 0.6835
Refinement method	Full-matrix least-squares on F^2
Goodness-of-fit on F^2	1.093
Final R indices [I>2sigma(I)]	R1 = 0.0261, wR2 = 0.0698
R indices (all data)	R1 = 0.0271, wR2 = 0.0705
Largest diff. peak and hole	0.183 and -0.423 e.A^-3

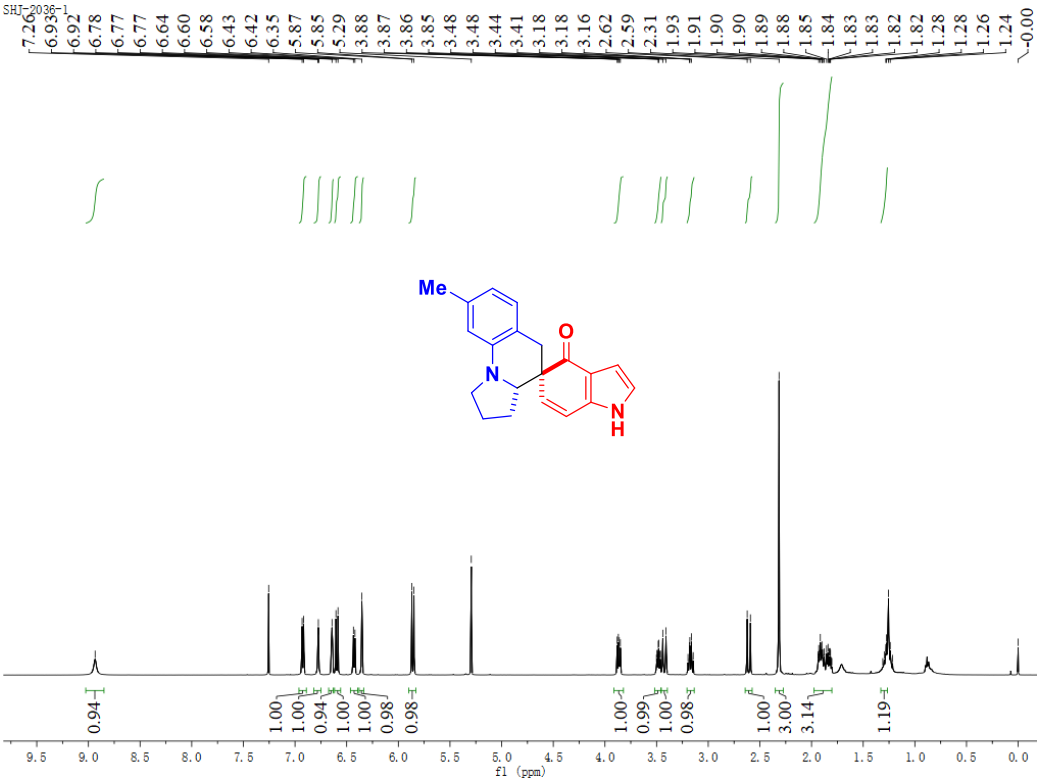
6. ^{1}H and ^{13}C-NMR Spectra

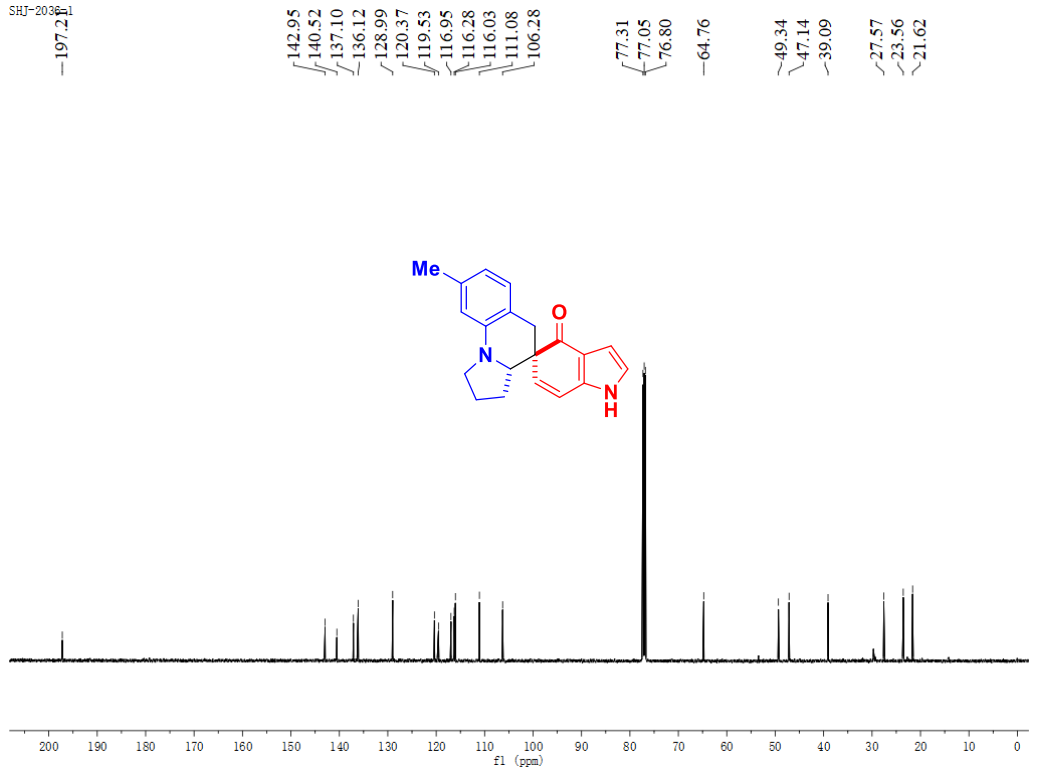
1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3a)



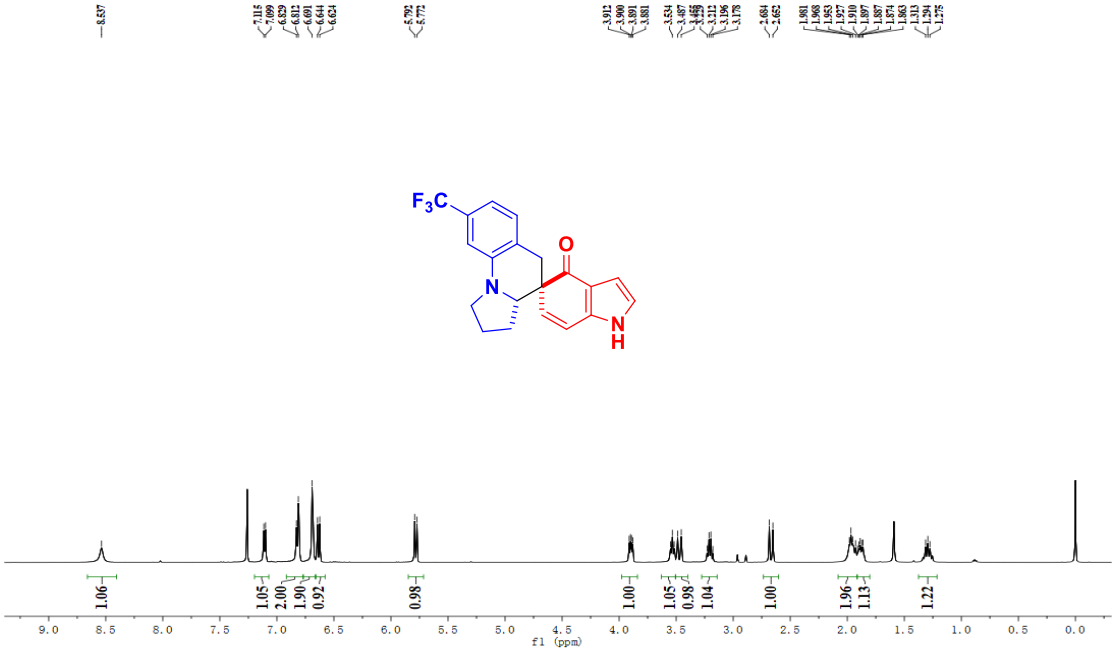


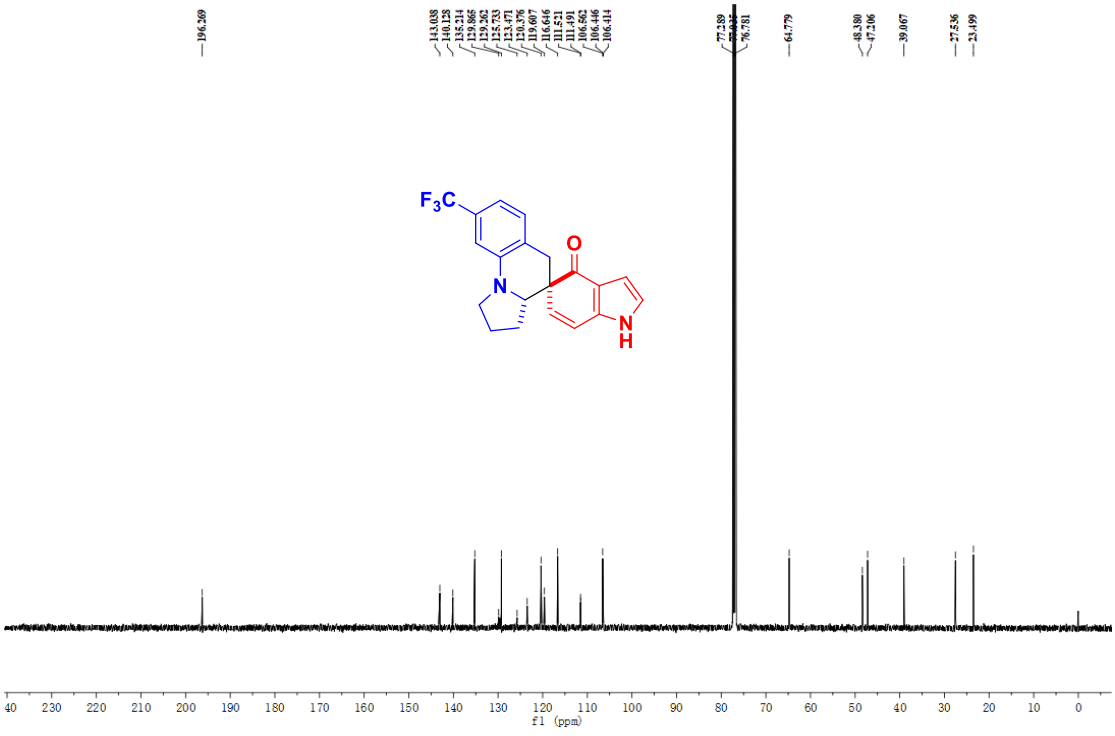
8'-methyl-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3b)



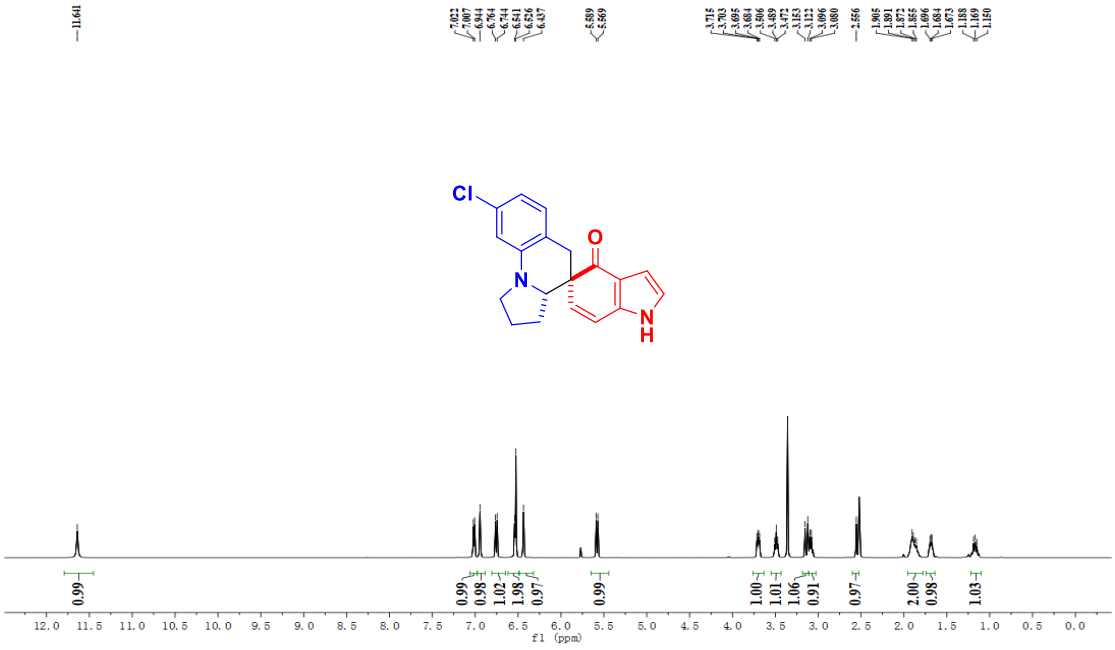


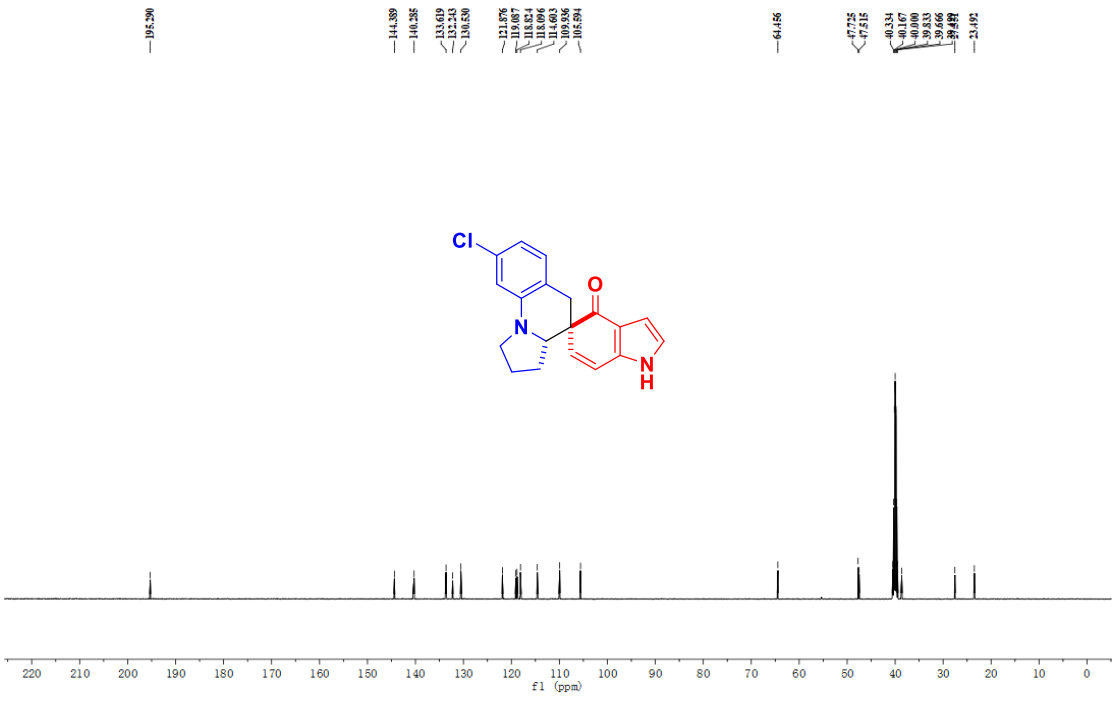
8'-(trifluoromethyl)-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3c)



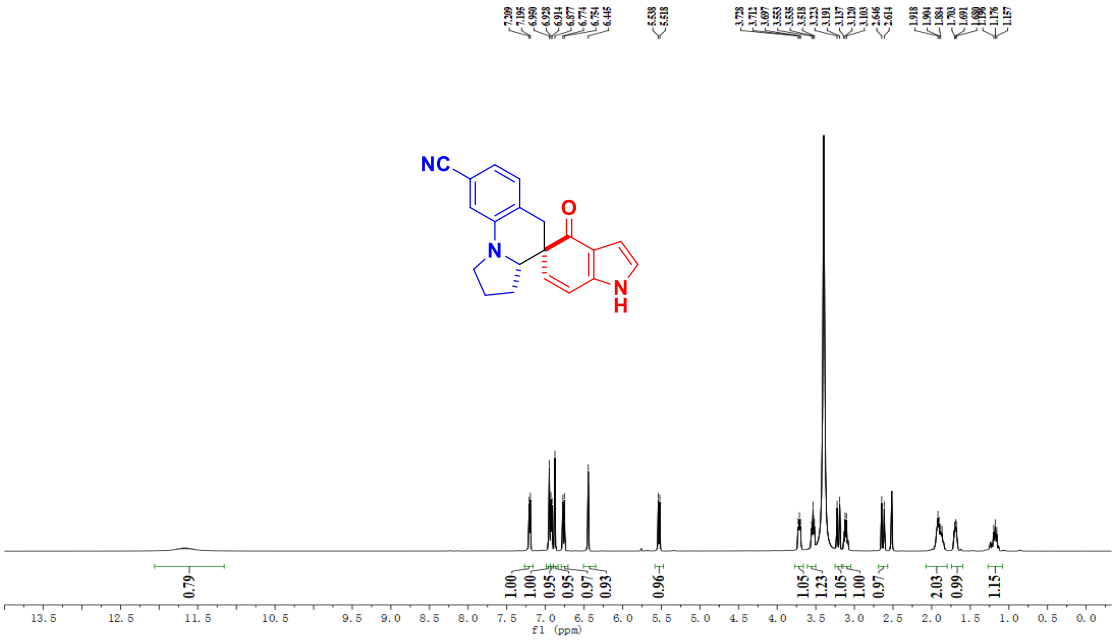


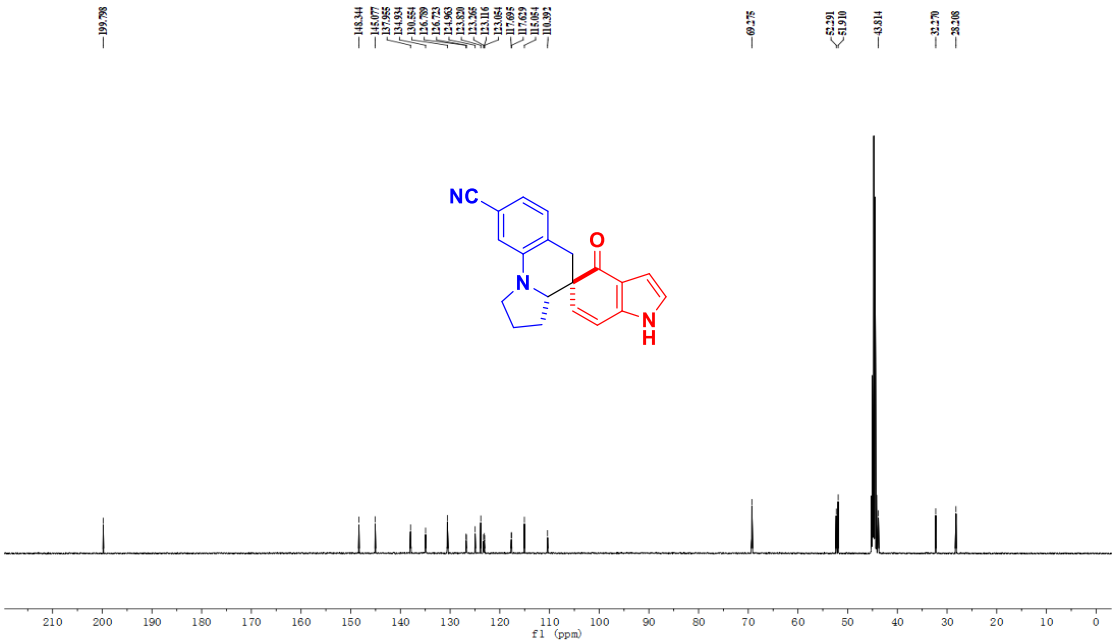
8'-chloro-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3d)



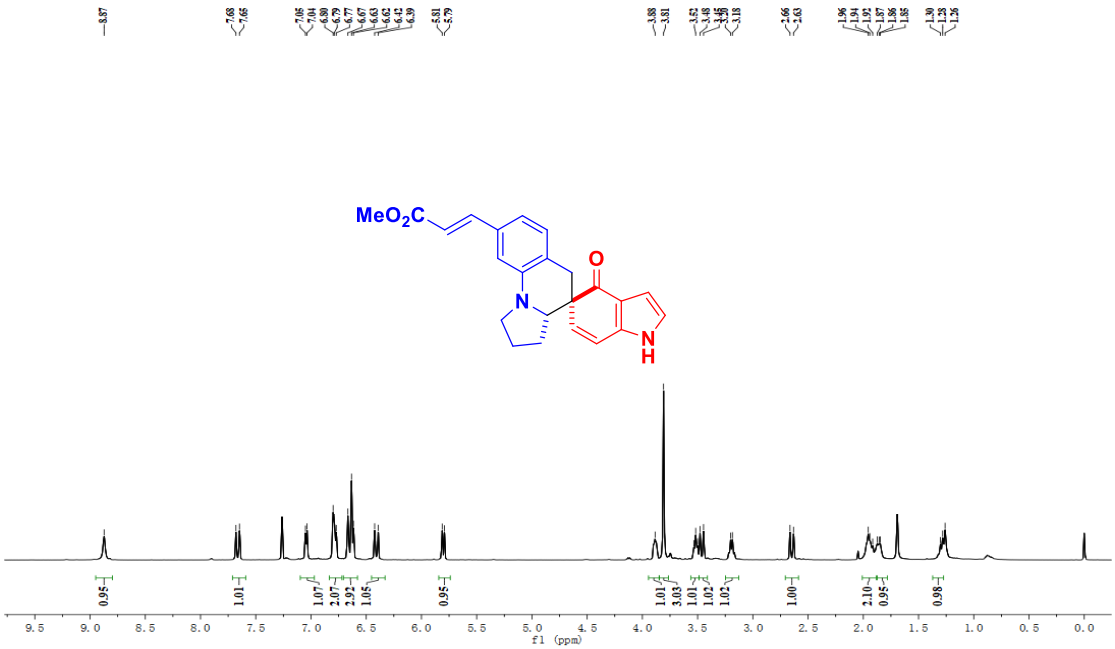


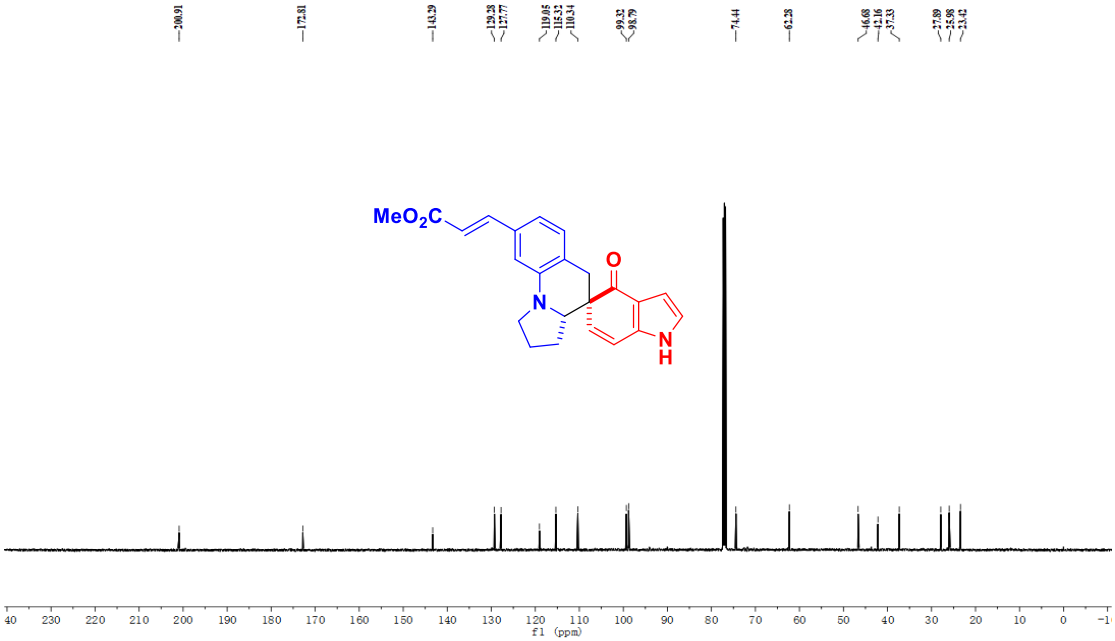
4-oxo-1,1',2',3',3a',4-hexahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinoline]-8'-carbonitrile (3e)



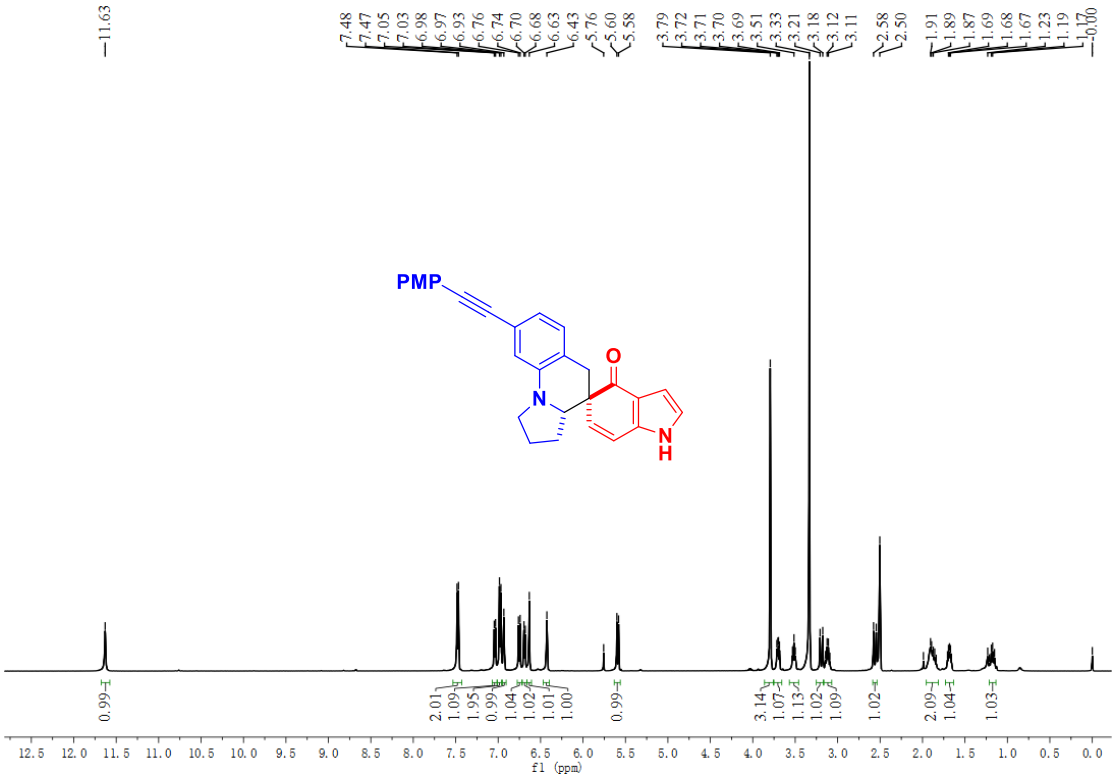


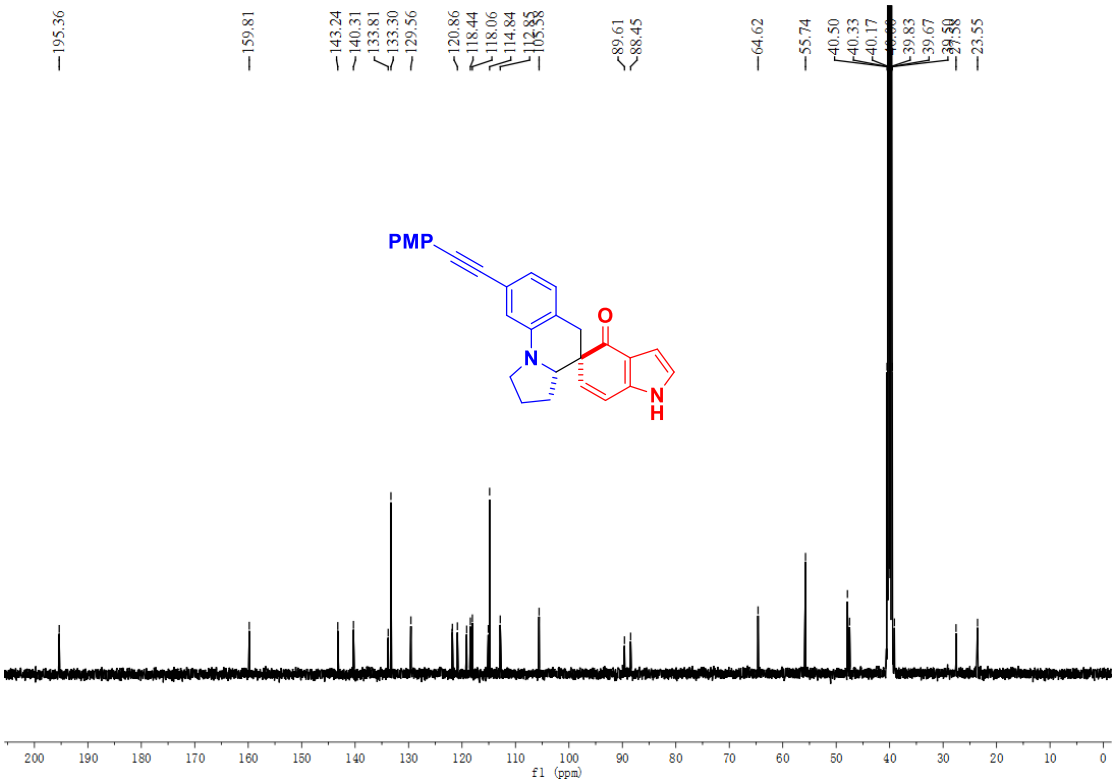
methyl(*E*)-3-(4-oxo-1,1',2',3',3a',4-hexahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-8'-yl)acrylate (3f)



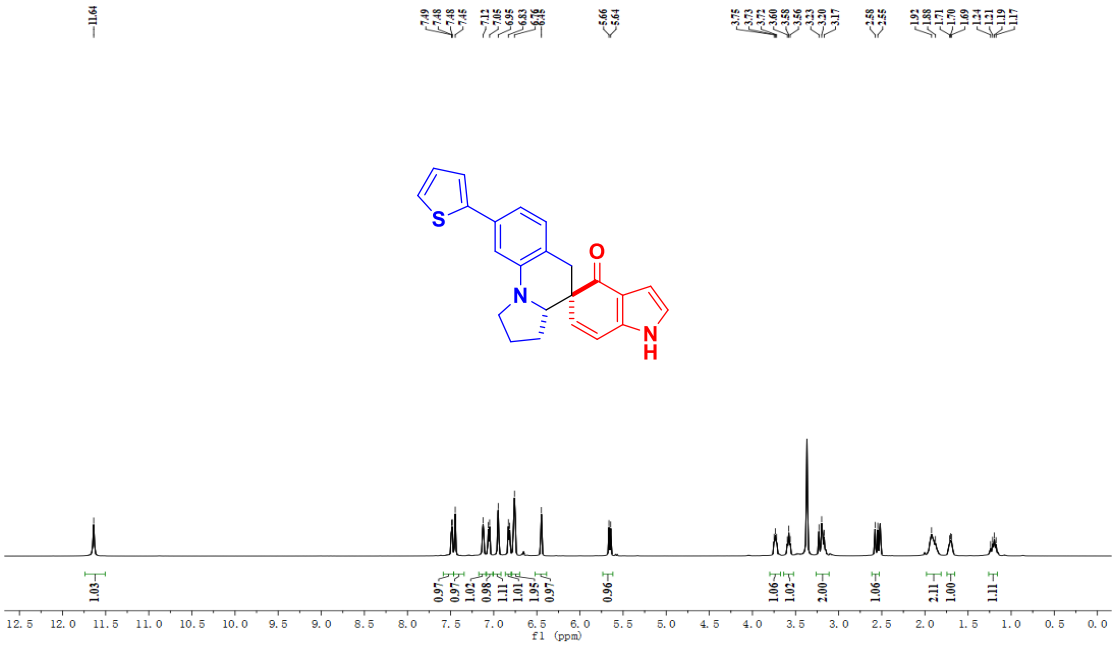


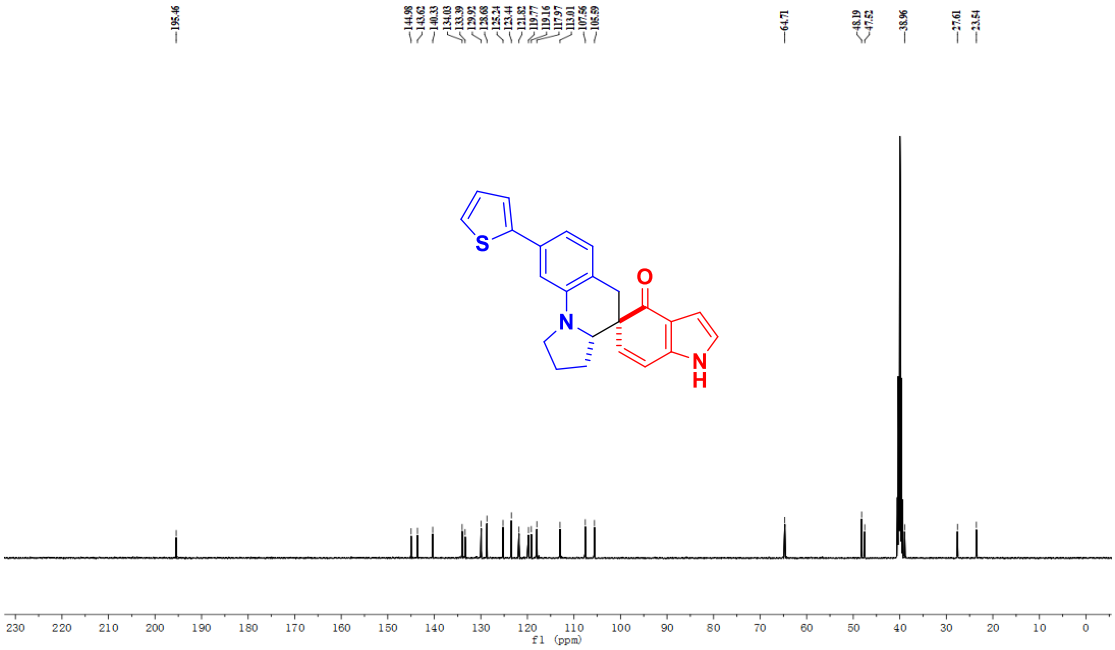
8'-((4-methoxyphenyl)ethynyl)-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3g)



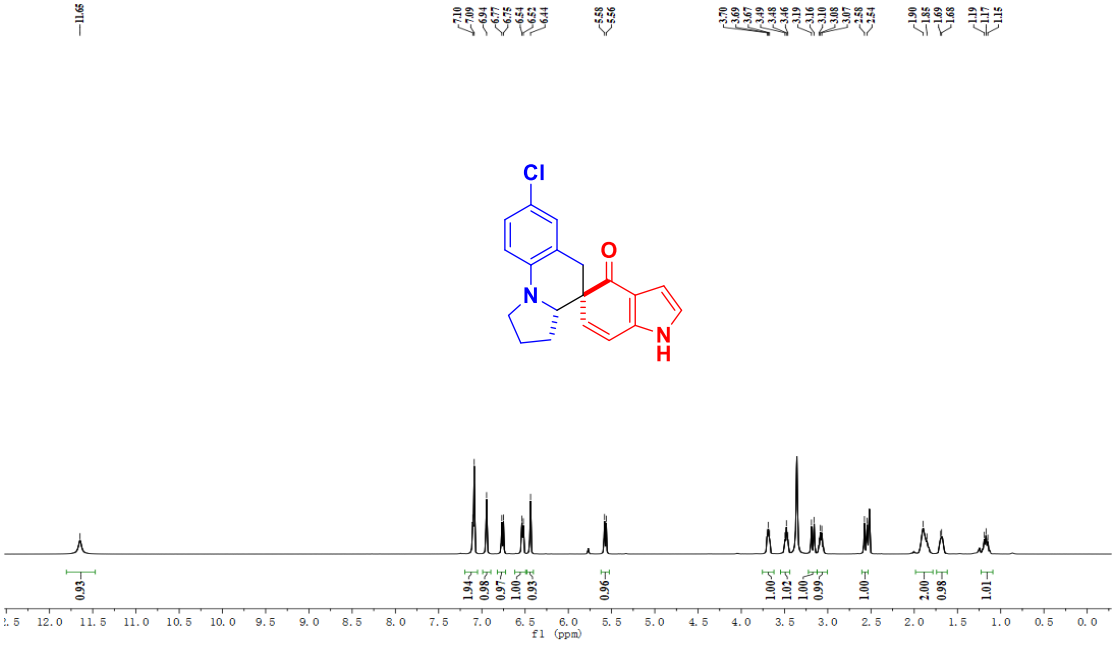


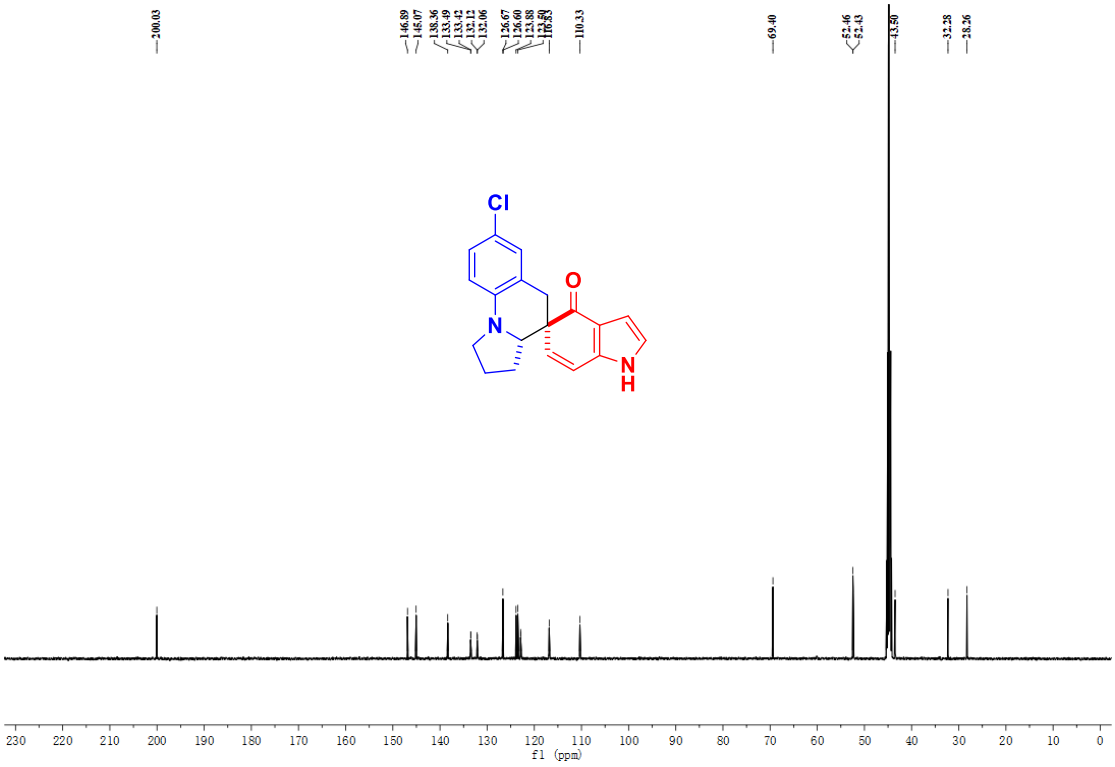
8'-(thiophen-2-yl)-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3h)



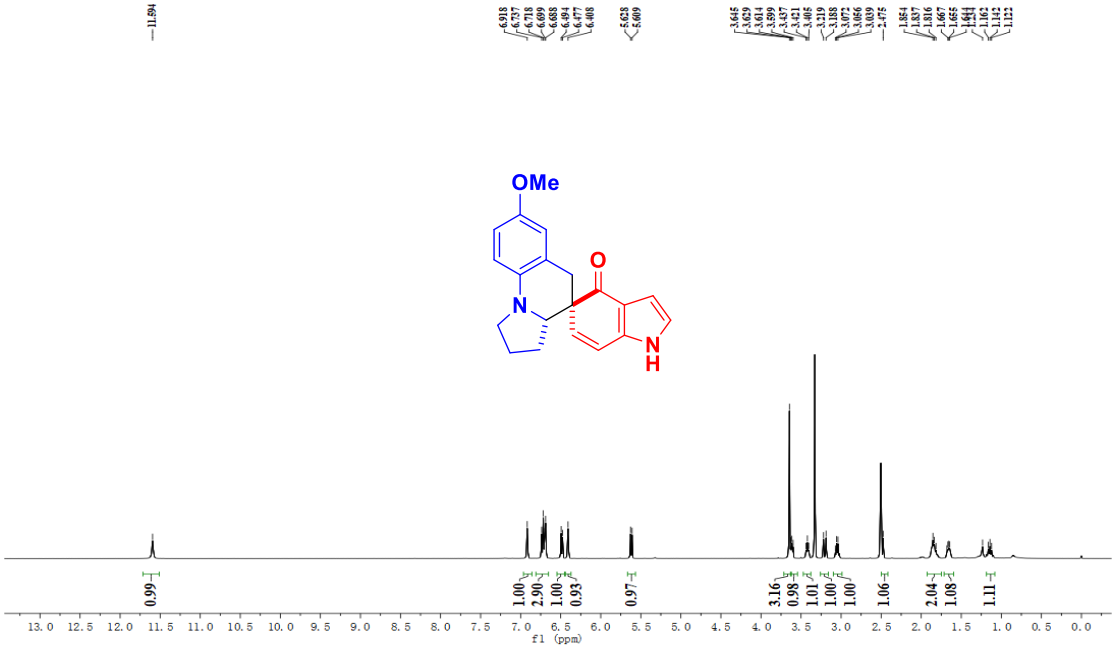


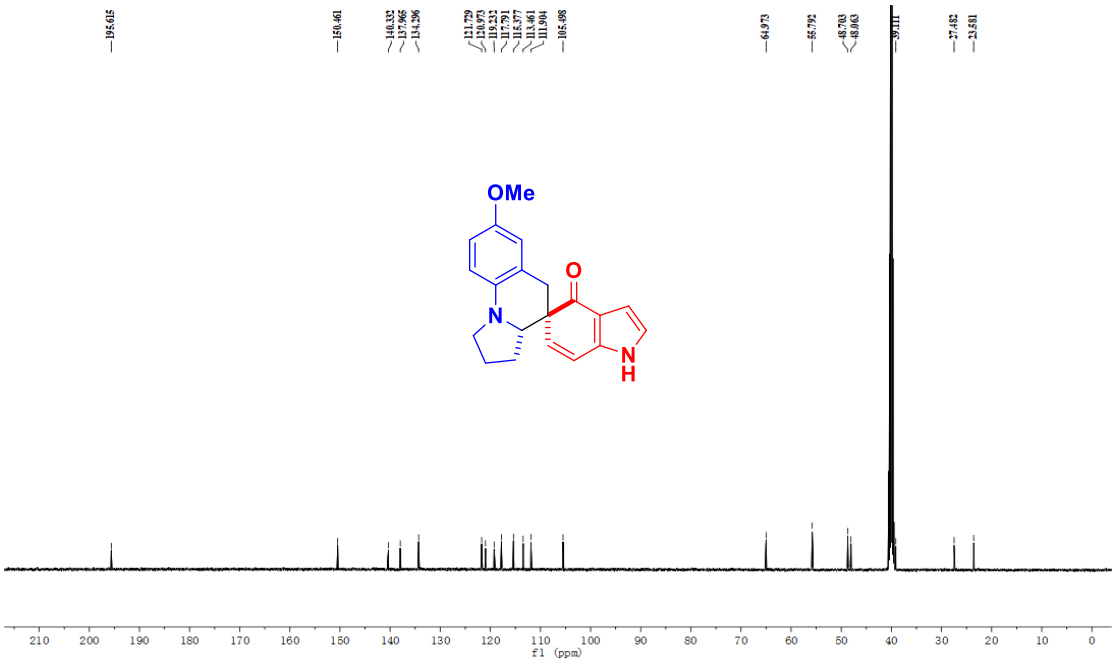
7'-chloro-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3i)



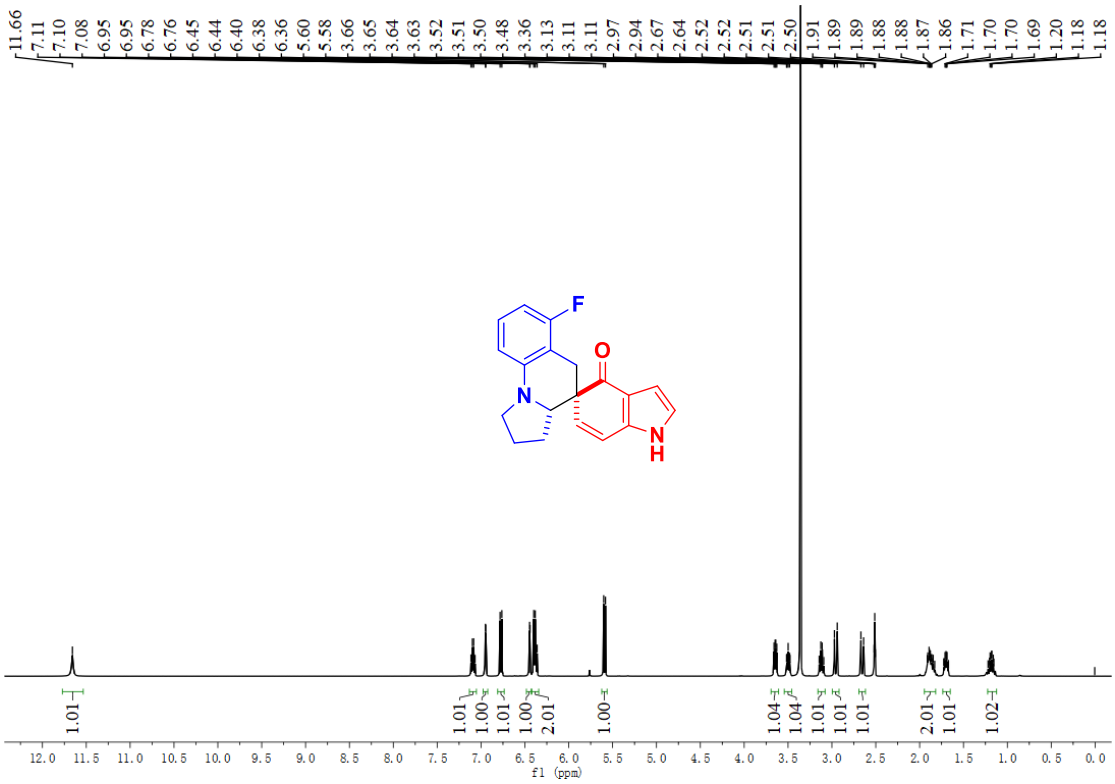


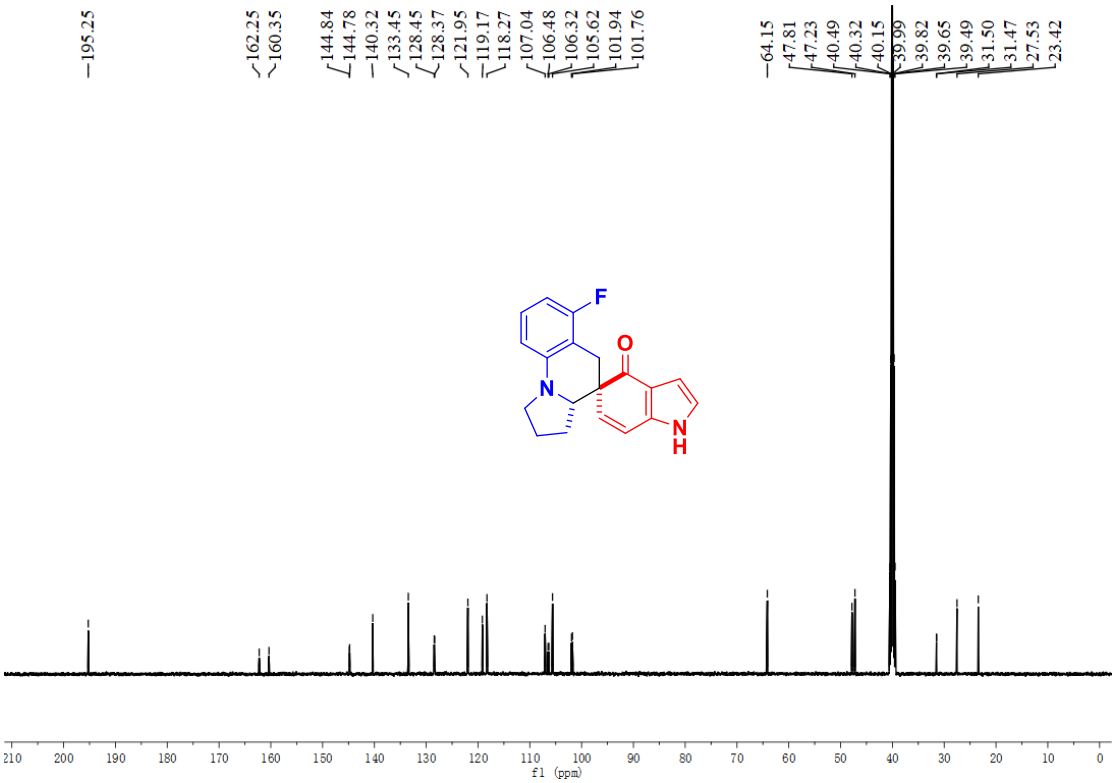
7'-methoxy-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3j)



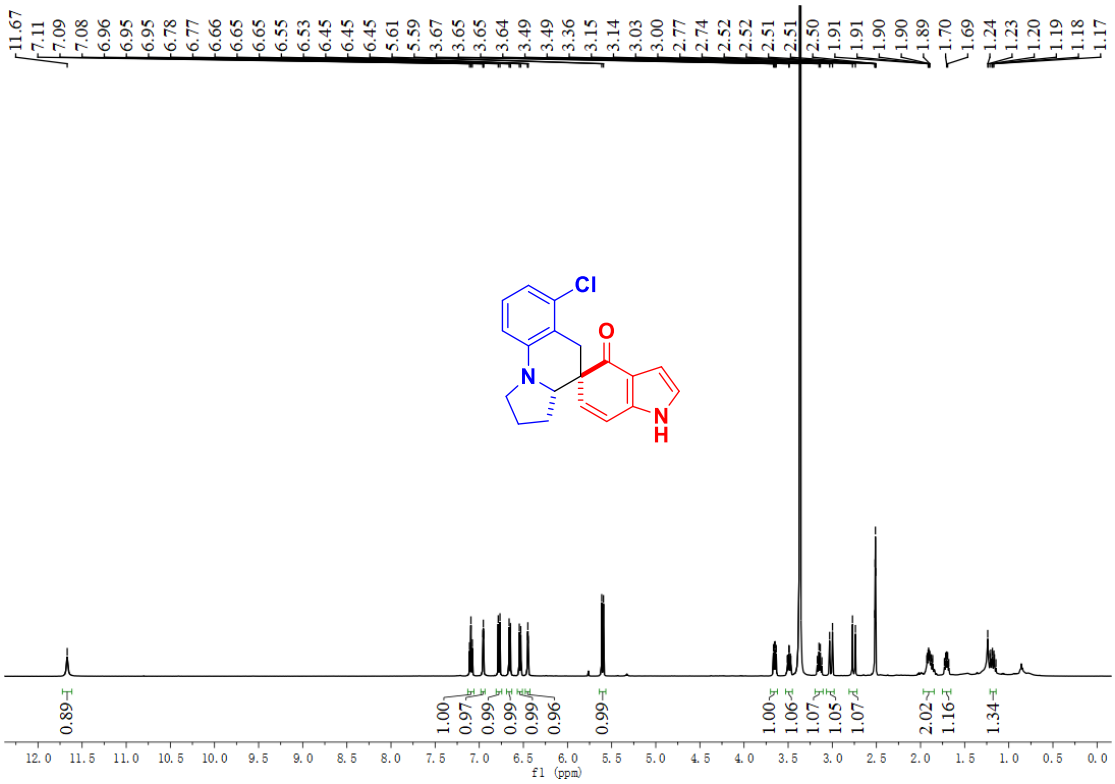


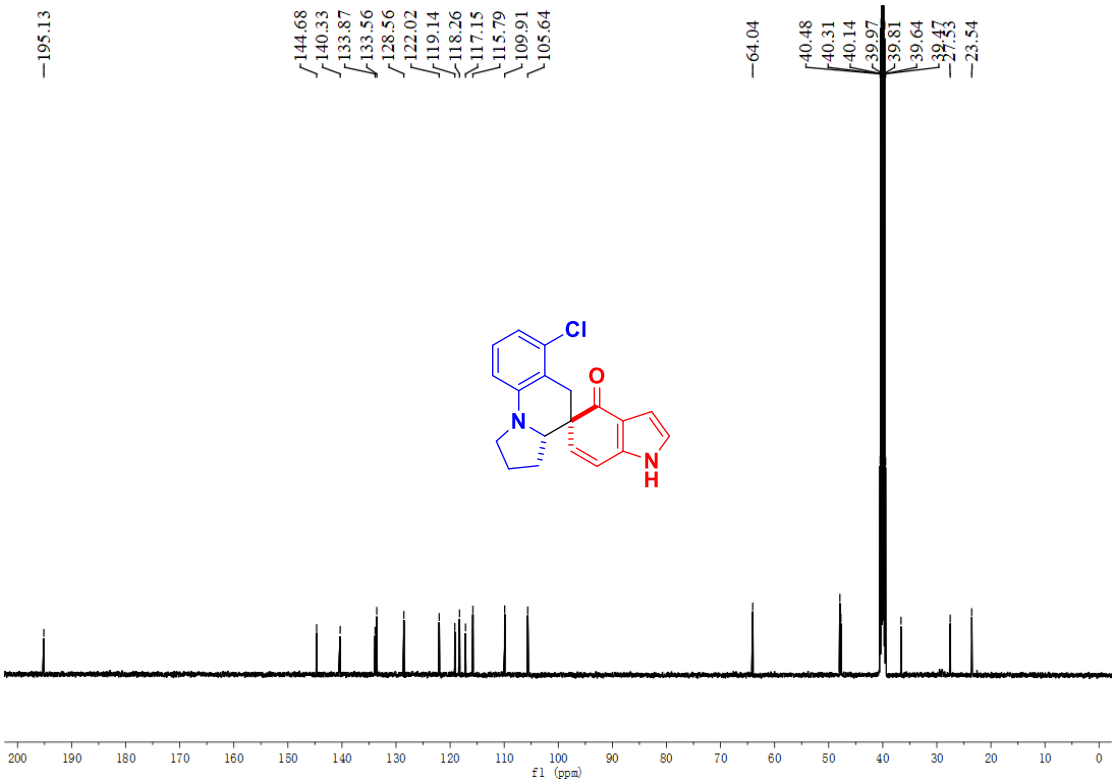
6'-fluoro-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3k)



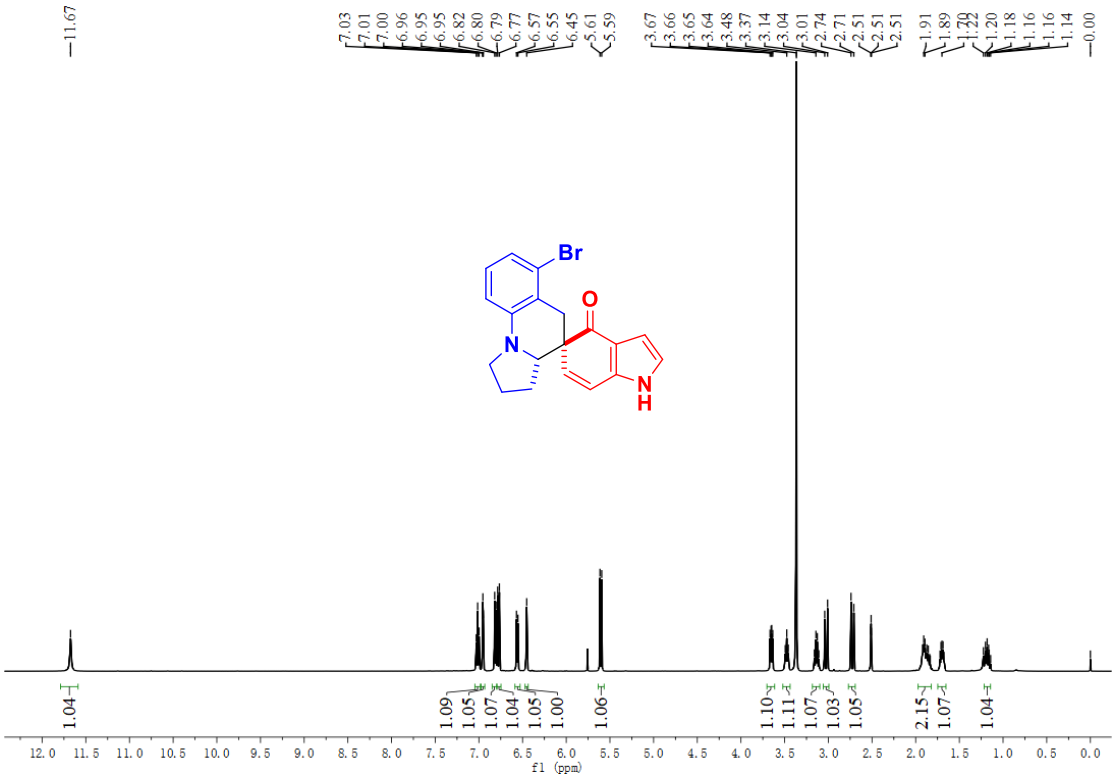


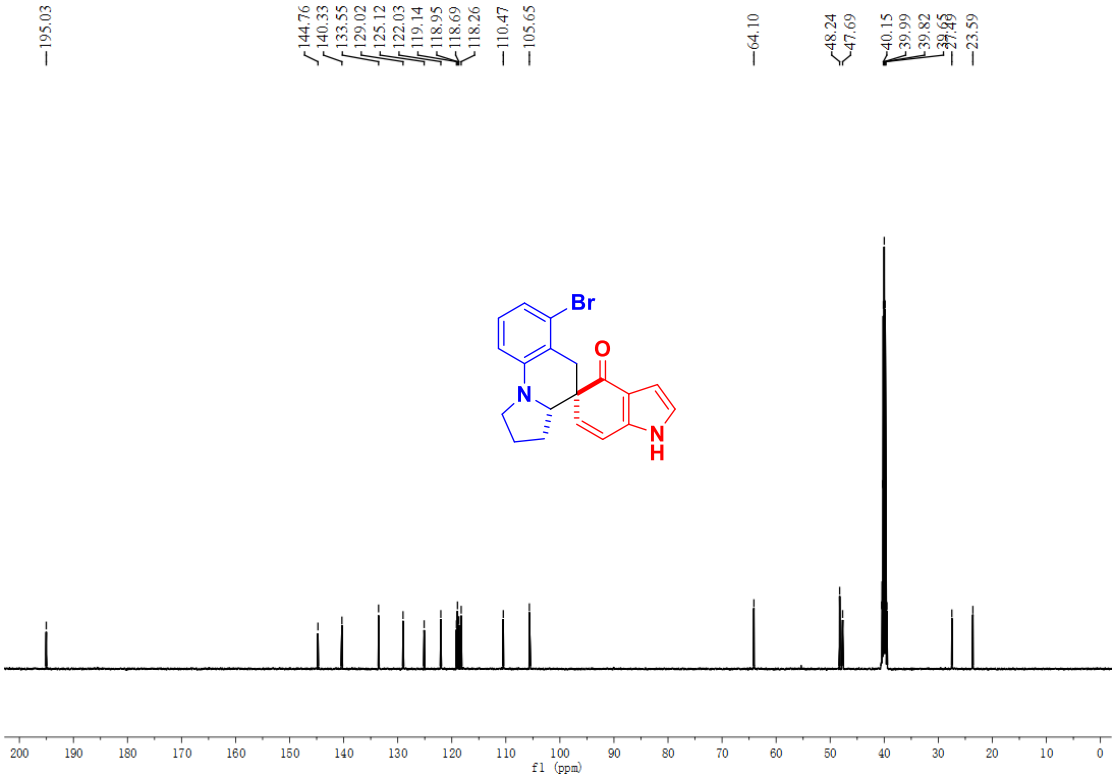
6'-chloro-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3l)



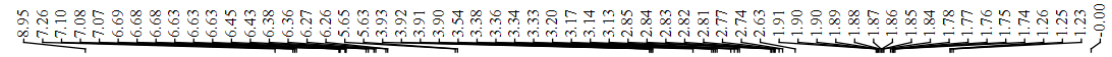


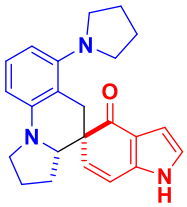
6'-bromo-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3m)

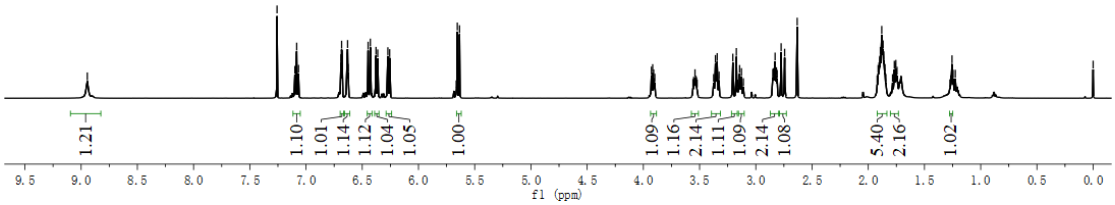


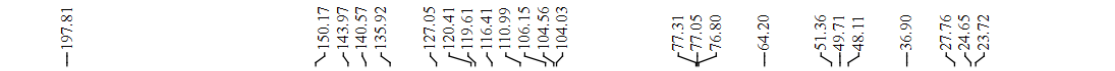


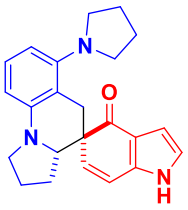
6'-(pyrrolidin-1-yl)-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3n)

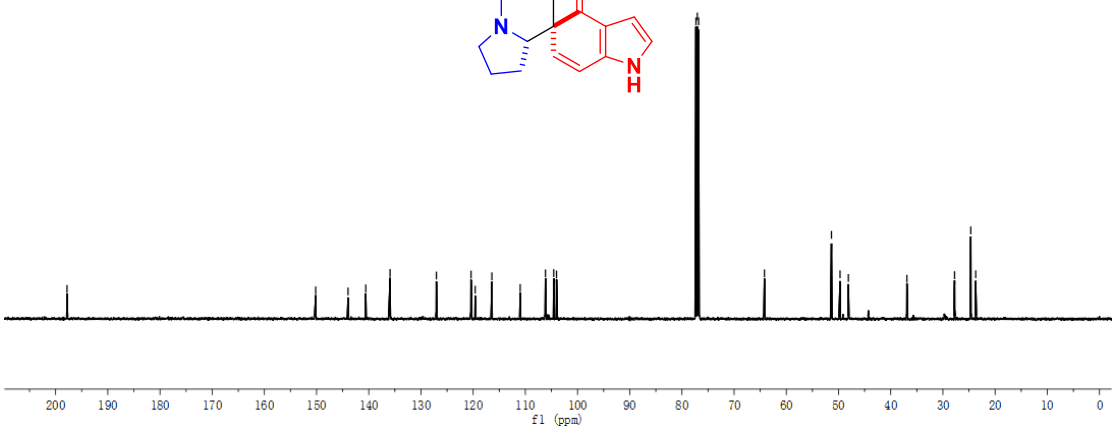




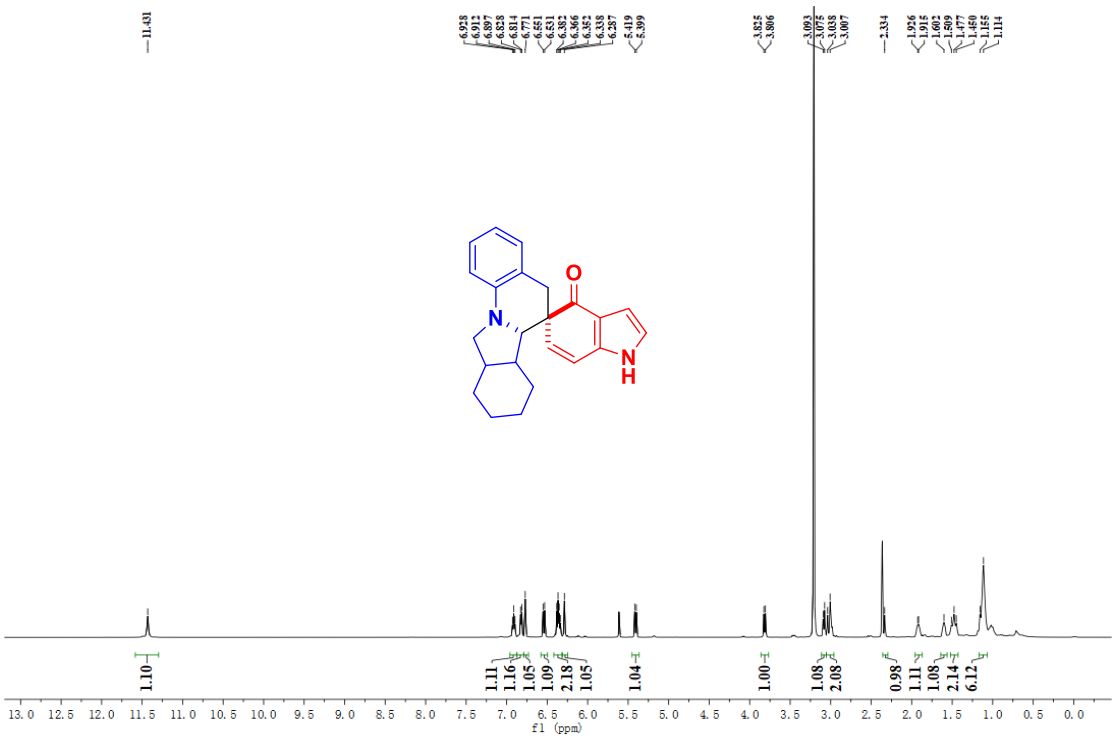


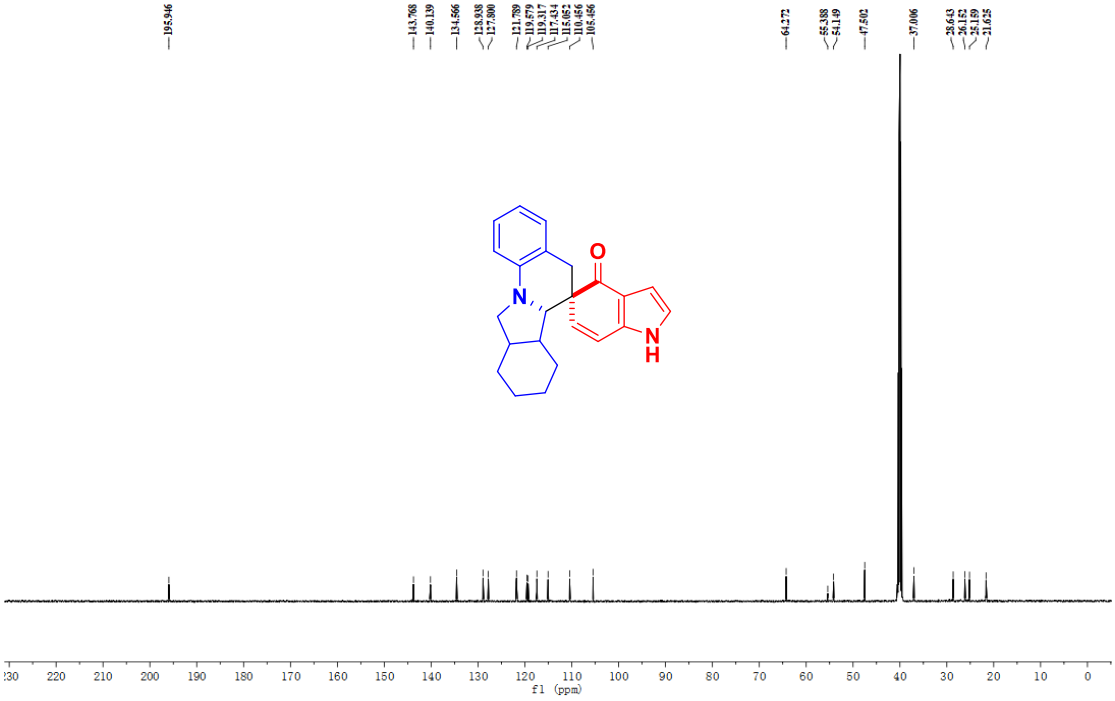




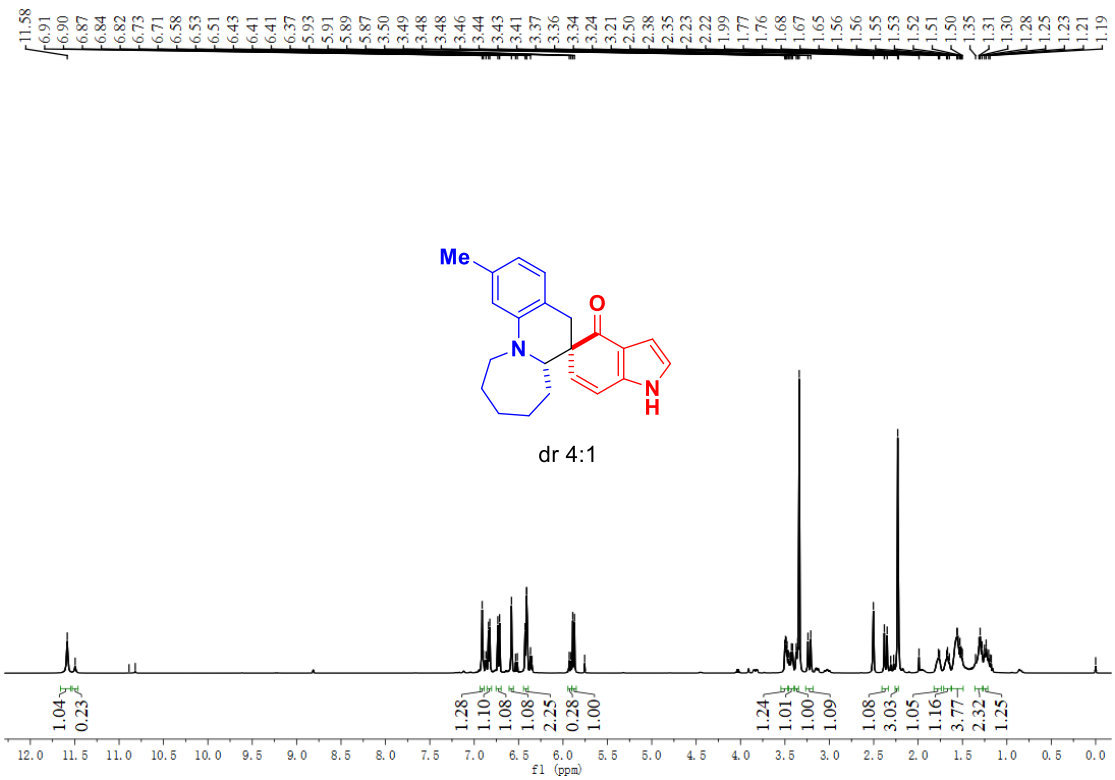


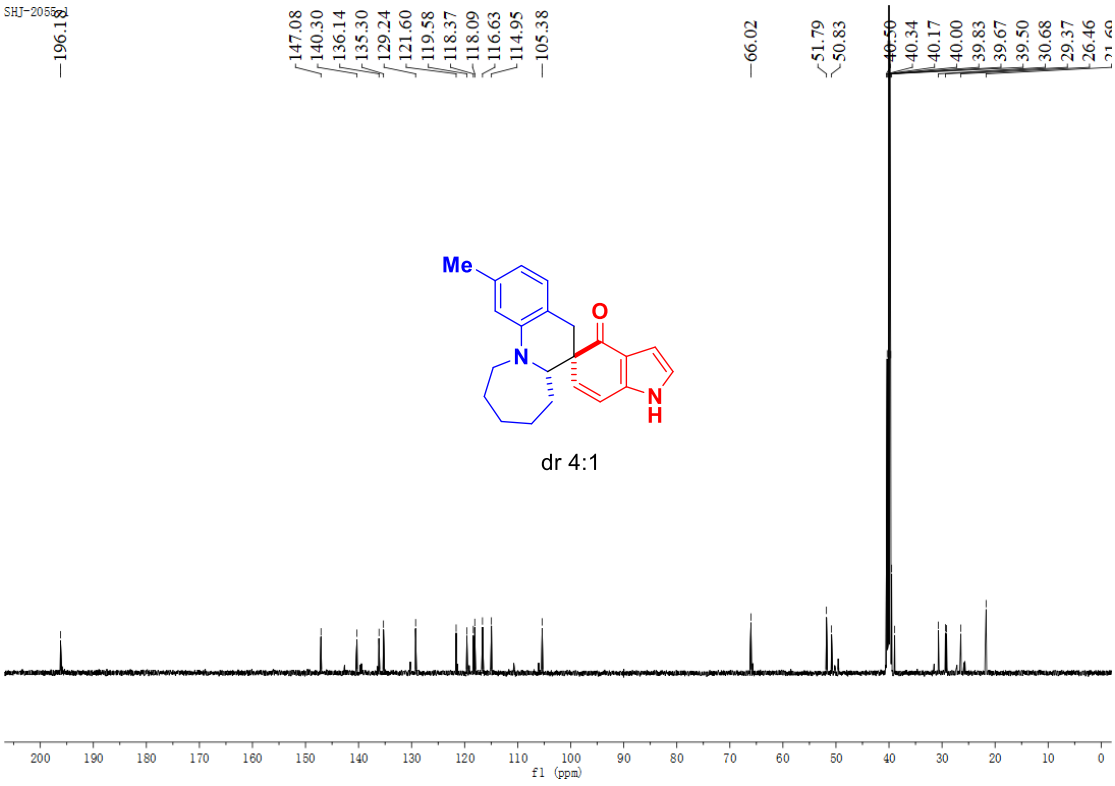
6a',6b',7',8',9',10',10a',11'-octahydro-5'*H*-spiro[indole-5,6'-isoindolo[2,1-*a*]quinolin]-4(1*H*)-one (3o)



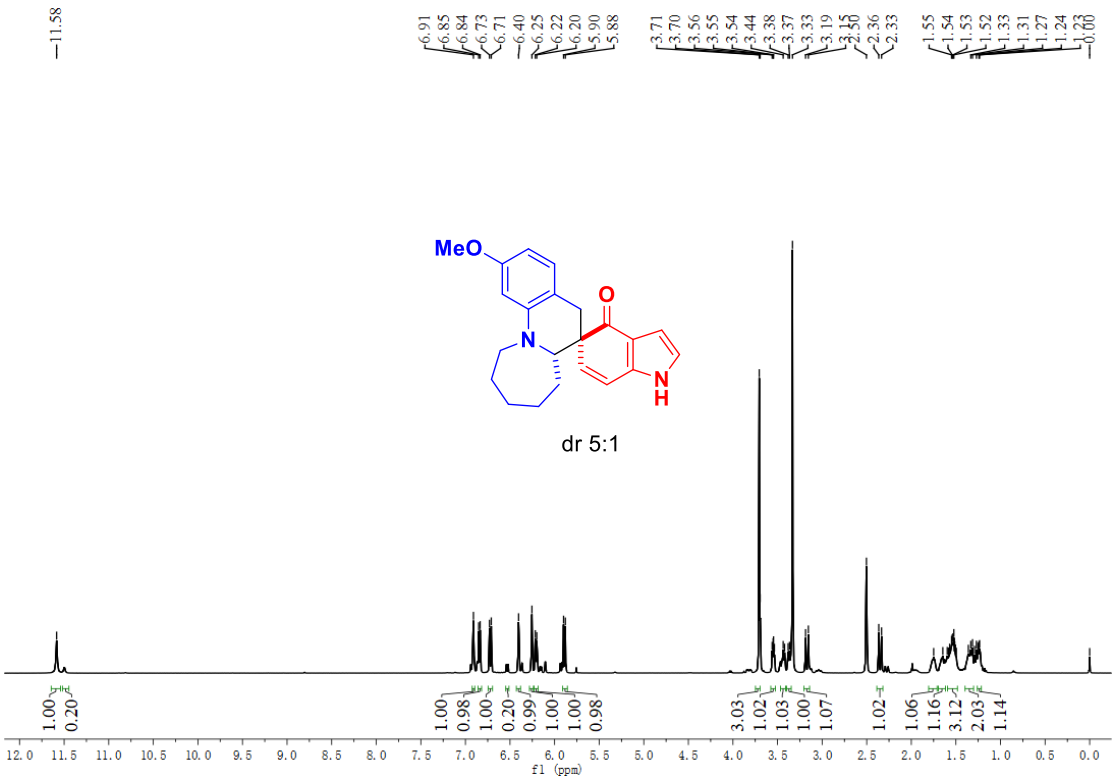


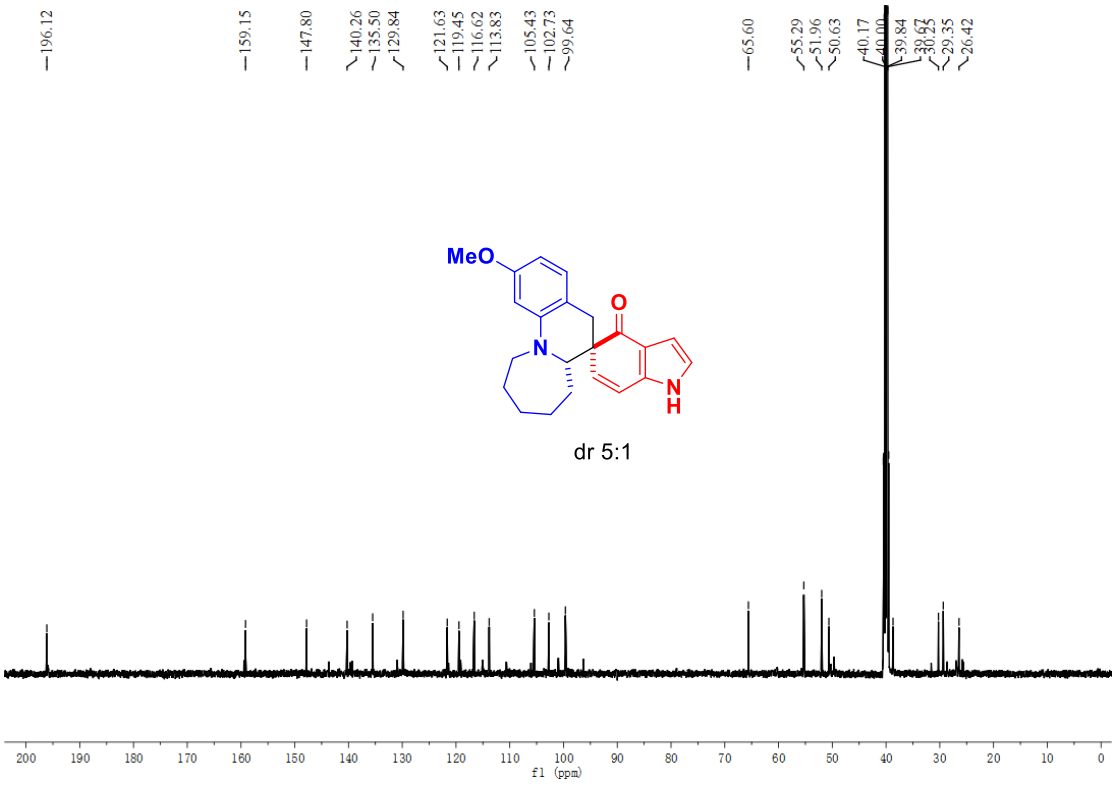
2-methyl-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,5'-indol]-4'(1'H)-one (3p)



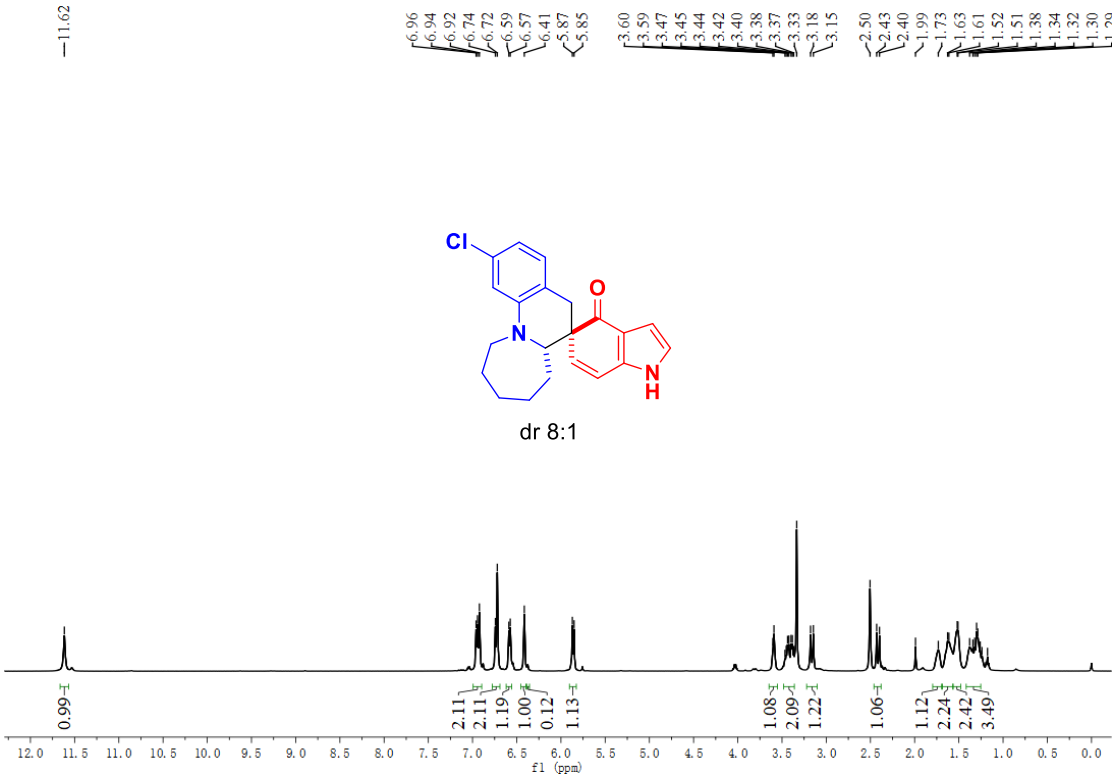


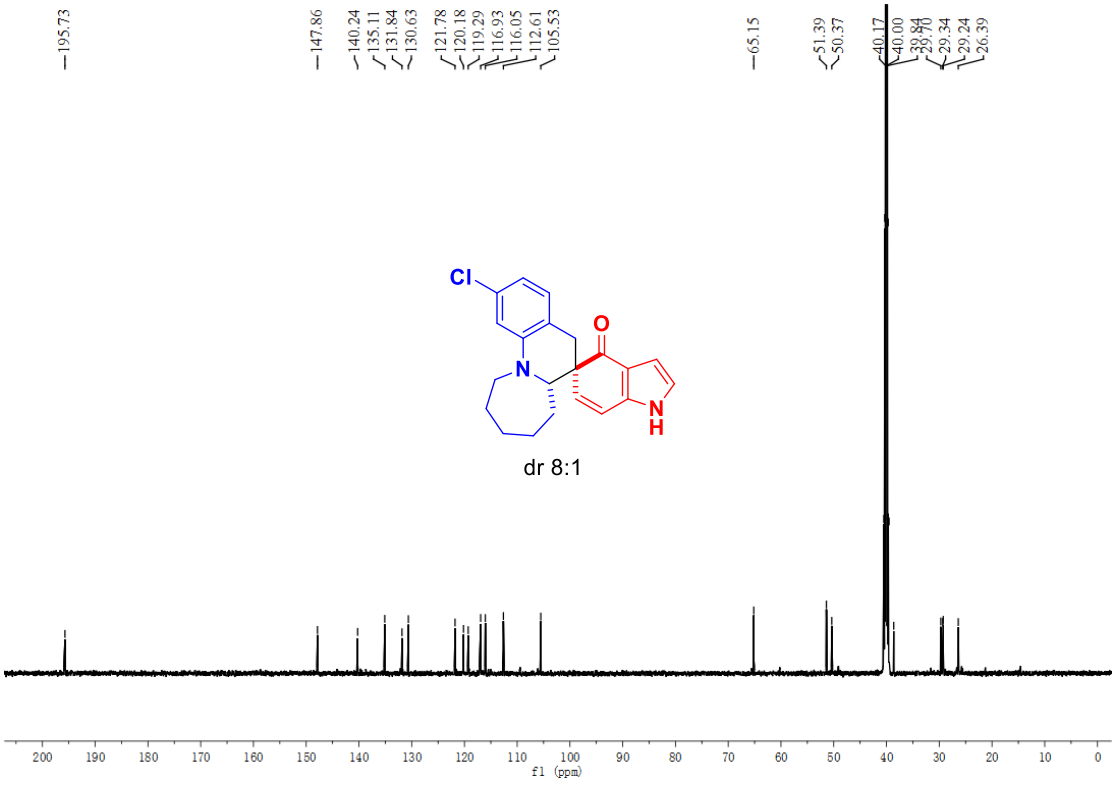
2-methoxy-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,5'-indol]-4'(1'H)-one (3q)



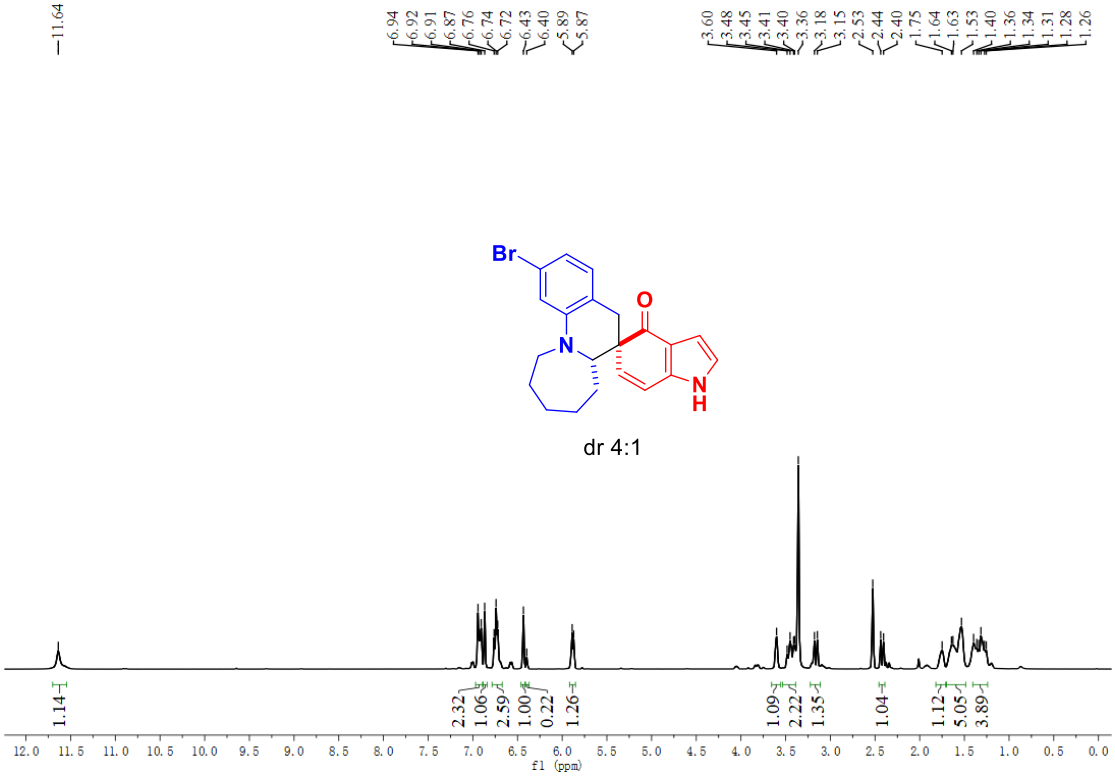


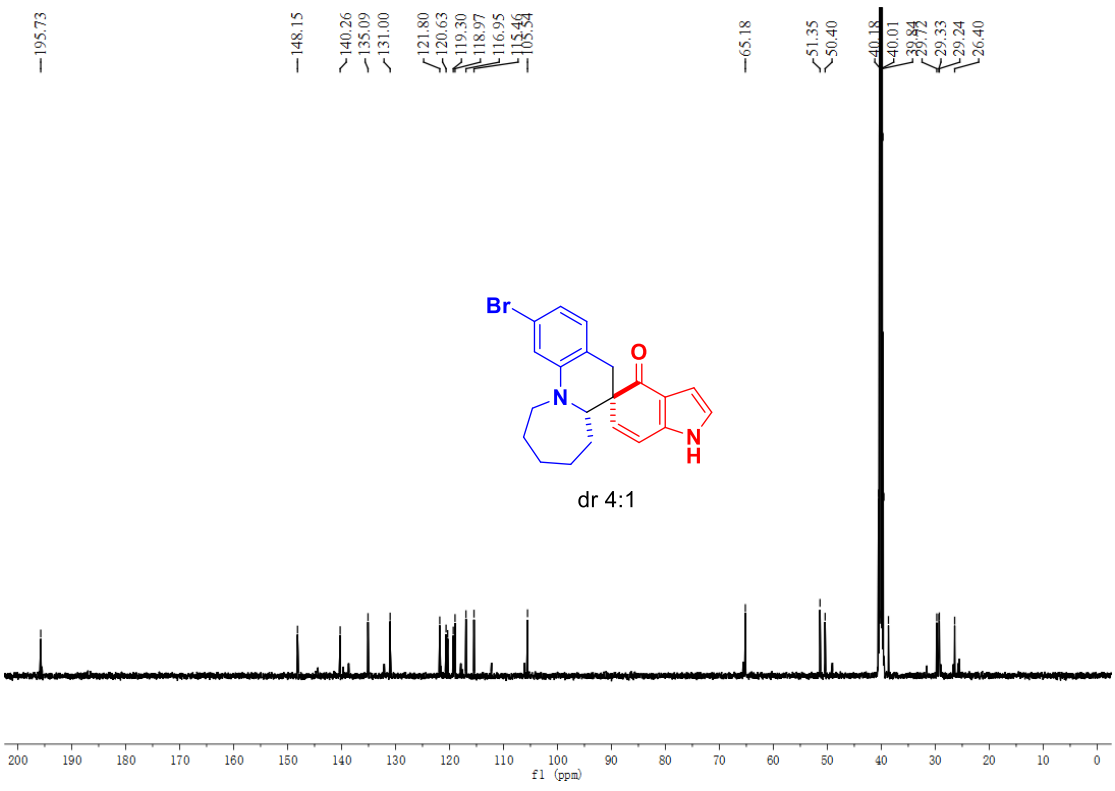
2-chloro-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,5'-indol]-4'(1'H)-one (3r)



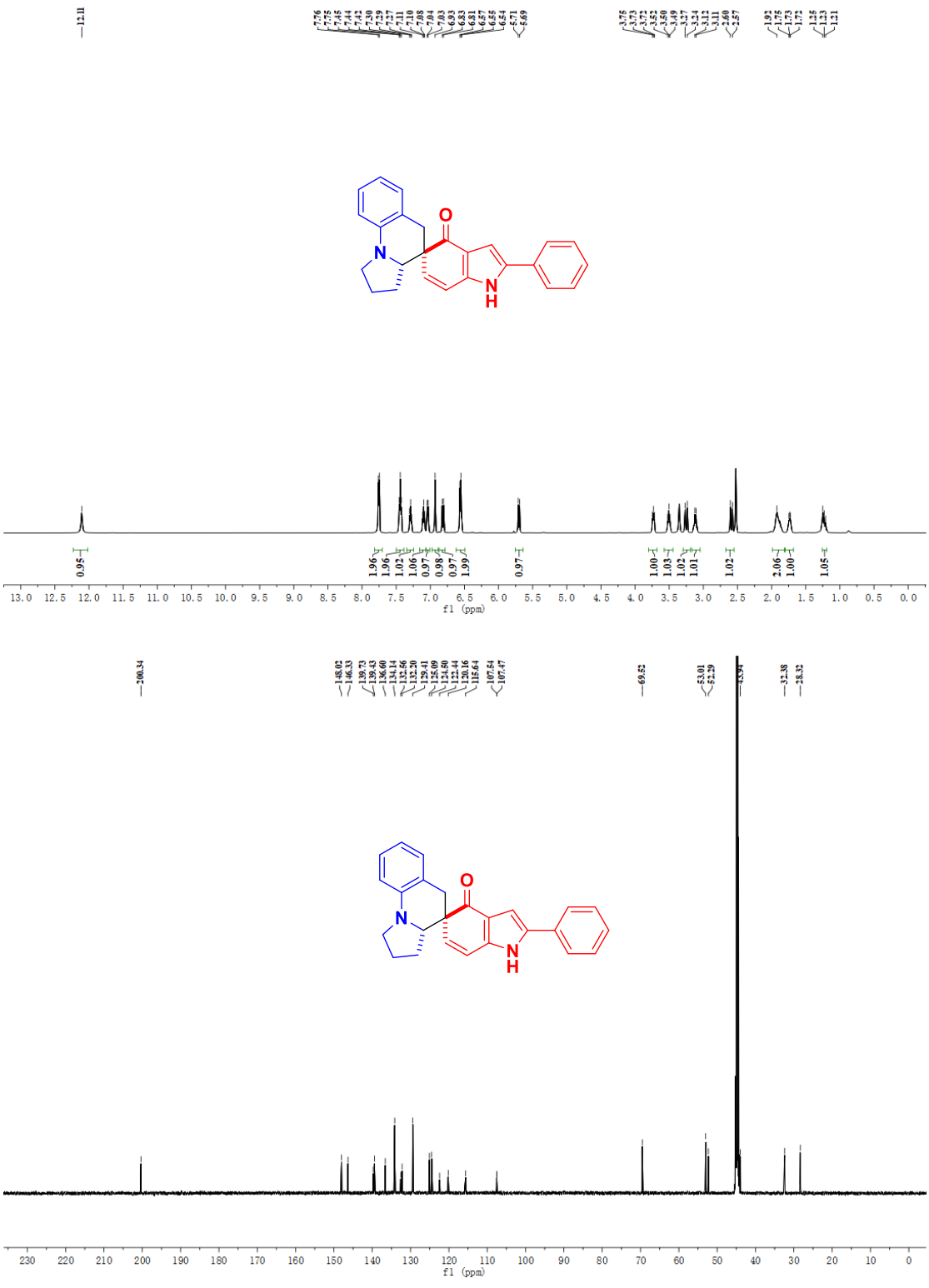


2-bromo-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,5'-indol]-4'(1'H)-one (3s)

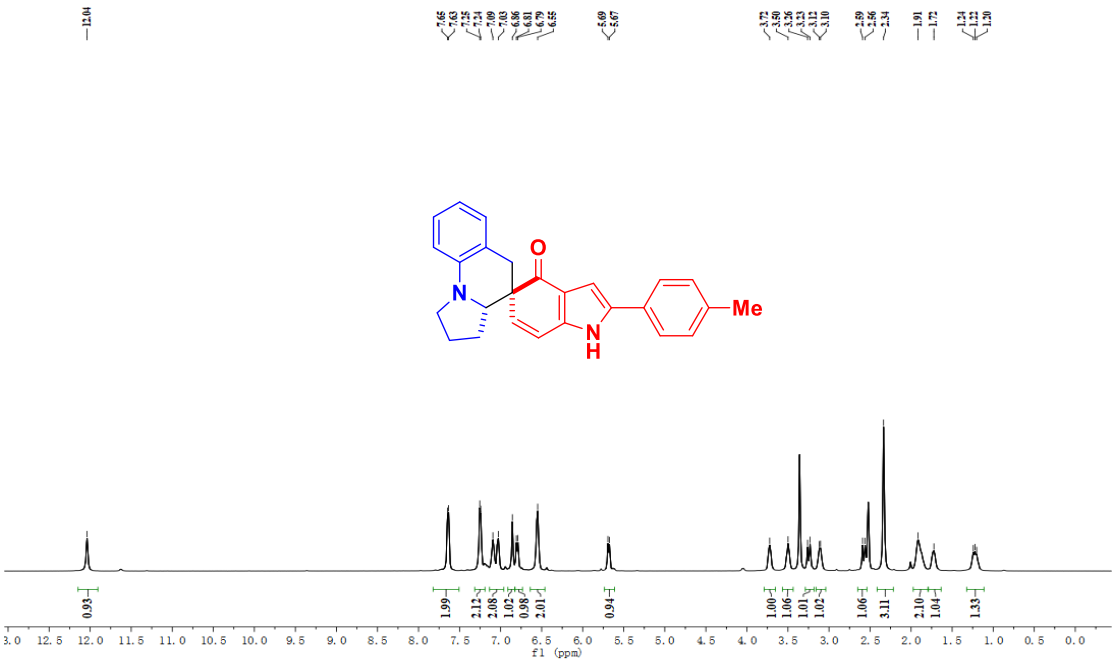


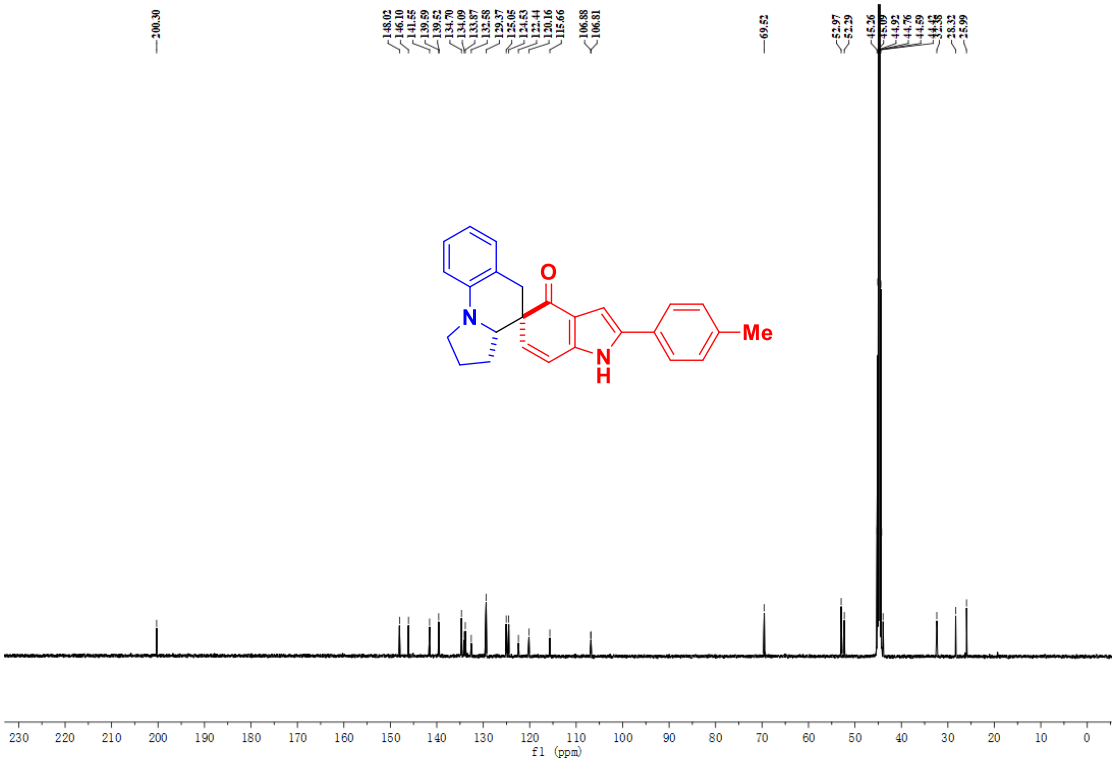


2-phenyl-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3t)

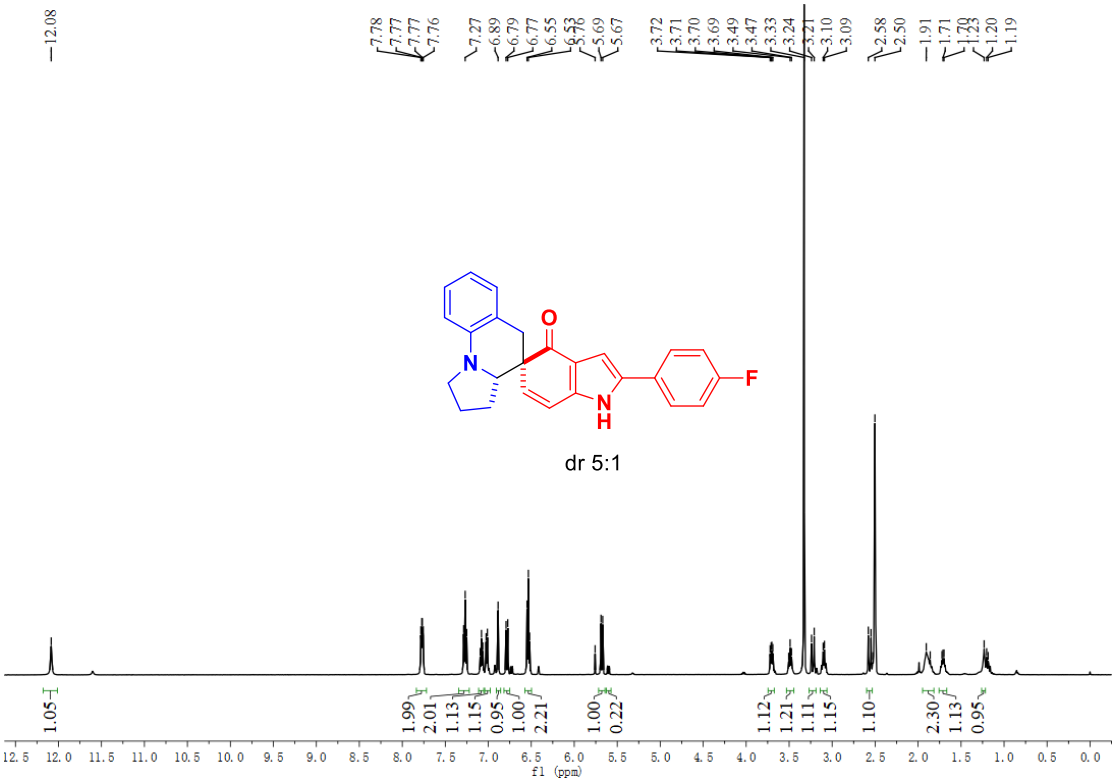


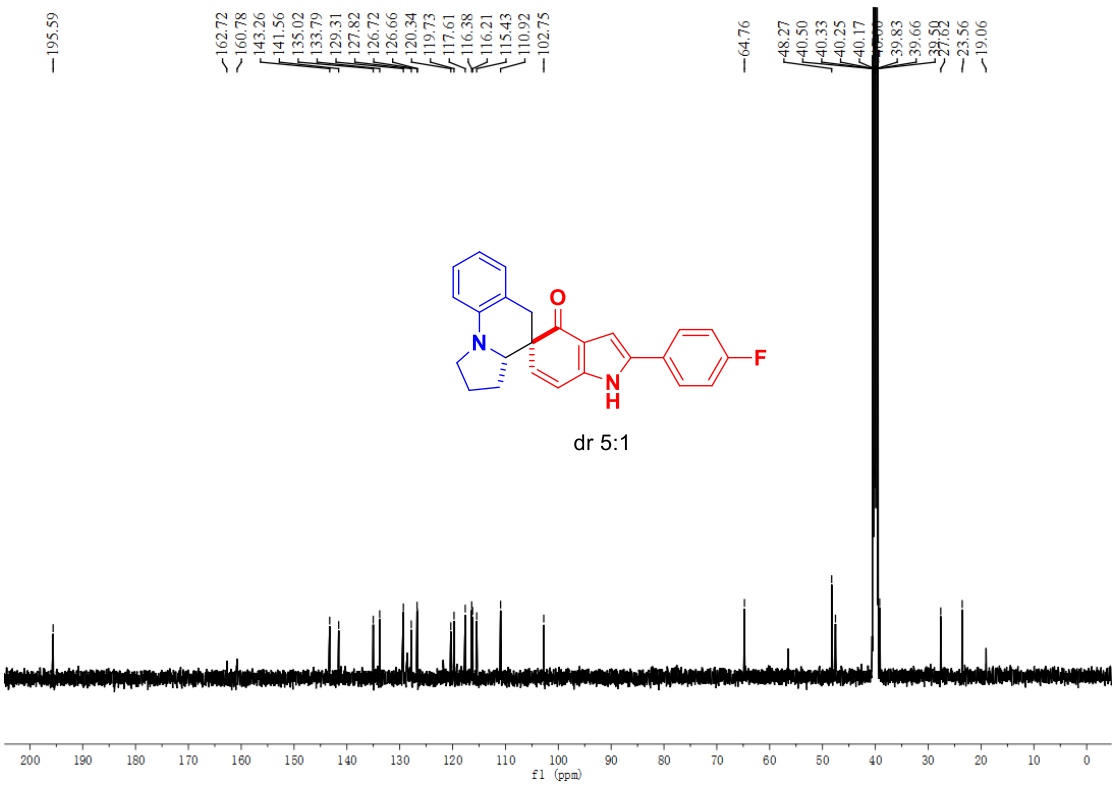
2-(*p*-tolyl)-1',2',3',3a'-tetrahydro-5'*H*-spiro[indole-5,4'-pyrrolo[1,2-*a*]quinolin]-4(1*H*)-one (3u)



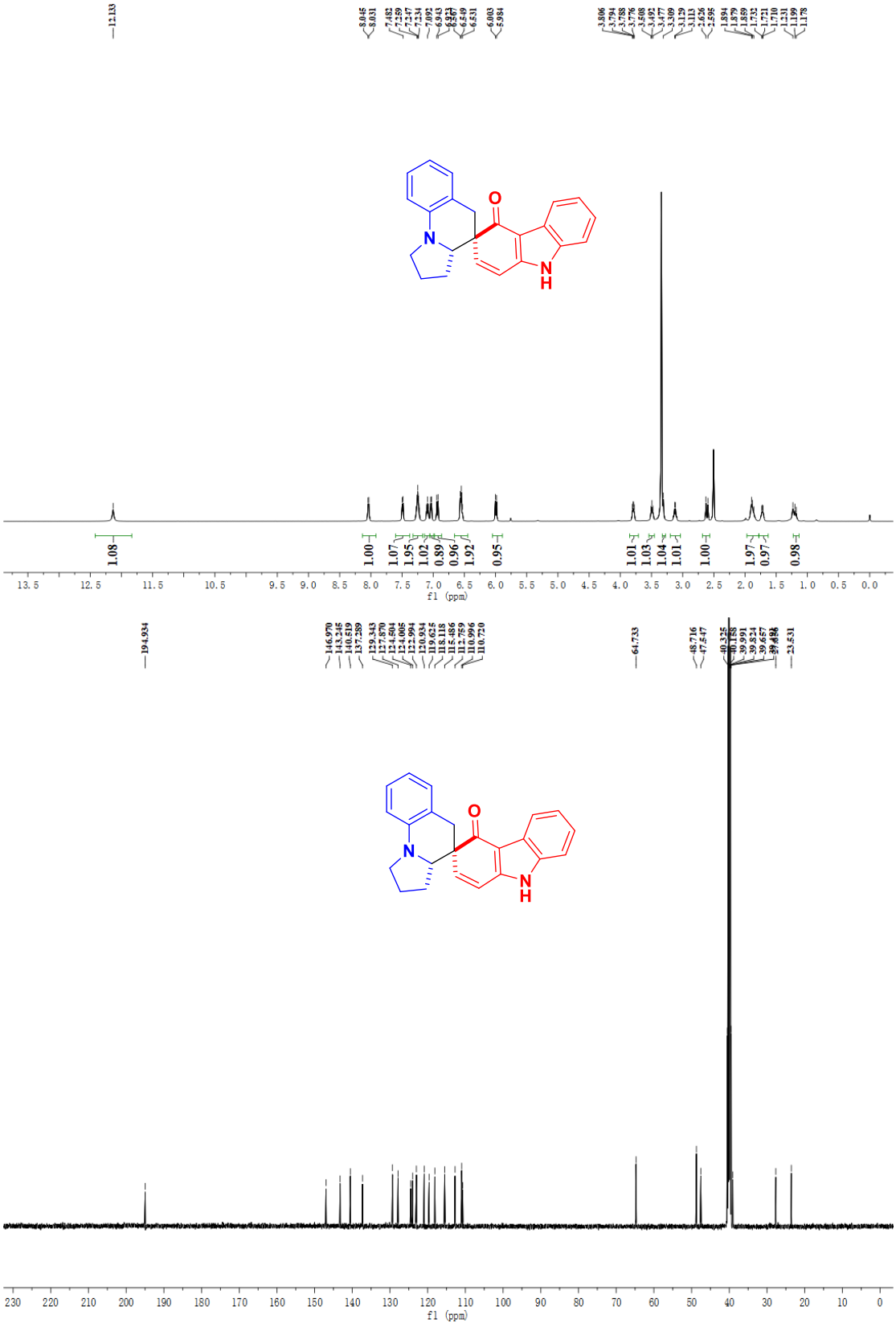


2-(4-fluorophenyl)-1',2',3',3a'-tetrahydro-5'H-spiro[indole-5,4'-pyrrolo[1,2-a]quinolin]-4(1H)-one (3v)

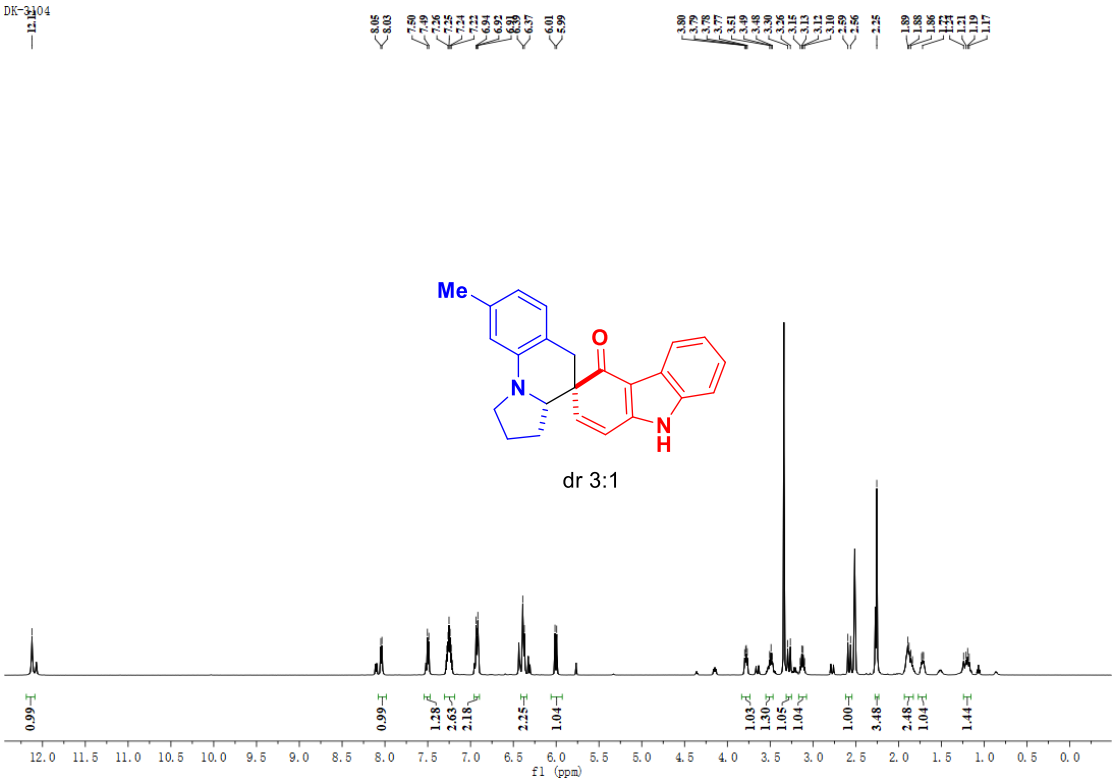


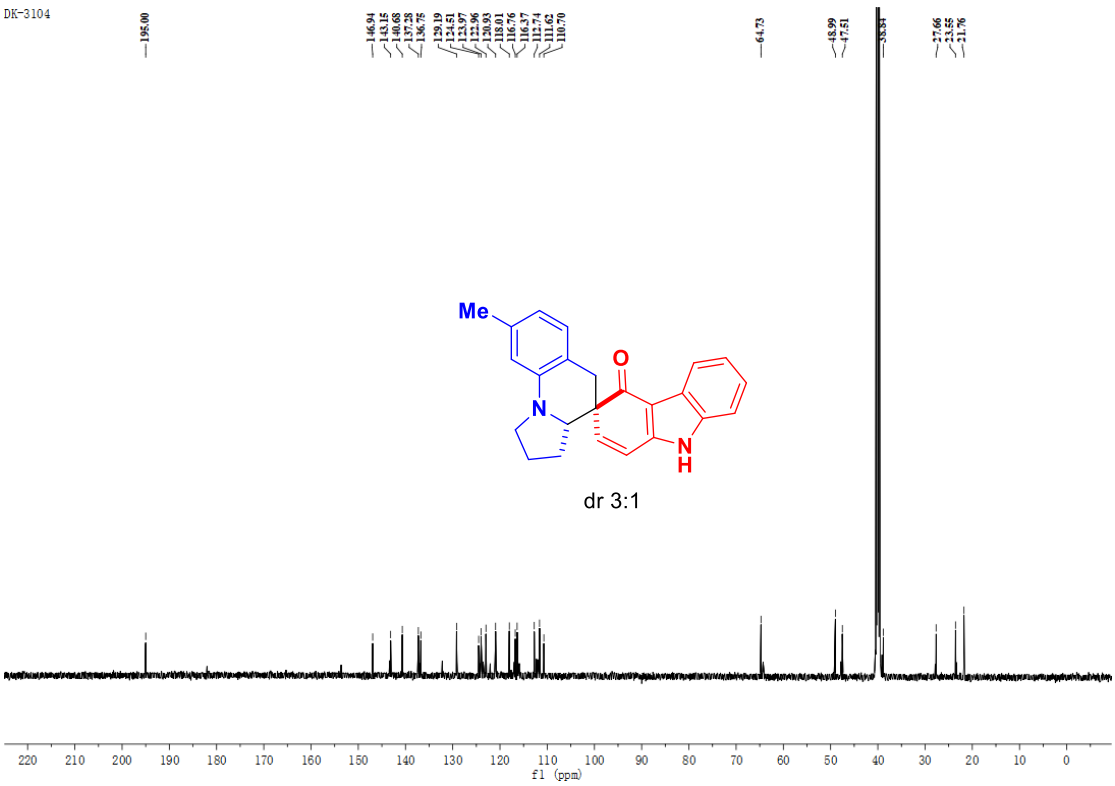


1',2',3',3a'-tetrahydro-5'*H*-spiro[carbazole-3,4'-pyrrolo[1,2-*a*]quinolin]-4(9*H*)-one (5a)

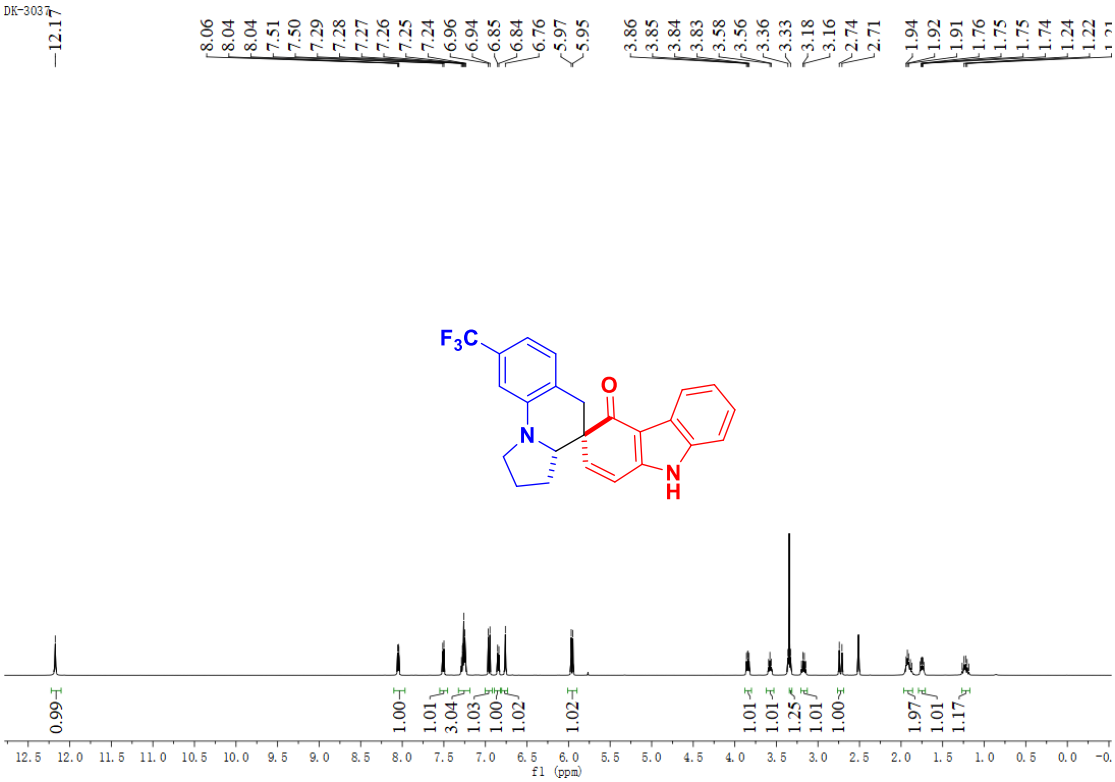


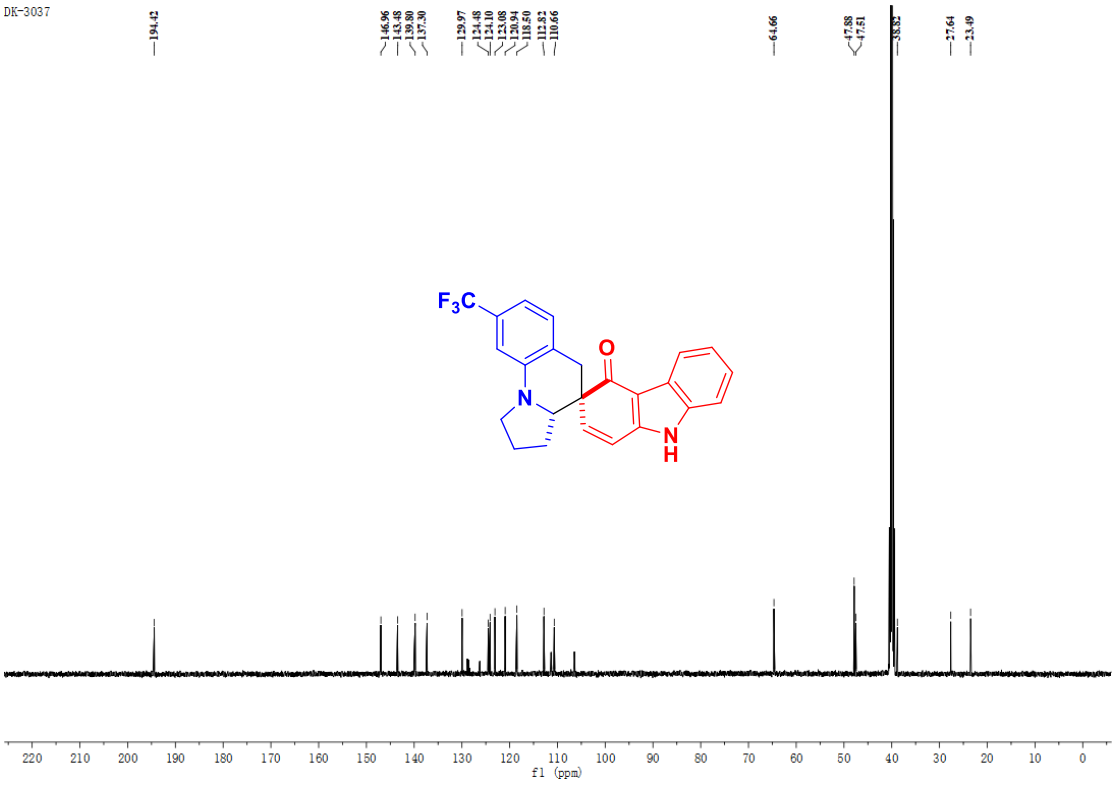
8'-methyl-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-4(9H)-one (5b)



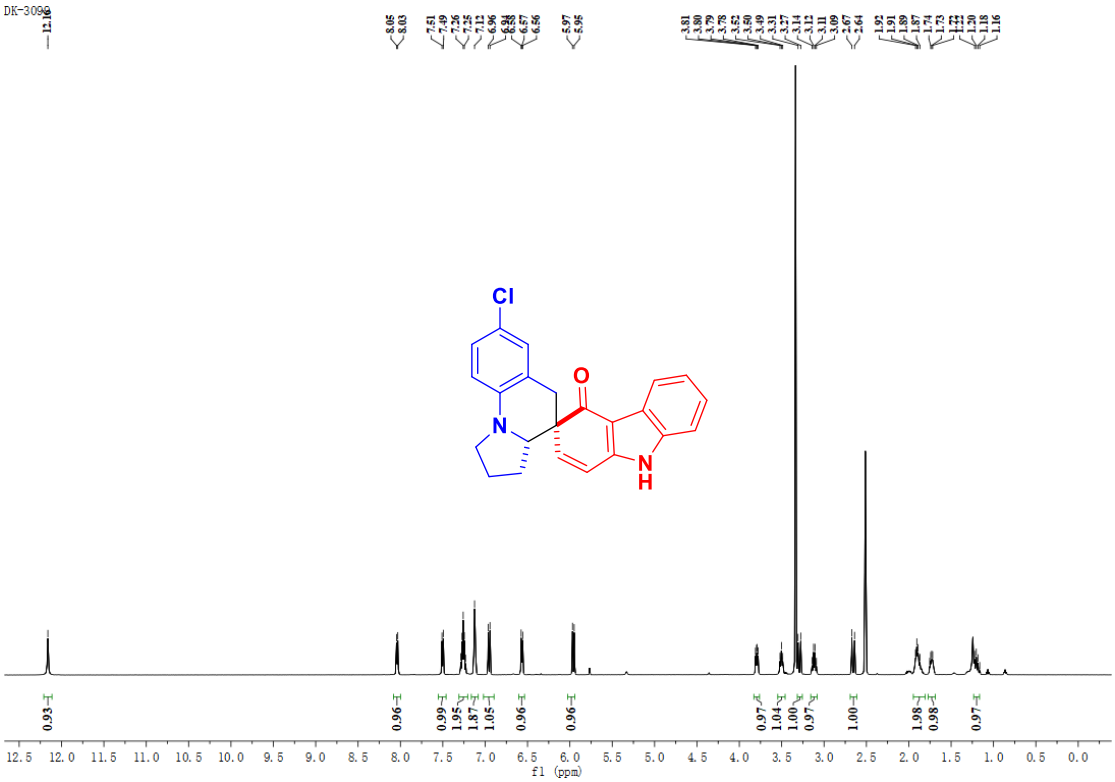


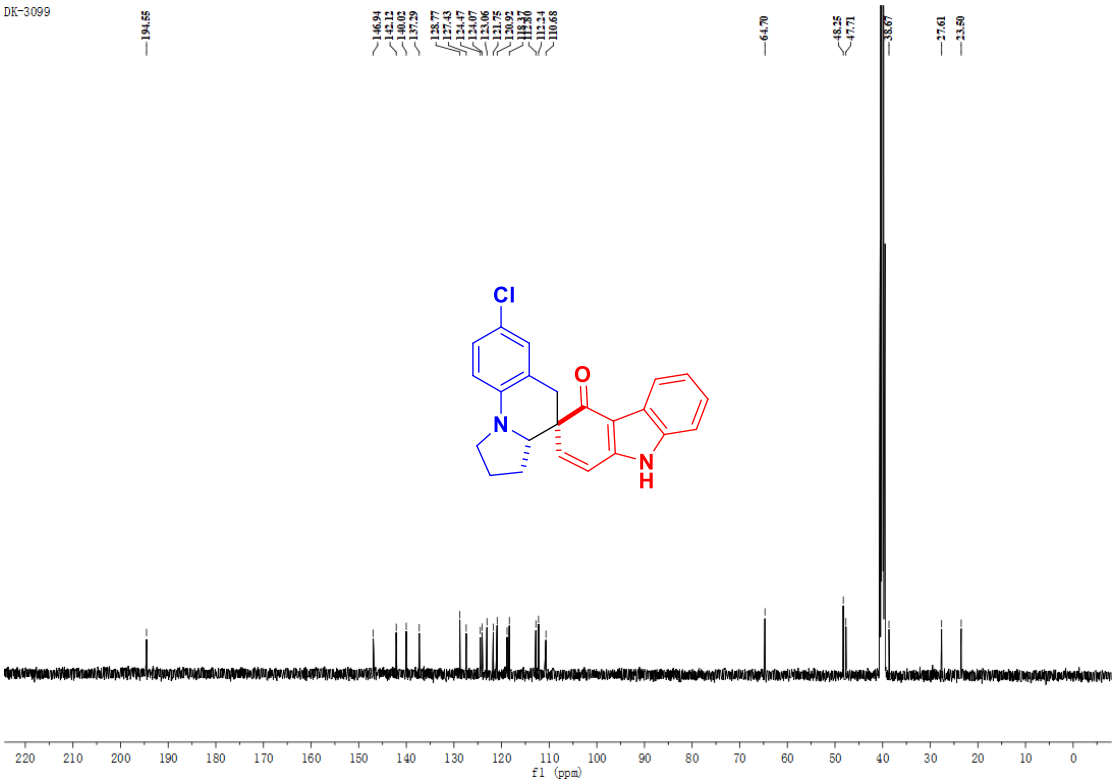
8'-(trifluoromethyl)-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-4(9H)-one (5c)



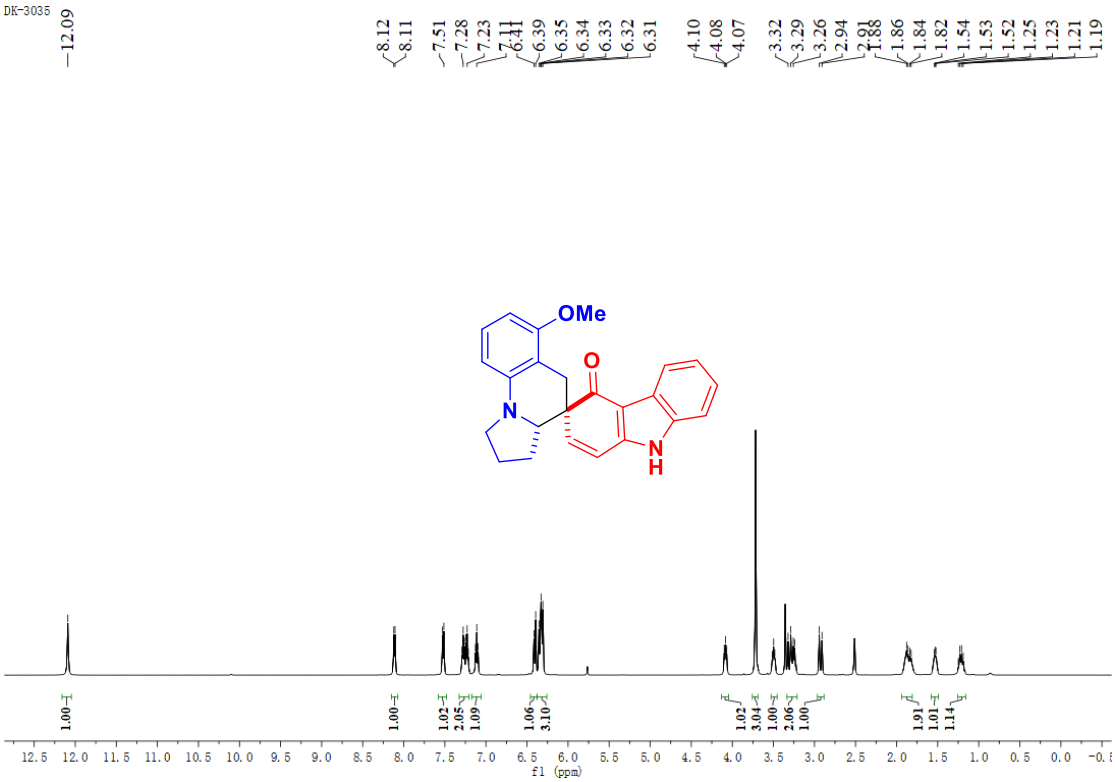


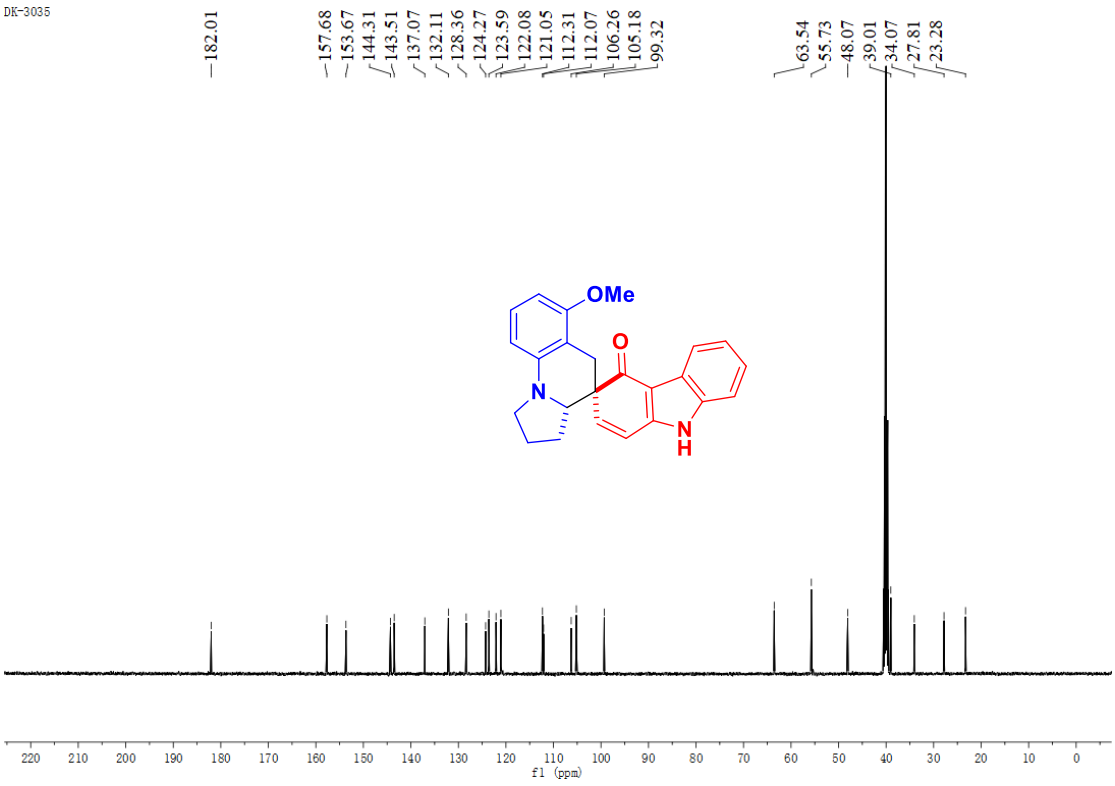
7'-chloro-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-4(9H)-one (5d)



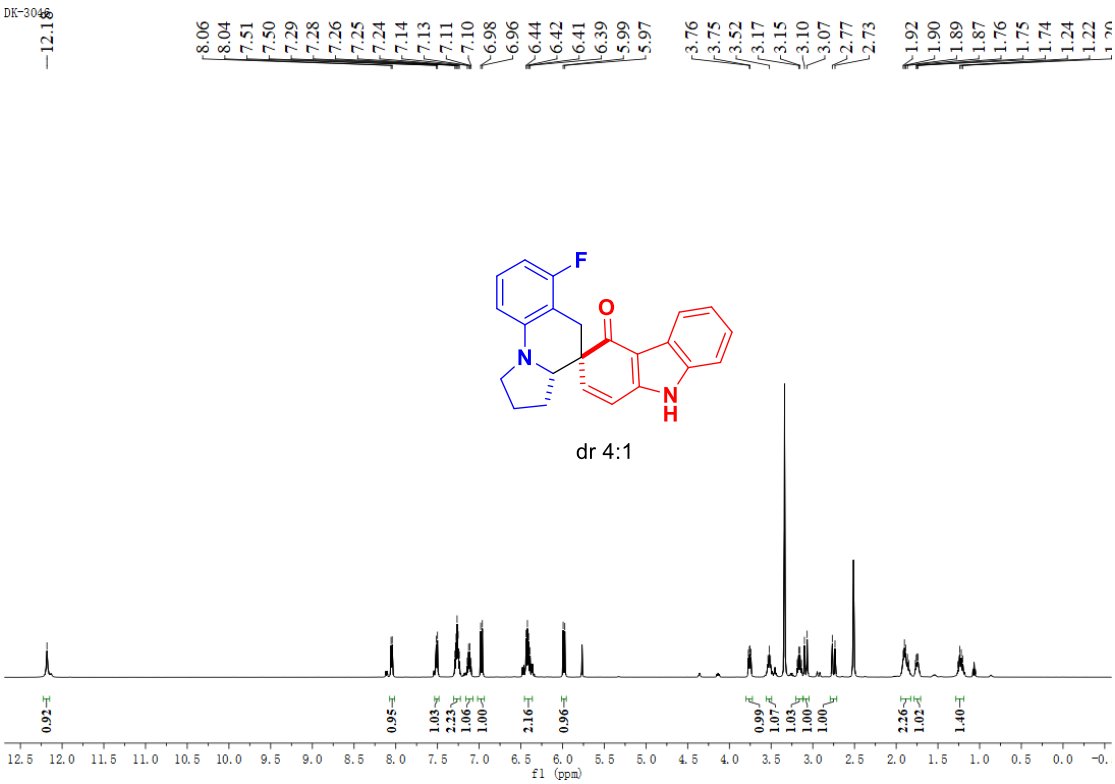


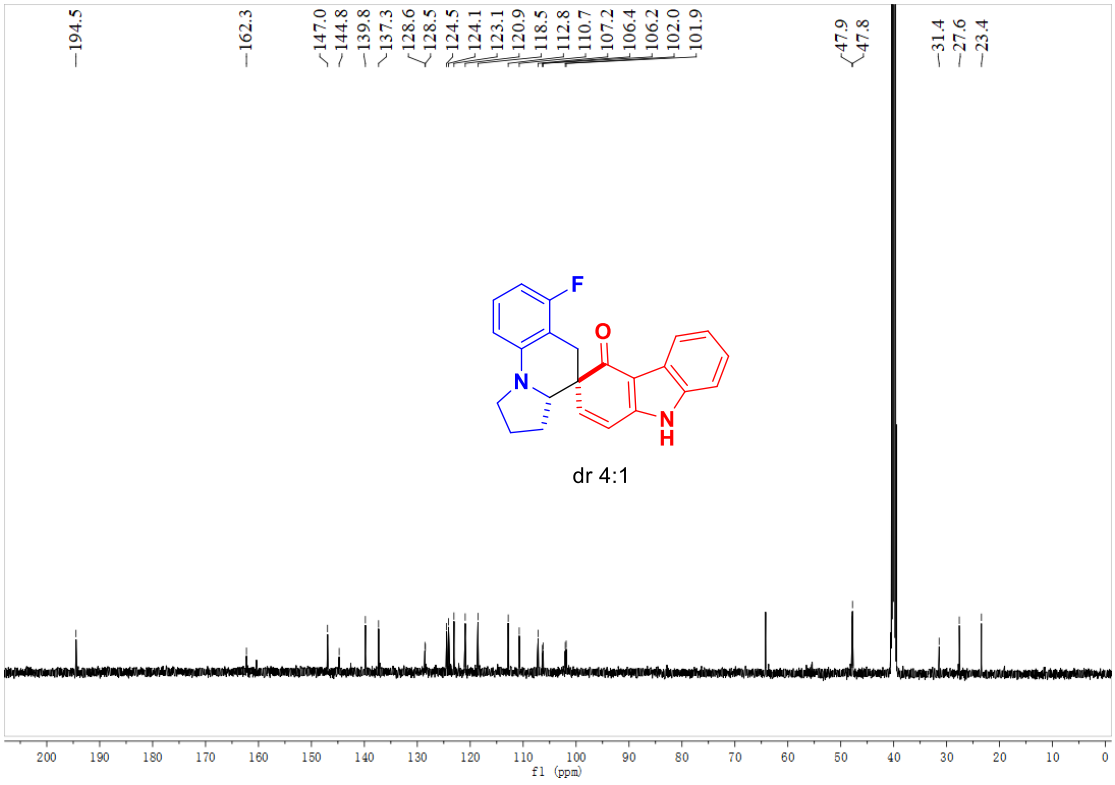
6'-methoxy-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-4(9H)-one (5e)



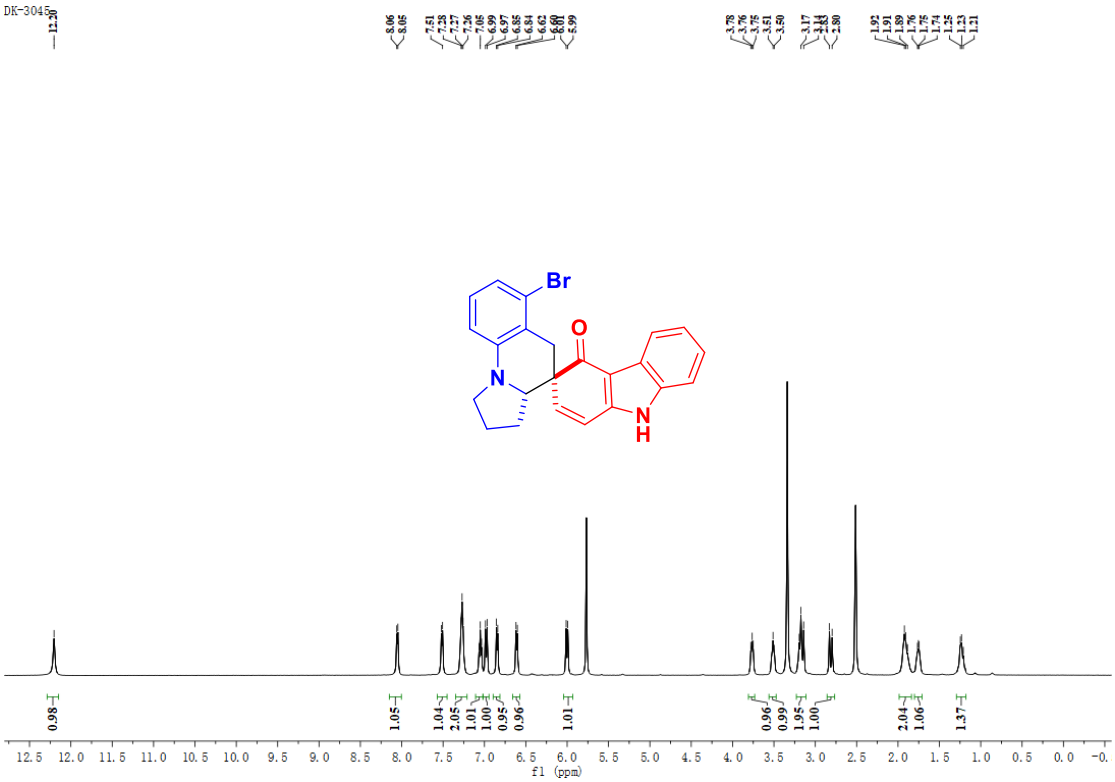


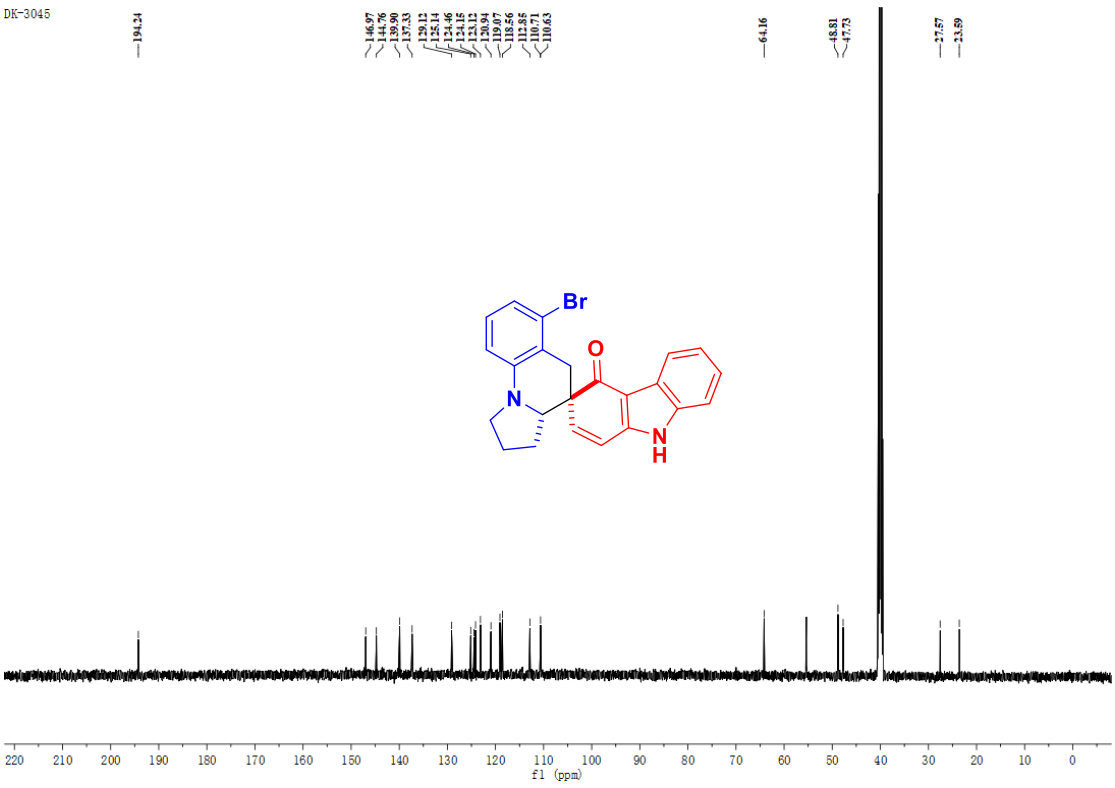
6'-fluoro-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-4(9H)-one (5f)



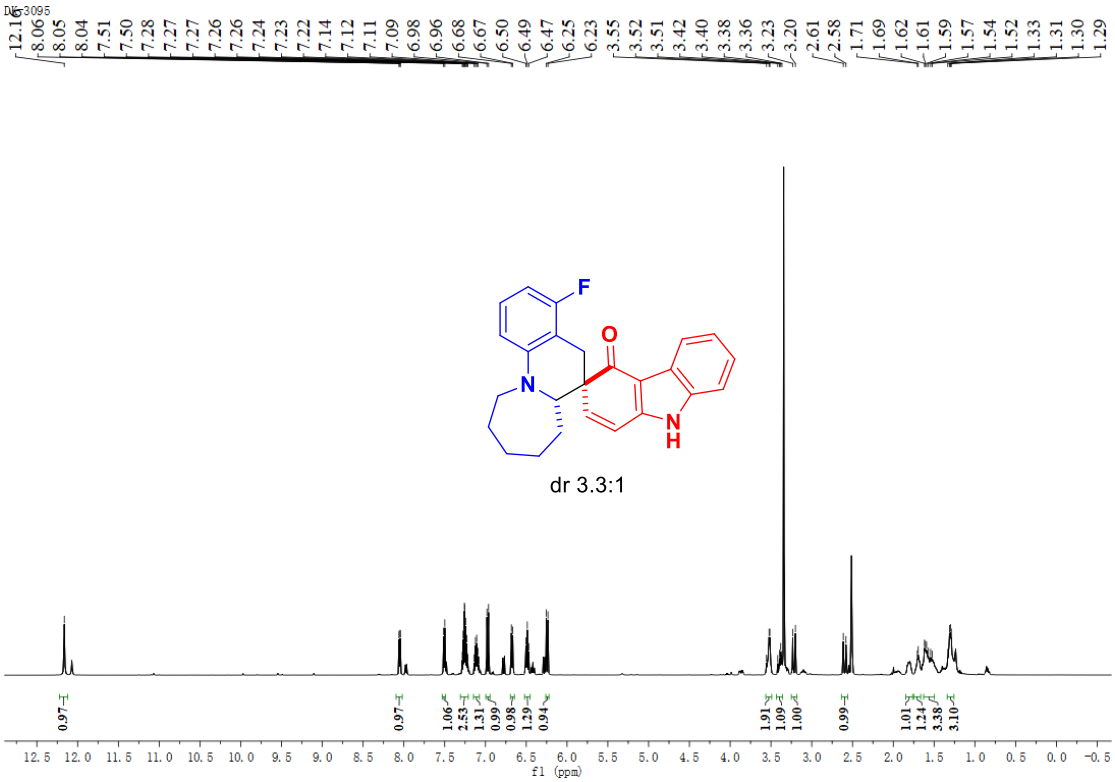


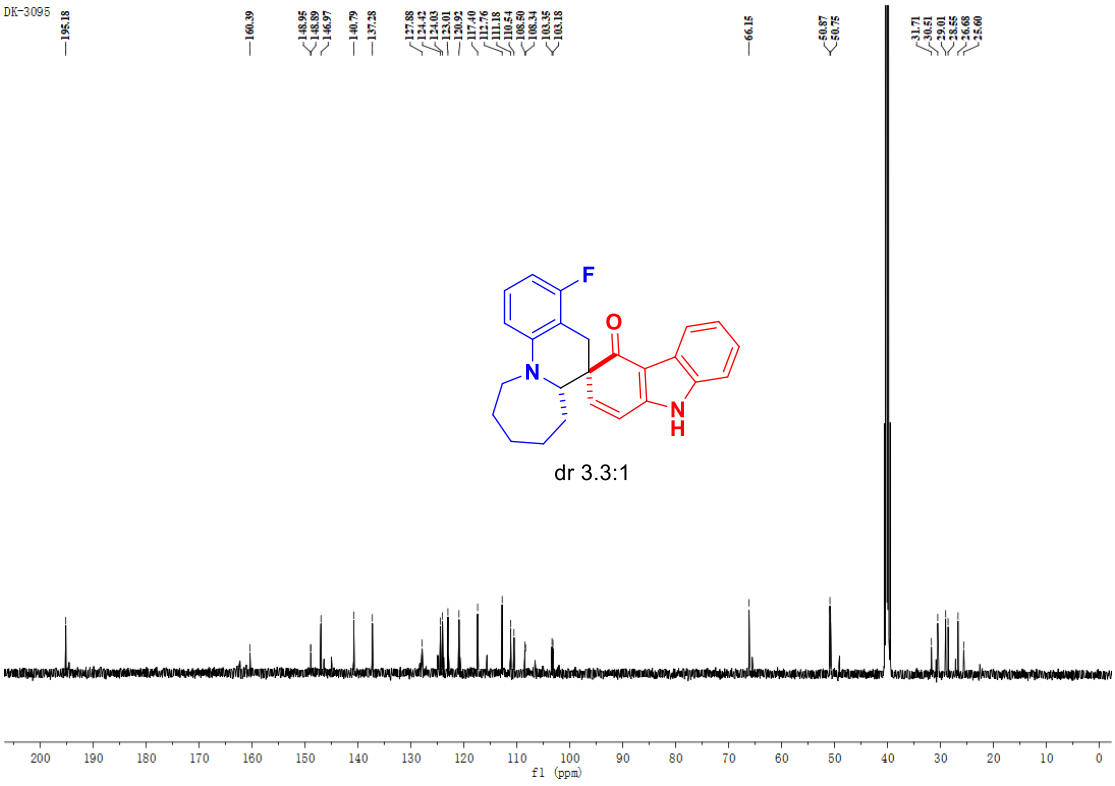
6'-bromo-1',2',3',3a'-tetrahydro-5'H-spiro[carbazole-3,4'-pyrrolo[1,2-a]quinolin]-4(9H)-one (5g)



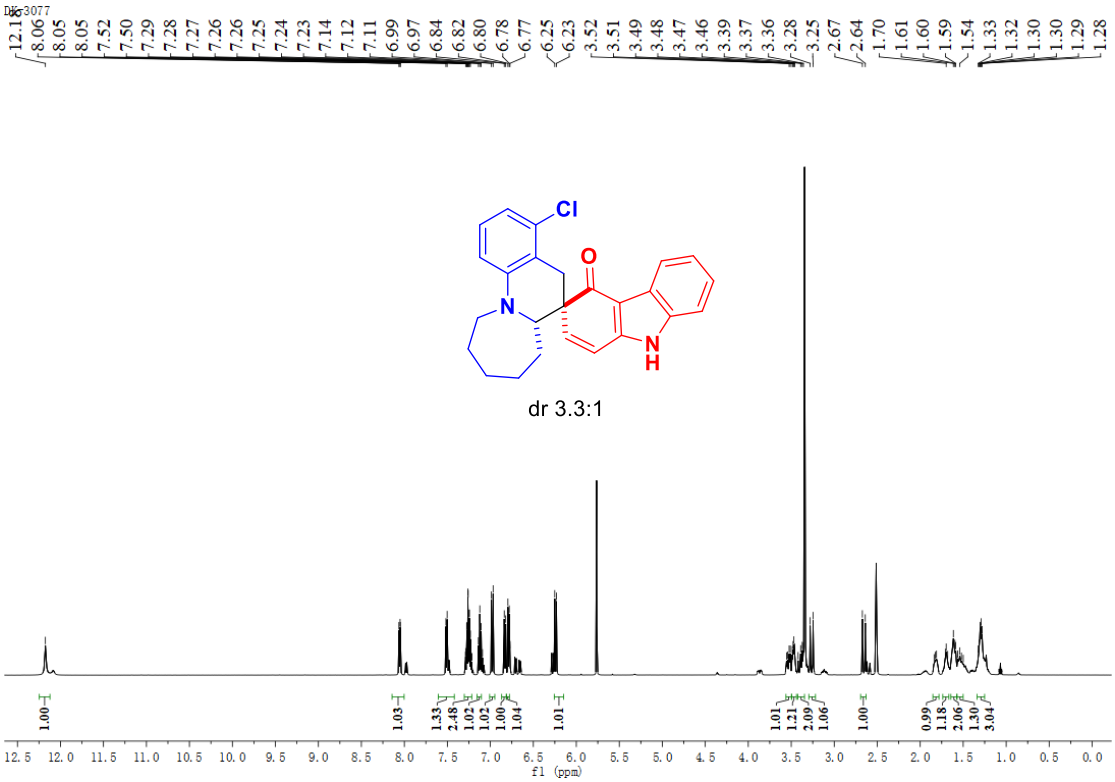


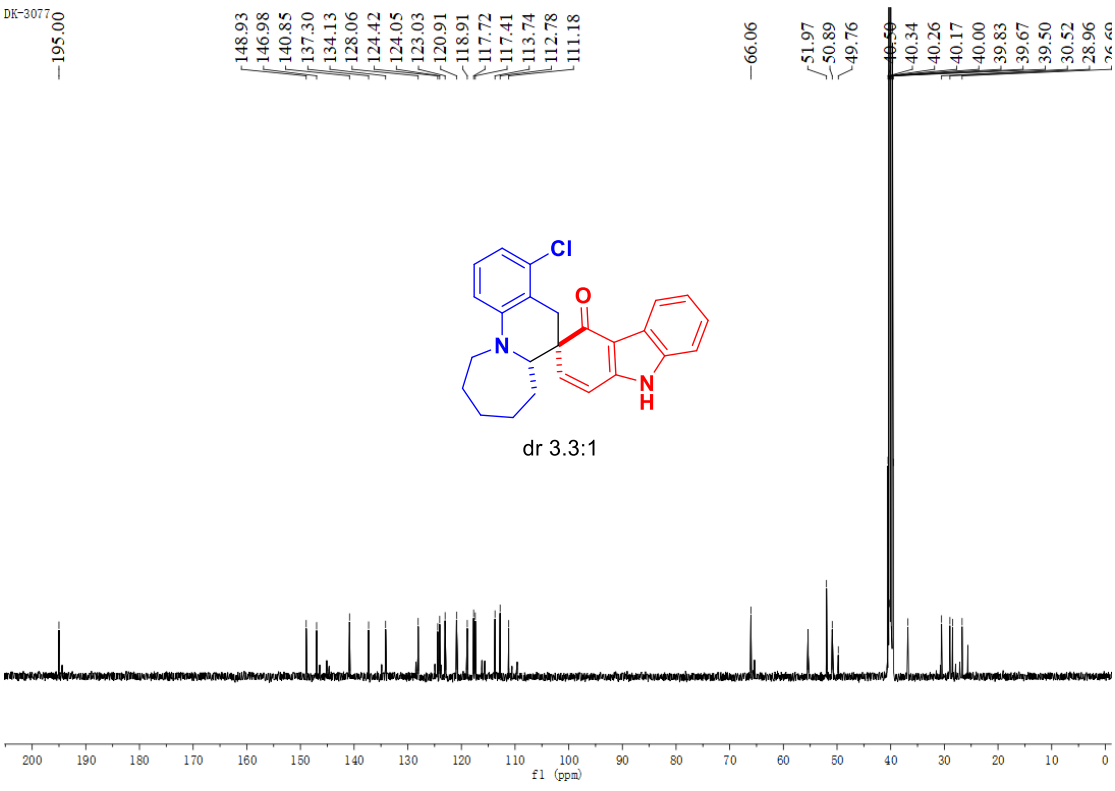
4-fluoro-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,3'-carbazol]-4'(9'H)-one (5h)



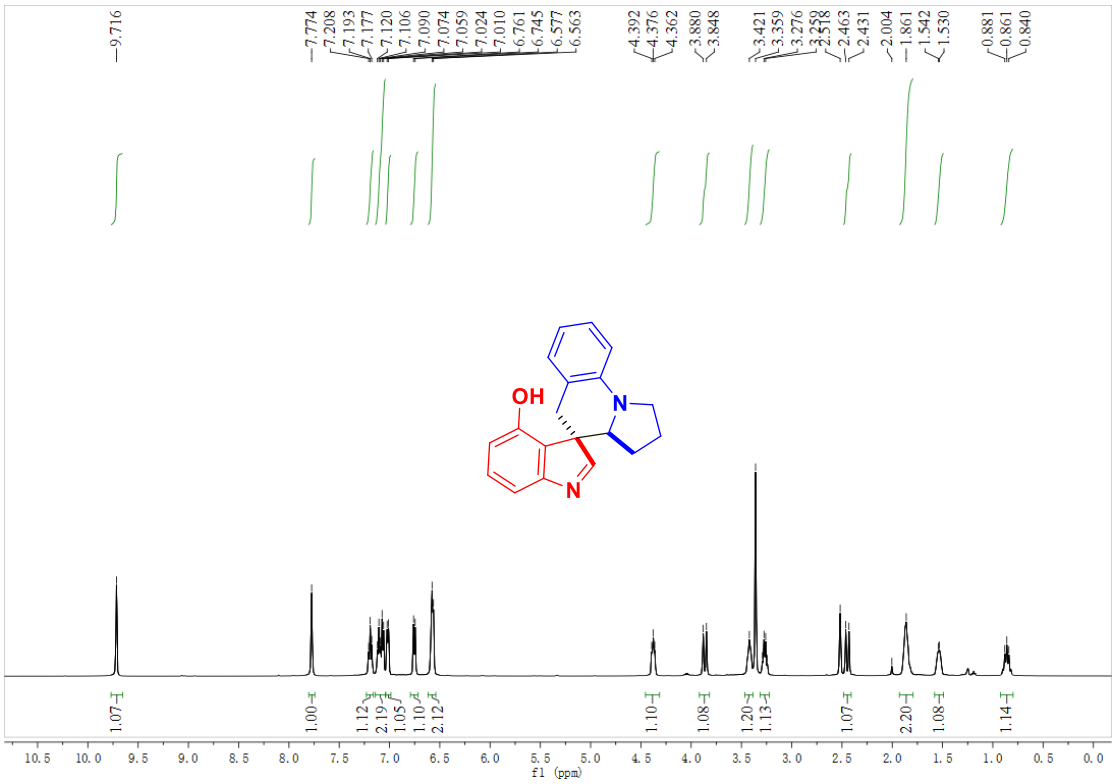


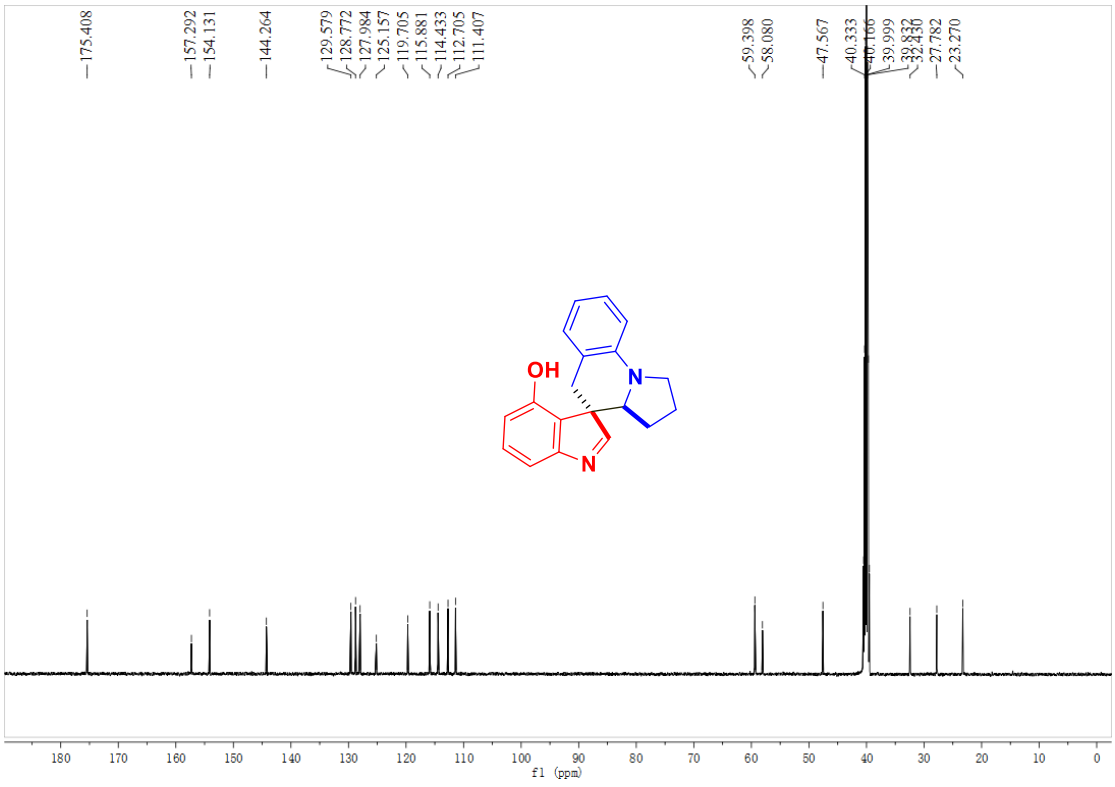
4-chloro-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,3'-carbazol]-4'(9'H)-one (5i)



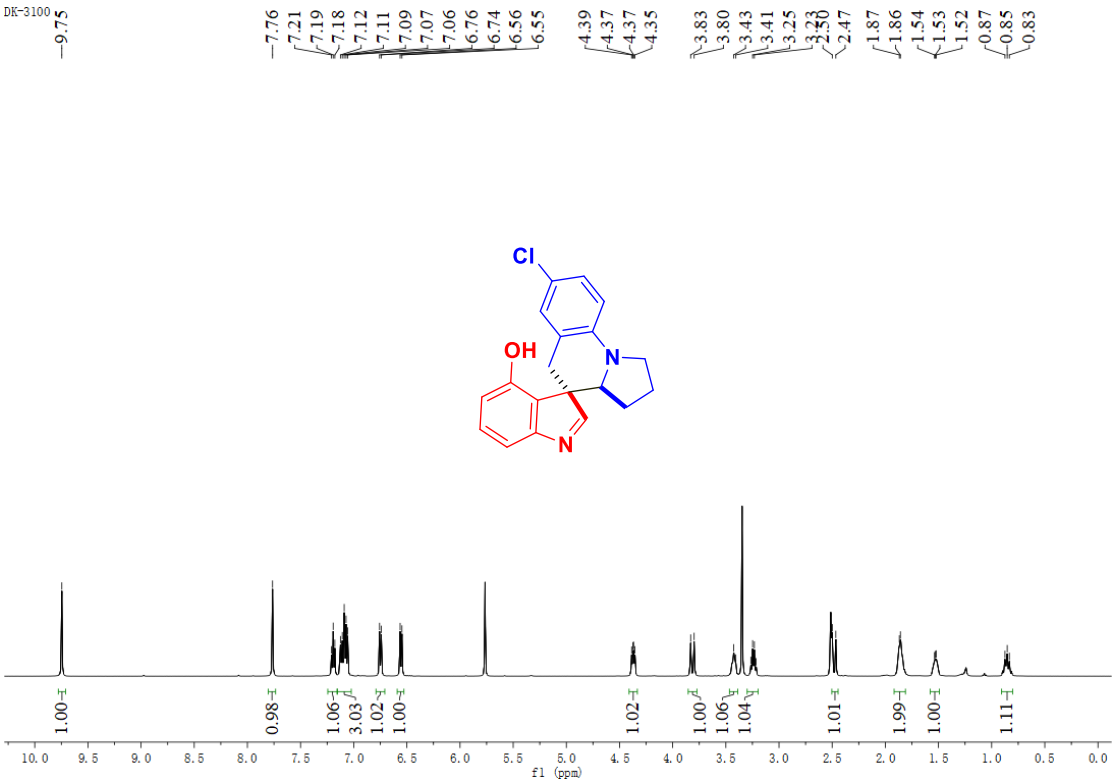


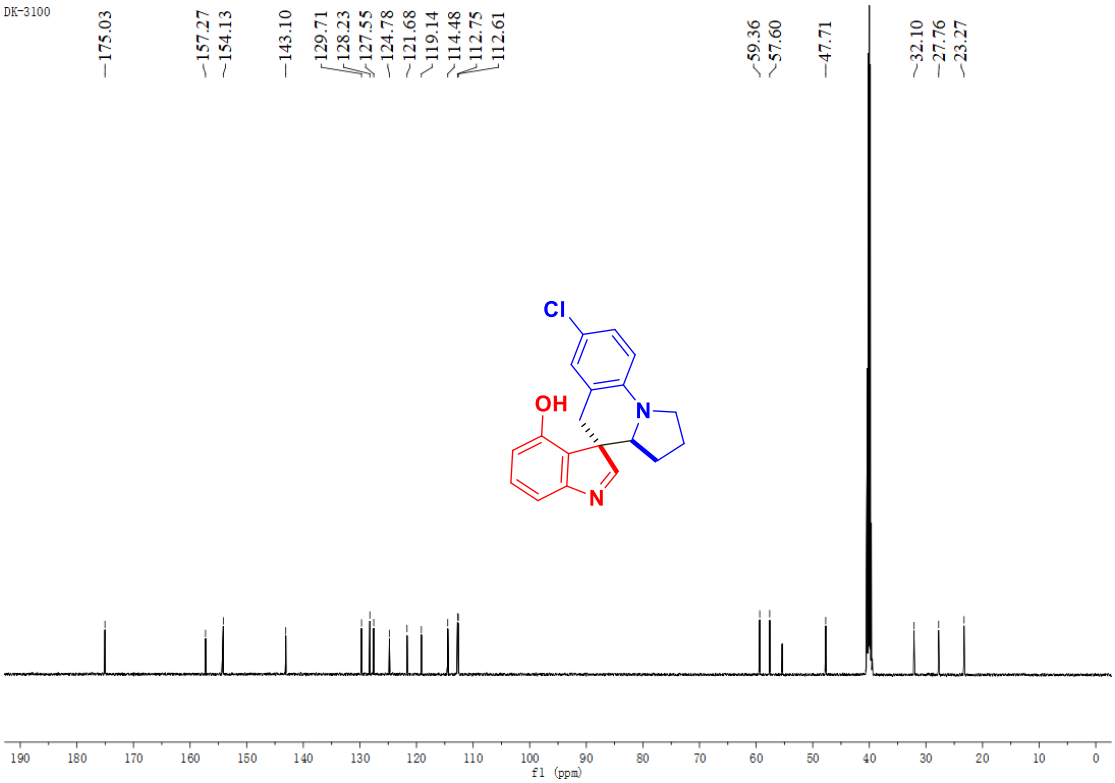
1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7a)



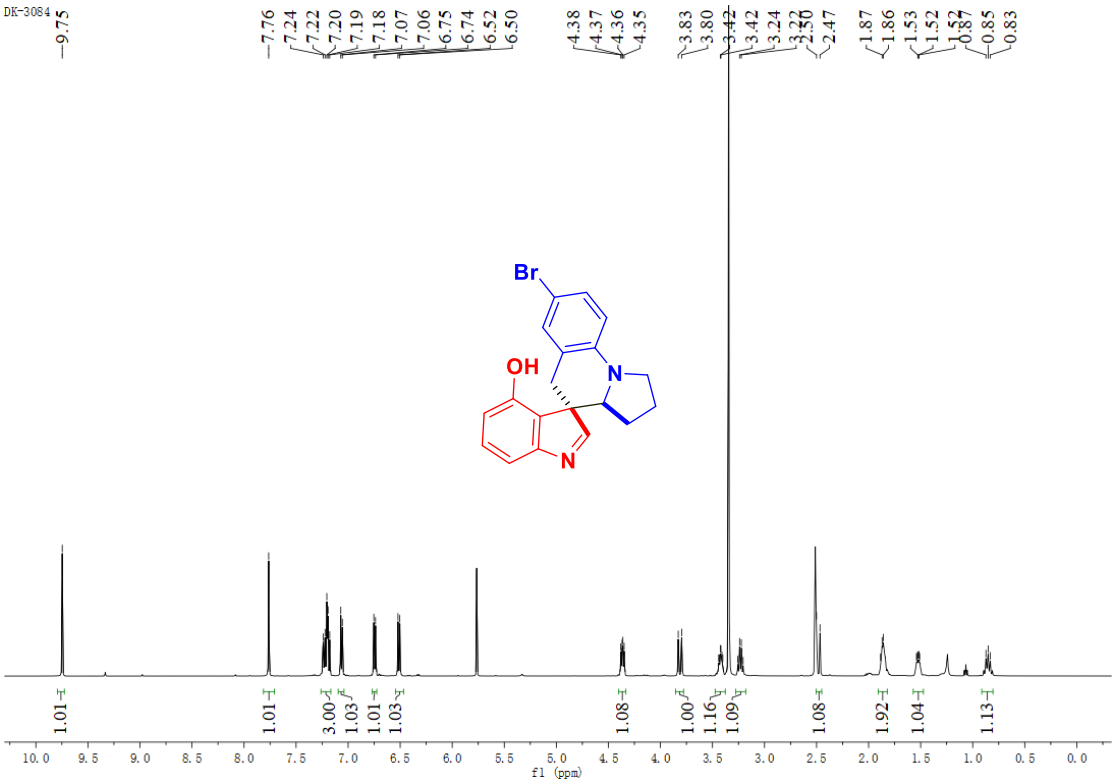


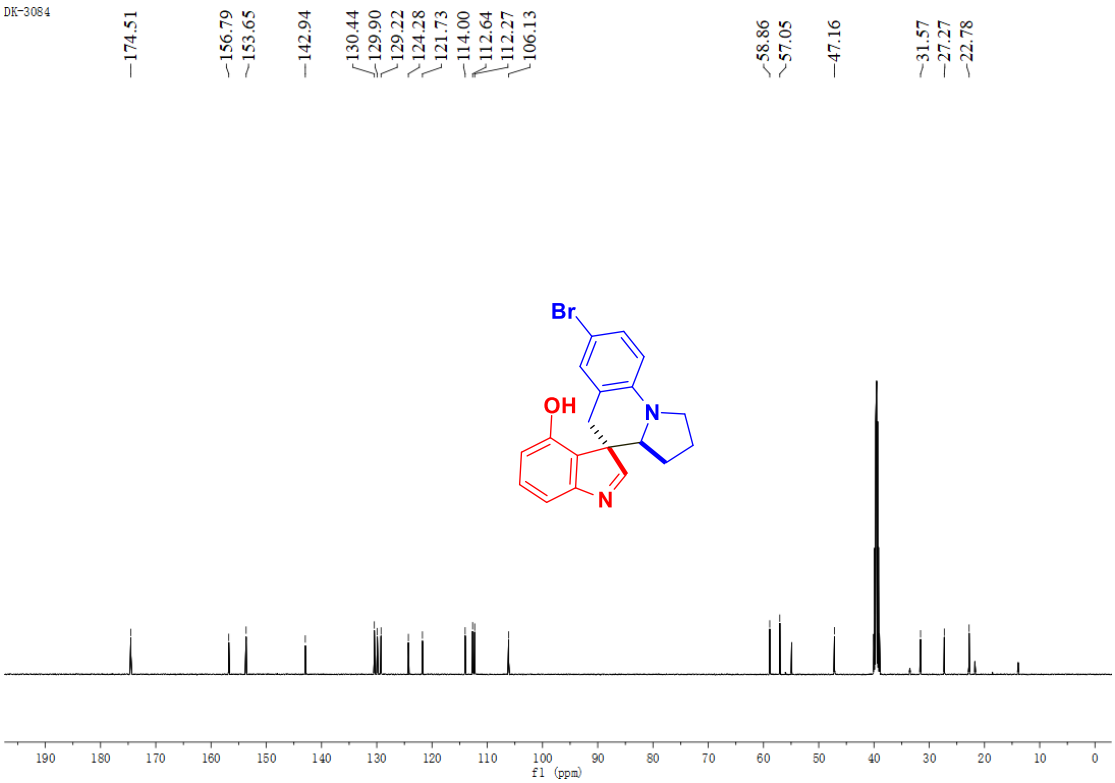
7'-chloro-1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7b)



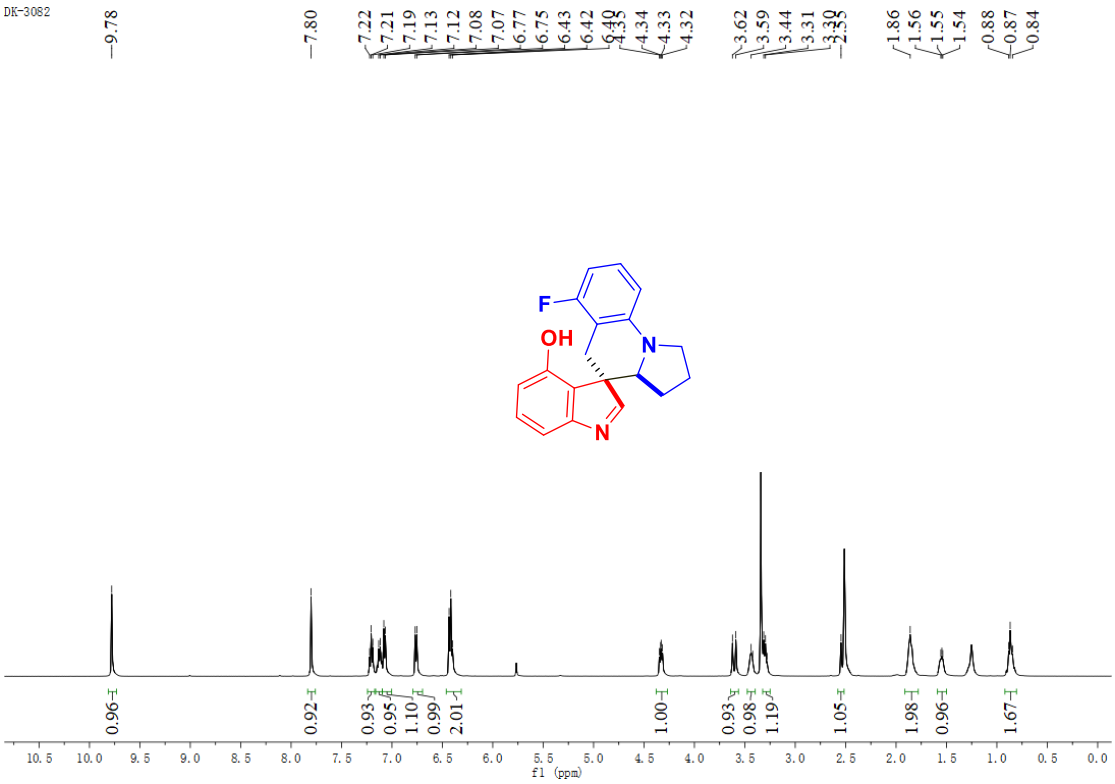


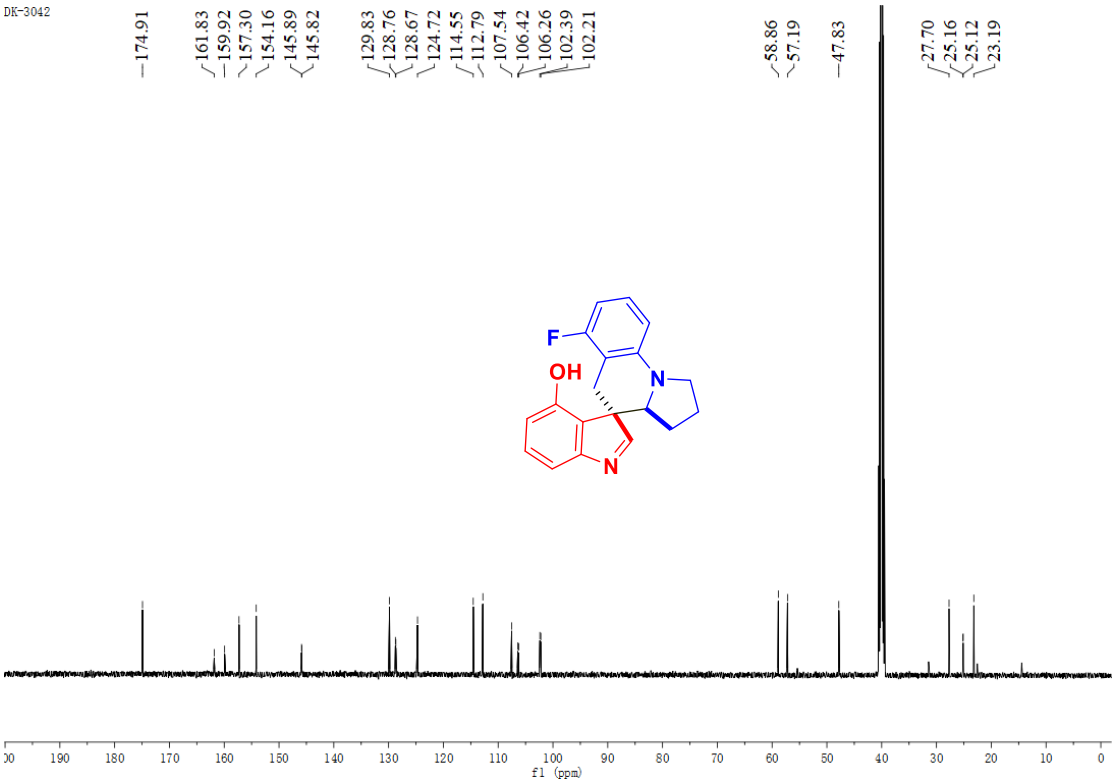
7'-bromo-1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7c)



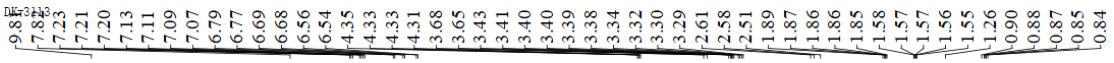


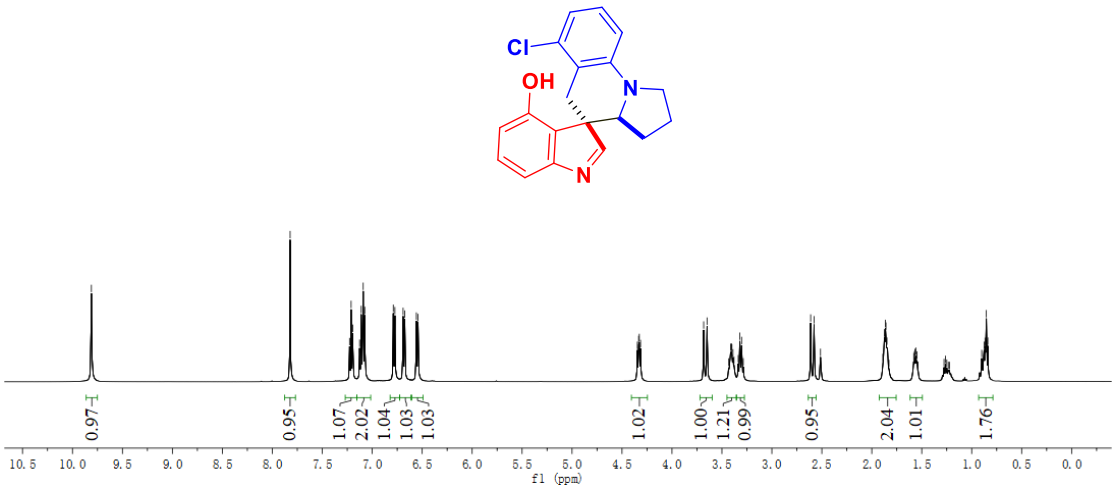
6'-fluoro-1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7d)

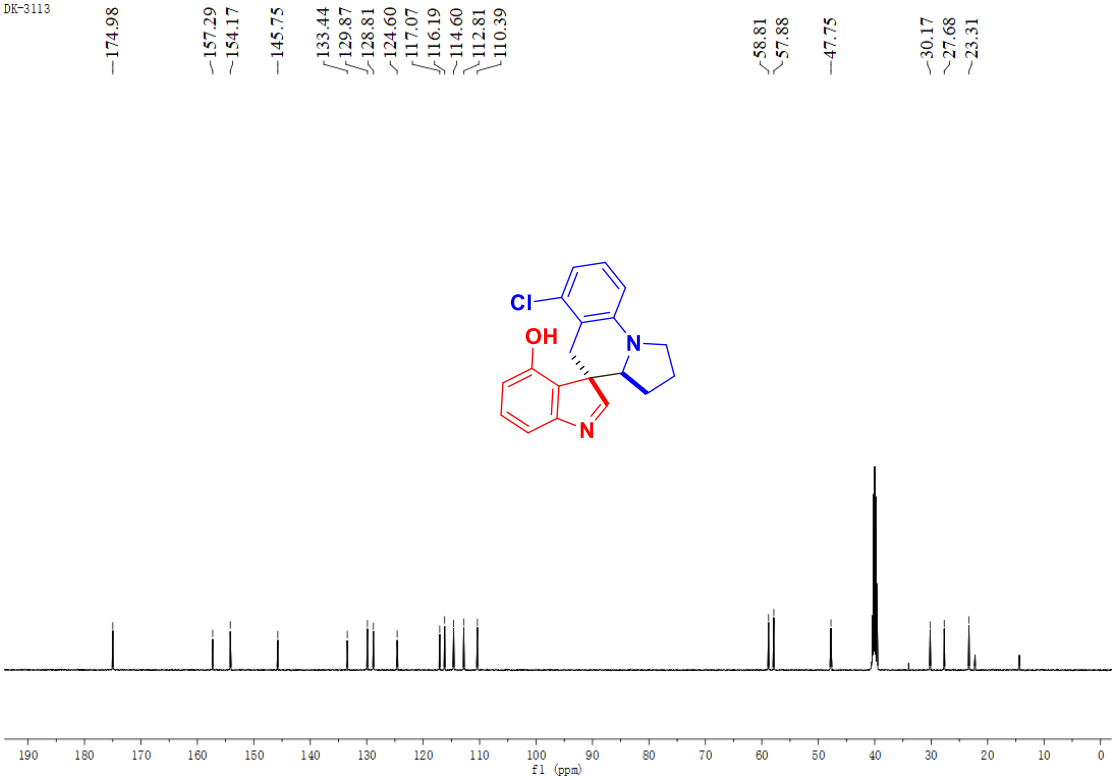




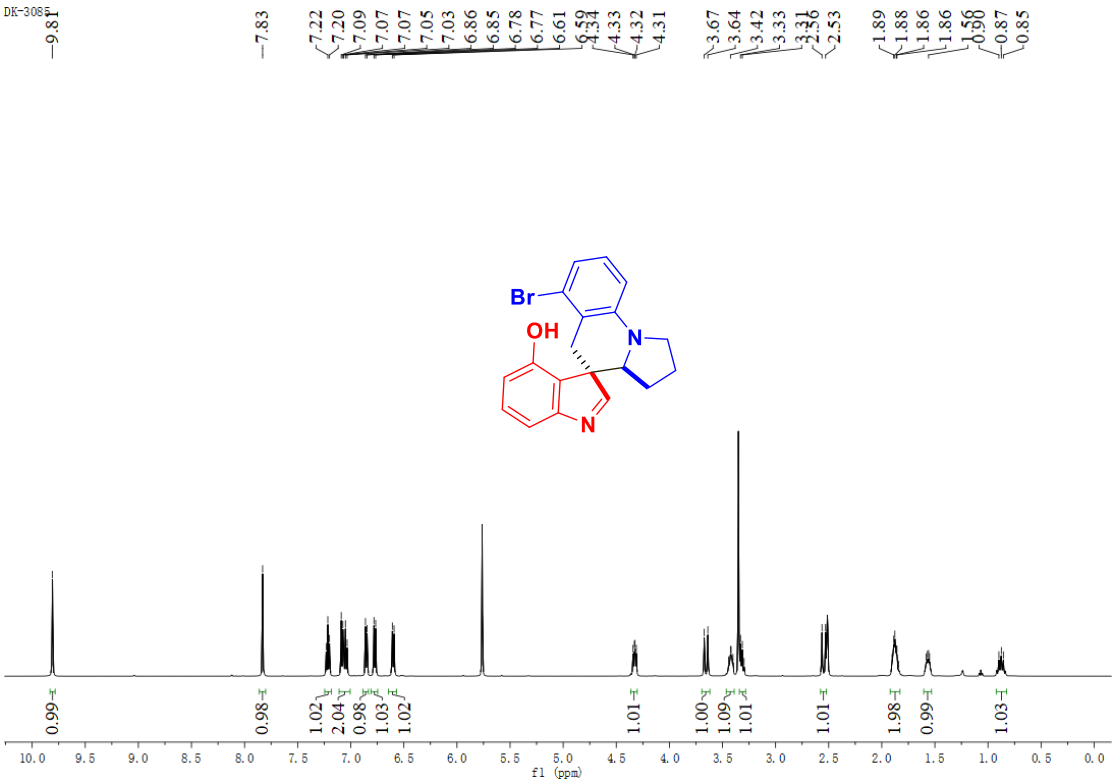
6'-chloro-1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7e)

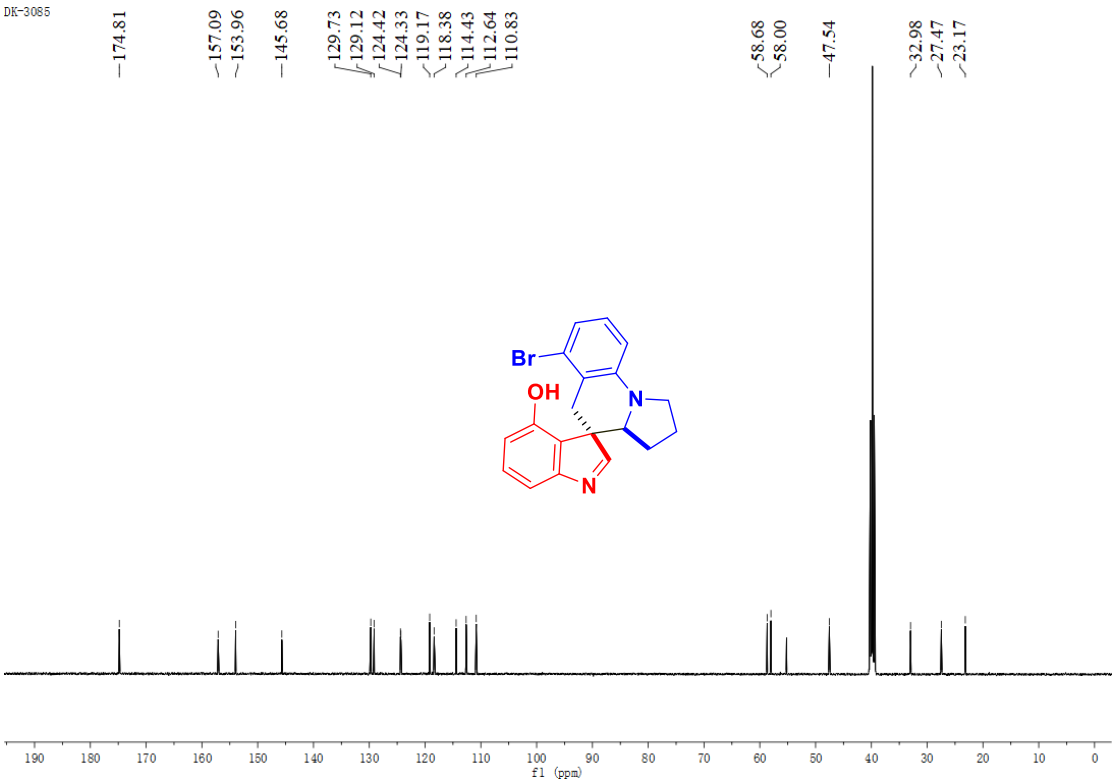






6'-bromo-1',2',3',3a'-tetrahydro-5'H-spiro[indole-3,4'-pyrrolo[1,2-a]quinolin]-4-ol (7f)





3-bromo-6a,7,8,9,10,11-hexahydro-5H-spiro[azepino[1,2-a]quinoline-6,3'-indol]-4'-ol (7g)