



Supporting Information

for

Copper-catalyzed multicomponent reactions for the efficient synthesis of diverse spirotetrahydrocarbazoles

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Characterization data and ^1H NMR, ^{13}C NMR, HRMS spectra of the compounds

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Single crystals were grown by slow evaporation of concentrated solution in CHCl_3 / DCM / EtOH (compounds **1f**, **2b**, **2g'**, **3a**, **4e**, **5b**) in glass vials, which were then sealed by plugs with needles on them.

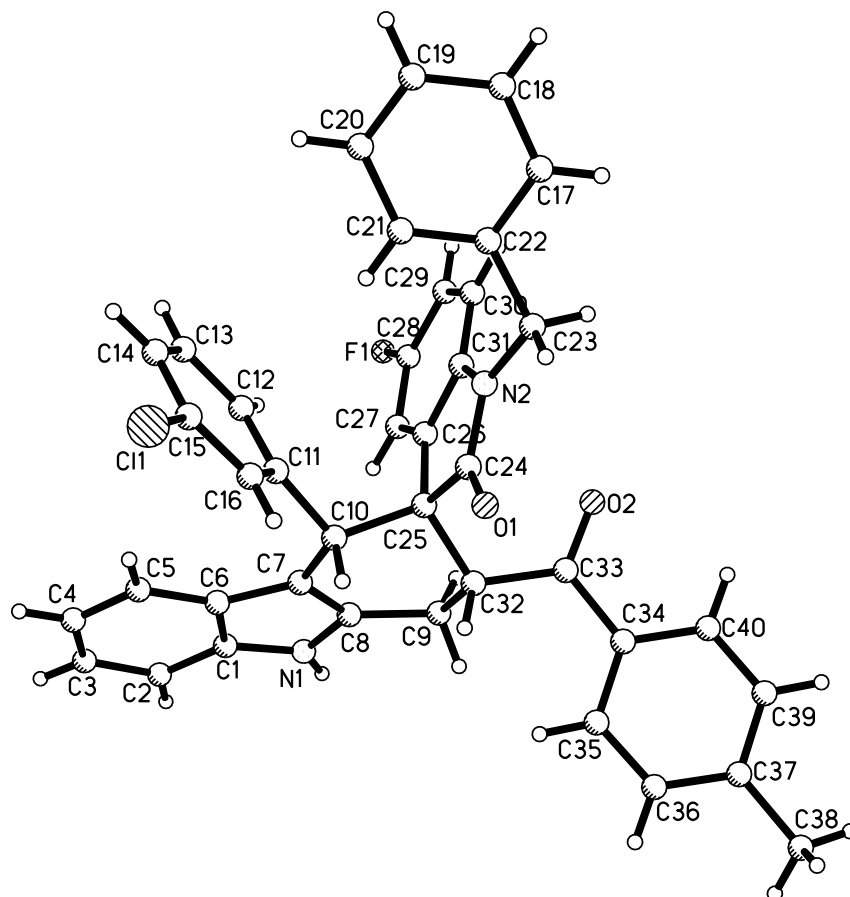


Fig. S1 ORTEP drawing (30%) of the crystal structure of **1f**

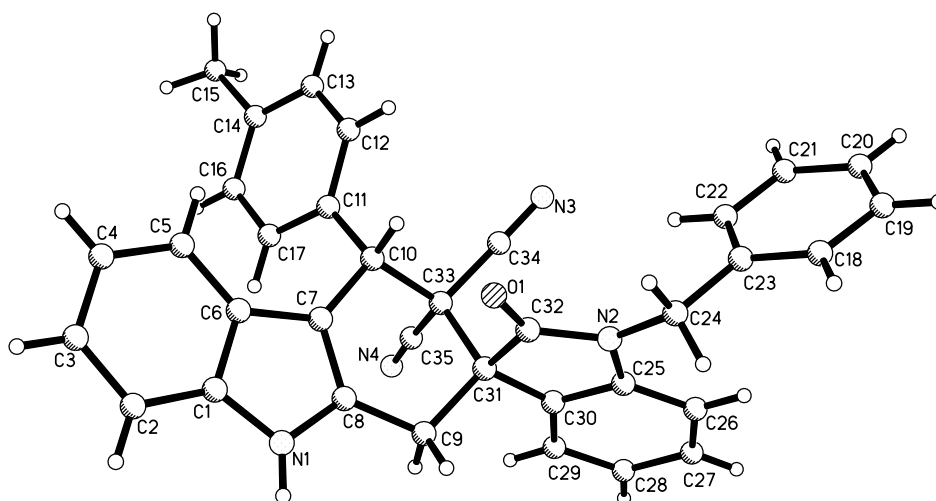


Fig. S2 ORTEP drawing (30%) of the crystal structure of **2b**

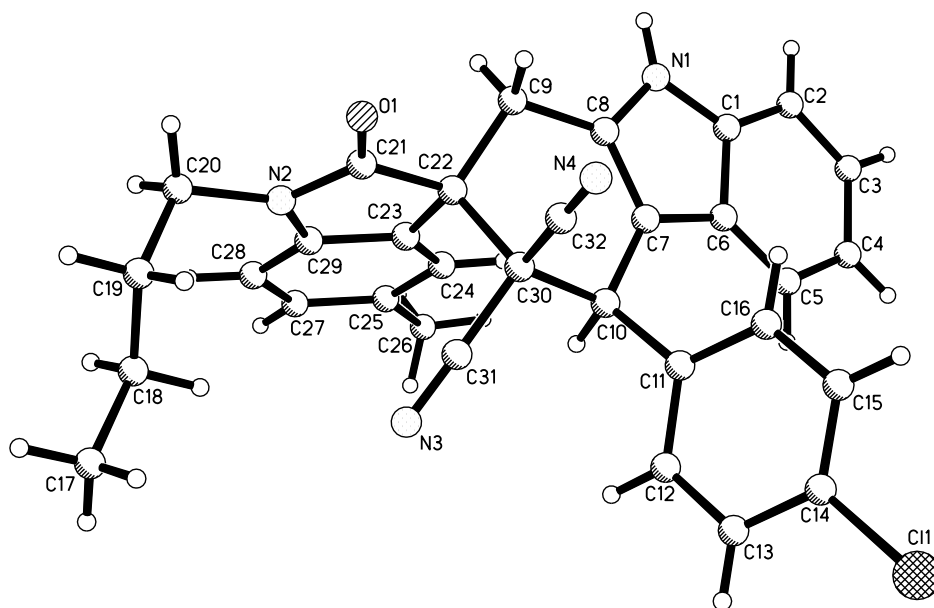


Fig. S3 ORTEP drawing (30%) of the crystal structure of **2g'**

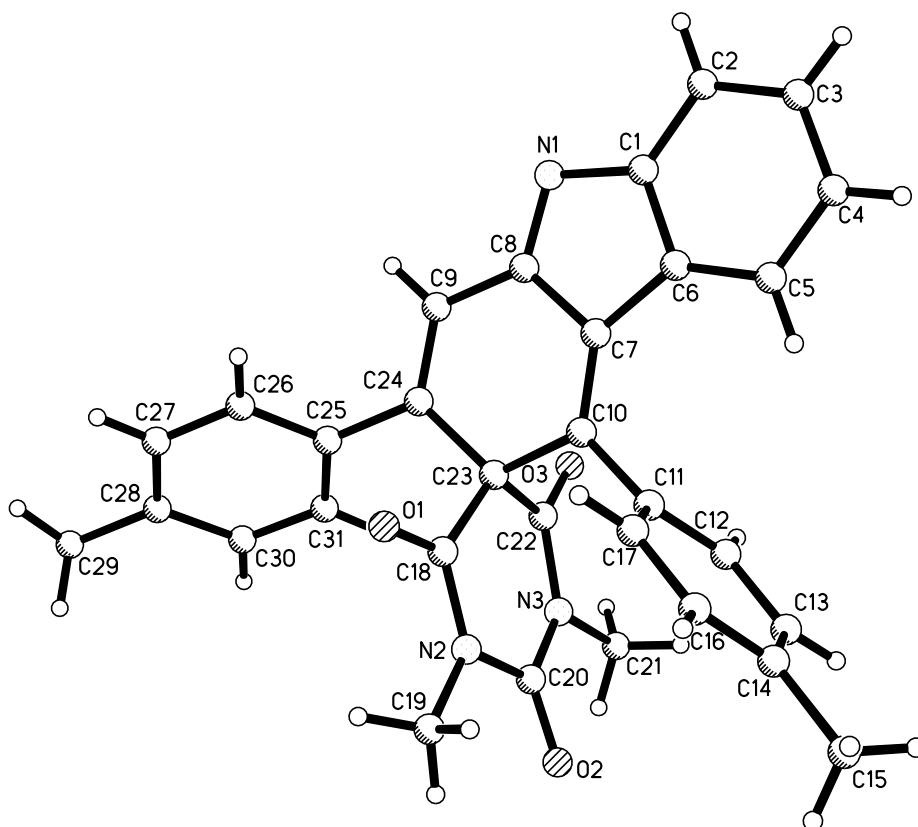


Fig. S4 ORTEP drawing (30%) of the crystal structure of **3a**

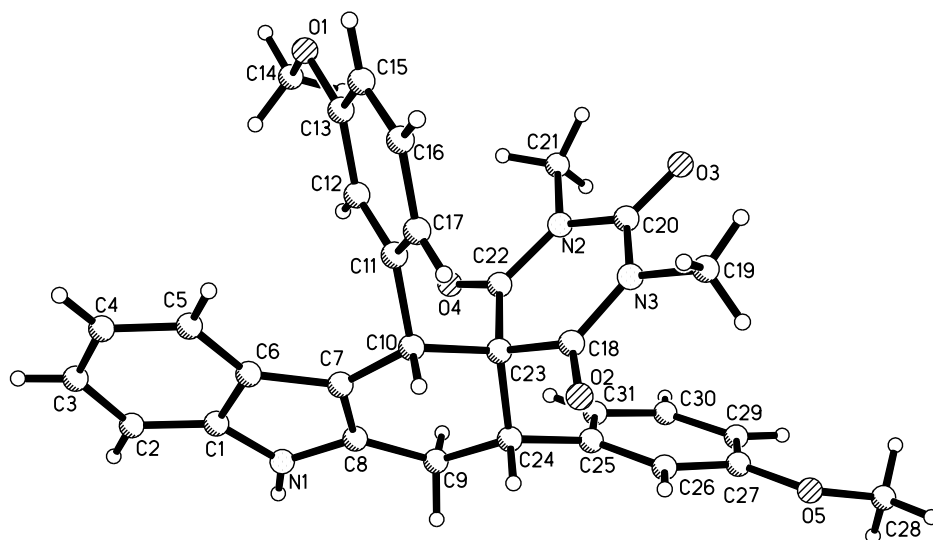


Fig. S5 ORTEP drawing (30%) of the crystal structure of **4e**

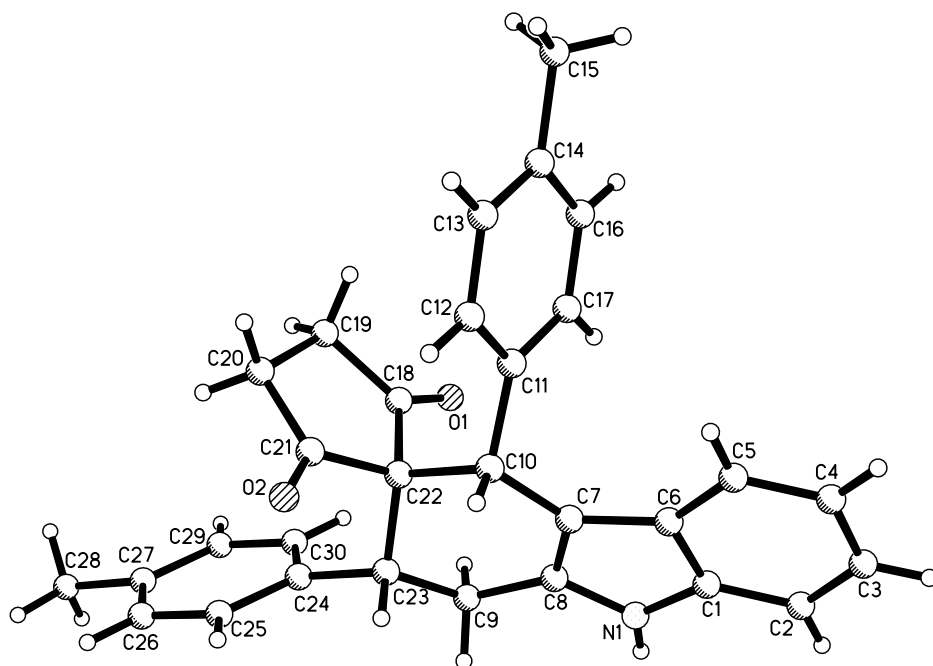


Fig. S6 ORTEP drawing (30%) of the crystal structure of **5b**

Table S1 The single crystal data of compounds **1f**, **2b**

Phase	1f	2b
Empirical formula	C ₄₀ H ₃₀ ClFN ₂ O ₂	C ₃₅ H ₂₆ N ₄ O
Formula weight	625.11	518.60
Temperature/K	296(2) K	296(2) K
Wavelength/ Å	0.71073	0.71073
Crystal system	Orthorhombic	Monoclinic
Space group	Pbca	P2(1)/n
<i>a</i> / Å	10.6504(5)	17.974(8)
<i>b</i> / Å	20.2099(12)	8.236(4)
<i>c</i> / Å	29.3774(14)	21.725(9)
α (°)	90	90
β (°)	90	109.319(11)
γ (°)	90	90
<i>V</i> (Å ³)	6323.3(6)	3035(2)
<i>Z</i>	8	4
Calculated density (g·cm ⁻³)	1.313	1.135
Absorption coefficient(mm ⁻¹)	0.166	0.070
<i>F</i> (000)	2608	1088
θ range /(°)	2.015 to 24.999 deg.	2.401 to 24.995
Limiting indices	- 12<= <i>h</i> <=10, - 21<= <i>k</i> <=24, -34<= <i>l</i> <=34	-21<= <i>h</i> <=21, -9<= <i>k</i> <=9, -25<= <i>l</i> <=23
Reflections collected/unique	49453 / 5558 [<i>R</i> (int) = 0.0996]	20790 / 5259 [<i>R</i> (int) = 0.1344]
Completeness to theta	99.9 %	98.4 %
Max. and min. transmission	0.7456 and 0.6685	0.7456 and 0.6320
Refinement method	Full-matrix least-squares on <i>F</i> ²	Full-matrix least-squares on <i>F</i> ²
Data/restraints/parameters	5558 / 0 / 417	5259 / 0 / 362
Goodness-of-fit on <i>F</i> ²	1.054	1.016
Final <i>R</i> indices[<i>I</i> >2σ(<i>I</i>)]	<i>R</i> 1 = 0.0796, <i>wR</i> 2 = 0.2216	<i>R</i> 1 = 0.0801, <i>wR</i> 2 = 0.2005
<i>R</i> indices (all data)	<i>R</i> 1 = 0.1537, <i>wR</i> 2 = 0.2491	<i>R</i> 1 = 0.1978, <i>wR</i> 2 = 0.2542
Largest diff. peak and hole /(e · Å ⁻³)	1.473 and -0.443	0.246 and -0.212

Table S2The single crystal data of compounds **2g'**, **3a**

Phase	2g'	3a
Empirical formula	C ₃₃ H ₂₈ Cl ₄ N ₄ O	C ₃₁ H ₂₅ N ₃ O ₃
Formula weight	638.39	487.54
Temperature/K	296(2) K	296(2) K
Wavelength/ Å	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic
Space group	P2(1)/c	P2(1)/n
<i>a</i> / Å	10.4591(4)	12.6929(8)
<i>b</i> / Å	12.0856(5)	14.6126(8)
<i>c</i> / Å	24.9411(10)	14.7027(7)
α (°)	90	90
β (°)	93.7861(13)	113.5420(18)
γ (°)	90	90
<i>V</i> (Å ³)	3145.8(2)	2500.0(2)
<i>Z</i>	8	4
Calculated density (g·cm ⁻³)	1.348	1.295
Absorption coefficient(mm ⁻¹)	0.409	0.085
<i>F</i> (000)	1320	1024
θ range /(°)	2.349 to 26.000 deg.	2.275 to 25.995
Limiting indices	-12<= <i>h</i> <=12, -12<= <i>k</i> <=14, -30<= <i>l</i> <=30	-15<= <i>h</i> <=13, -18<= <i>k</i> <=17, -18<= <i>l</i> <=17
Reflections collected/unique	29030 / 6165 [<i>R</i> (int) = 0.0353]	23220 / 4903 [<i>R</i> (int) = 0.0554]
Completeness to theta	99.8 %	99.7 %
Max. and min. transmission	0.7456 and 0.6750	0.7456 and 0.6972
Refinement method	Full-matrix least-squares on <i>F</i> ²	Full-matrix least-squares on <i>F</i> ²
Data/restraints/parameters	6165 / 0 / 381	4903 / 0 / 339
Goodness-of-fit on <i>F</i> ²	1.020	1.029
Final <i>R</i> indices[<i>I</i> >2sigma(<i>I</i>)]	<i>R</i> 1 = 0.0572, <i>wR</i> 2 = 0.1346	<i>R</i> 1 = 0.0495, <i>wR</i> 2 = 0.1057
<i>R</i> indices (all data)	<i>R</i> 1 = 0.0882, <i>wR</i> 2 = 0.1538	<i>R</i> 1 = 0.1077, <i>wR</i> 2 = 0.1285
Largest diff. peak and hole /(e · Å ⁻³)	0.598 and -0.516	0.194 and -0.149

Table S3 The single crystal data of compounds **4e**, **5b**

Phase	4e	5b
Empirical formula	C ₃₁ H ₂₉ N ₃ O ₅	C ₃₁ H ₂₈ Cl ₃ NO ₂
Formula weight	523.57	552.89
Temperature/K	296(2) K	296(2) K
Wavelength/ Å	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic
Space group	P2(1)/c	P2(1)/c
<i>a</i> / Å	9.1112(5)	9.0519(14)
<i>b</i> / Å	32.6212(16)	12.354(2)
<i>c</i> / Å	17.7777(10)	25.164(5)
α (°)	90	90
β (°)	91.0593(17)	99.596(5)
γ (°)	90	90
<i>V</i> (Å ³)	5283.0(5)	2774.6(8)
<i>Z</i>	8	4
Calculated density (g·cm ⁻³)	1.317	1.324
Absorption coefficient(mm ⁻¹)	0.090	0.359
<i>F</i> (000)	2208	1152
θ range /(°)	2.196 to 25.999	2.327 to 25.998
Limiting indices	-11<= <i>h</i> <=11, -40<= <i>k</i> <=40, -21<= <i>l</i> <=21	-11<= <i>h</i> <=10, -14<= <i>k</i> <=15, -31<= <i>l</i> <=27
Reflections collected/unique	46537 / 10364 [<i>R</i> (int) = 0.0880]	23988 / 5440 [<i>R</i> (int) = 0.1160]
Completeness to theta	99.8 %	99.9 %
Max. and min. transmission	0.7455 and 0.6933	0.7456 and 0.4919
Refinement method	Full-matrix least-squares on <i>F</i> ²	Full-matrix least-squares on <i>F</i> ²
Data/restraints/parameters	10364 / 65 / 711	5440 / 0 / 336
Goodness-of-fit on <i>F</i> ²	1.022	1.020
Final <i>R</i> indices[<i>I</i> >2σ(<i>I</i>)]	<i>R</i> 1 = 0.0745, <i>wR</i> 2 = 0.1512	<i>R</i> 1 = 0.0803, <i>wR</i> 2 = 0.1909
<i>R</i> indices (all data)	<i>R</i> 1 = 0.1792, <i>wR</i> 2 = 0.1920	<i>R</i> 1 = 0.1849, <i>wR</i> 2 = 0.2480
Largest diff. peak and hole /(e · Å ⁻³)	0.600 and -0.238	0.392 and -0.495

Experimental section

Unless noted, the commercial reagents and solvents were used without further purification. Melting points were recorded with a micromelting point apparatus and are uncorrected. IR spectra were recorded using a Bruker Tensor 27 spectrometer (KBr disc). The ^1H and $^{13}\text{C}\{^1\text{H}\}$ NMR spectra were recorded with a Varian 400 spectrometer at 400 or 100 MHz. High-resolution mass spectra (HRMS) were recorded in ESI mode using a MicroTOF mass spectrometer. Single-crystal X-ray data were collected with a Bruker Smart APEX-2 CCD diffractometer. All reactions were monitored by thin-layer chromatography (TLC) using silica gel plates (silica gel 60 F254 0.25 mm), and components were monitored by observation under UV light (254 and 365 nm).

1. **General procedure for the synthesis of functionalized spiro[carbazole-3,3'-inolines]:** A mixture of 2-methyl-1*H*-indole (0.5 mmol, 1.0 equiv), aldehyde (0.6 mmol, 1.2 equiv), 3-methyleneoxindole (0.5 mmol, 1.0 equiv) and CuSO_4 (0.1 mmol, 0.2 equiv) in dry toluene (6.0 mL) was stirred at 110 °C for about three hours. After removing the solvent by evaporating at reduced pressure, the residue was subjected to column chromatography with ethyl acetate and light petroleum (v/v = 1:5–1:8) as eluent to give pure **1a–j** and **1a'–j'**.

2. **General procedure for the synthesis of functionalized spiro[carbazole-2,3'-indolines]:** A mixture of 2-methyl-1*H*-indole (0.5 mmol, 1.0 equiv), aldehyde (0.6 mmol, 1.2 equiv), 2-(1-benzyl-2-oxoindolin-3-ylidene)malononitrile (0.5 mmol, 1.0 equiv) and CuSO_4 (0.1 mmol, 0.2 equiv) in dry toluene (6.0 mL) was stirred at 110 °C for about three hours. After removing the solvent by evaporating at reduced pressure, the residue was subjected to column chromatography with ethyl acetate and light petroleum (v/v = 1:5-1:8) as eluent to give pure **2a–g** and **2a'–g'**.

3. **General procedure for the synthesis of functionalized spiro[carbazole-3,5'-pyrimidine]**

reaction: A mixture of 2-methyl-1*H*-indole (0.5 mmol, 1.0 equiv), aldehyde (0.6 mmol, 1.2 equiv), 5-arylidene-1,3-dimethylbaribituric acid (0.5 mmol, 1.0 equiv) and CuSO₄ (0.1 mmol, 0.2 equiv) in dry toluene (6.0 mL) was stirred at 110°C for about three hours. After removing the solvent by evaporating at reduced pressure, the mixture of the above obtained product and DDQ (1.0 mmol, 0.227 g, 2.0 equiv) in dry acetonitrile (10.0 mL) was stirred at room temperature for about four hours. After removing the solvent by evaporating at reduced pressure, the residue was subjected to column chromatography with ethyl acetate and light petroleum (v/v = 1:3–1:6) as eluent to give pure products **3a–c**.

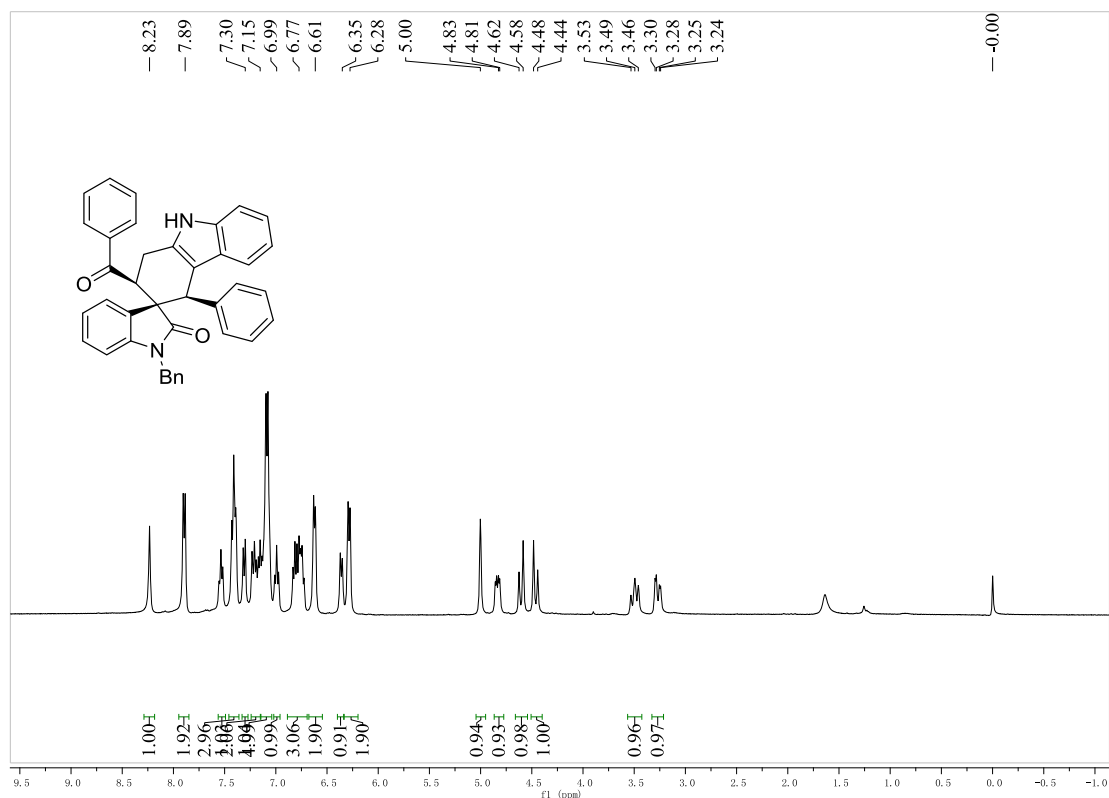
4. **General procedures for the synthesis of functionalized spiro[carbazole-3,3'-inolines]:** A mixture of 2-methyl-1*H*-indole (0.5 mmol, 1.0 equiv), aldehyde (1.2 mmol, 1.2 equiv) and 1,3-dimethylbaribituric acid (0.5 mmol, 1.0 equiv)) and CuSO₄ (0.1 mmol, 0.2 equiv) in dry toluene (6.0 mL) was stirred at 110°C for about three hours. After removing the solvent by evaporating at reduced pressure, the residue was subjected to column chromatography with ethyl acetate and light petroleum (v/v = 1:5–1:8) as eluent to give pure **4a–h**.

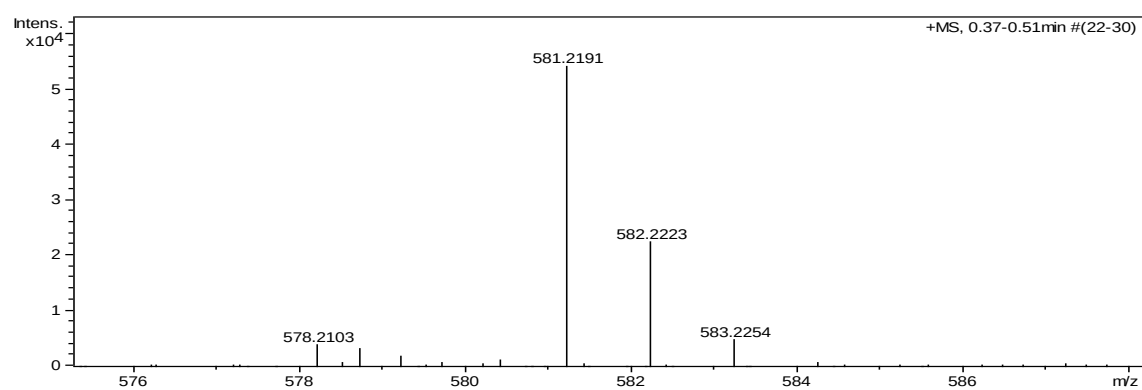
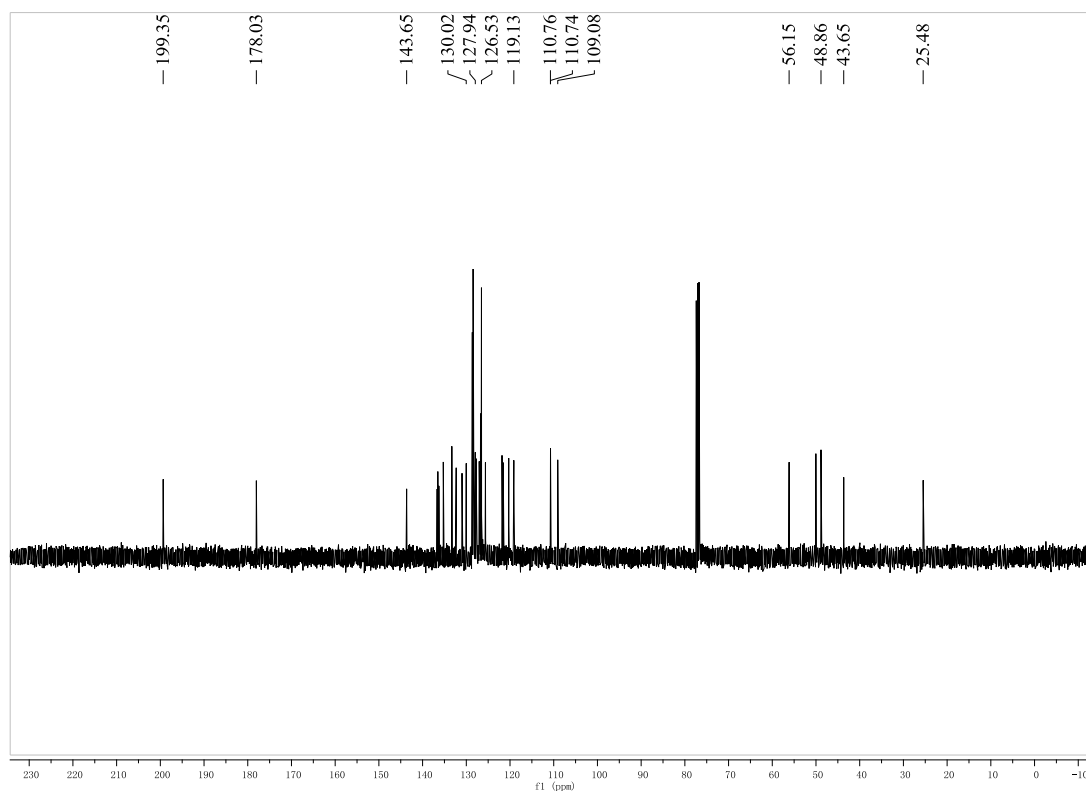
5. **General procedures for the synthesis of functionalized tetrahydrospiro[carbazole-3,1'-cyclopentane]-2',5'-diones and**

tetrahydrospiro[carbazole-3,1'-cyclohexane]-2',6'-diones: A mixture of 2-methyl-1*H*-indole (0.5 mmol, 1.0 equiv), aldehyde (1.2 mmol, 2.4 equiv), 1,3-diones (0.5 mmol, 1.0 equiv) and CuSO₄ (0.1 mmol, 0.2 equiv) in dry toluene (6.0 mL) was stirred at 110 °C for about three hours. After removing the solvent by evaporating at reduced pressure, the residue was subjected to column chromatography with ethyl acetate and light petroleum (v/v = 1:5–1:8) as eluent to give pure **5a–k**.

2-Benzoyl-1'-benzyl-4-phenyl-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1a):

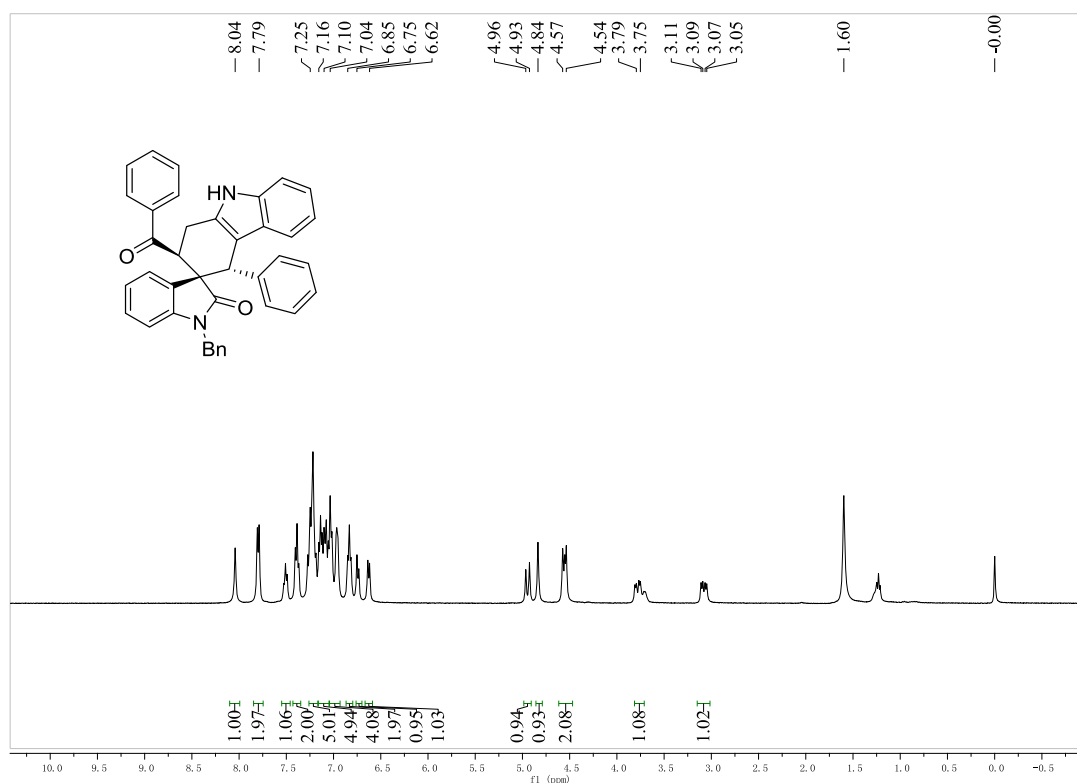
purple solid, 61%, m.p. 182-185 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.23 (s, 1H, NH), 7.89 (d, J = 7.2 Hz, 1H, ArH), 7.54 (t, J = 7.2 Hz, 1H, ArH), 7.43-7.39 (m, 3H, ArH), 7.30 (d, J = 8.0 Hz, 1H, ArH), 7.23-7.15 (m, 2H, ArH), 7.14-7.08 (m, 5H, ArH), 6.99 (t, J = 7.6 Hz, 1H, ArH), 6.83-6.73 (m, 3H, ArH), 6.63-6.62 (m, 2H, ArH), 6.36 (d, J = 7.6 Hz, 1H, ArH), 6.28 (t, J = 7.2 Hz, 2H, ArH), 5.00 (s, 1H, CH), 4.83 (dd, J_1 = 12.4 Hz, J_2 = 5.2 Hz, 1H, CH), 4.60 (d, J = 16.0 Hz, 1H, CH), 4.46 (d, J = 16.0 Hz, 1H, CH), 3.49 (t, J = 12.4 Hz, 1H, CH), 3.26 (dd, J_1 = 16.8 Hz, J_2 = 5.2 Hz, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 199.3, 178.0, 143.6, 136.6, 136.5, 136.2, 135.2, 133.3, 132.3, 130.9, 130.0, 128.7, 128.6, 128.4, 128.2, 127.9, 127.7, 127.0, 126.9, 126.7, 126.5, 125.6, 121.8, 121.5, 120.2, 119.1, 110.7, 110.7, 109.0, 56.1, 50.0, 48.8, 43.6, 25.4; IR (KBr) ν : 3367, 3210, 3155, 3017, 2980, 2831, 2864, 1877, 1623, 1611, 1507, 1456, 1355, 1241, 1178, 1143, 955, 931, 849, 789 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{39}\text{H}_{30}\text{N}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 581.2199, found: 581.2191.

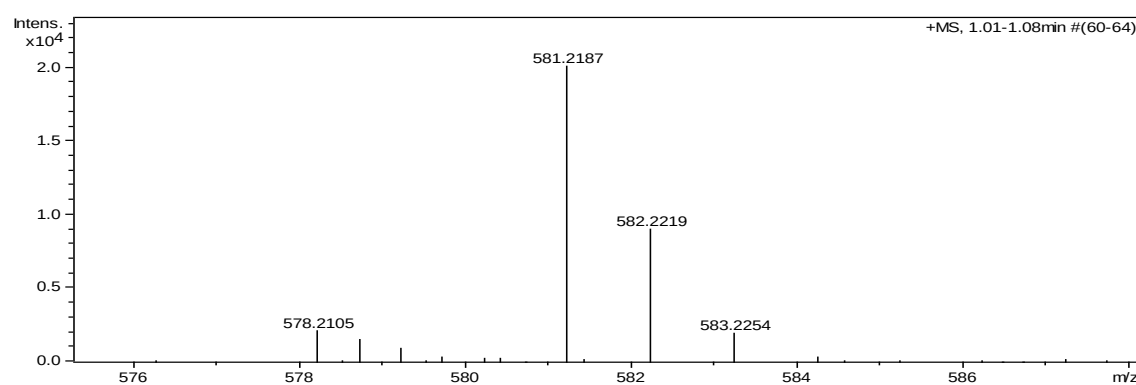
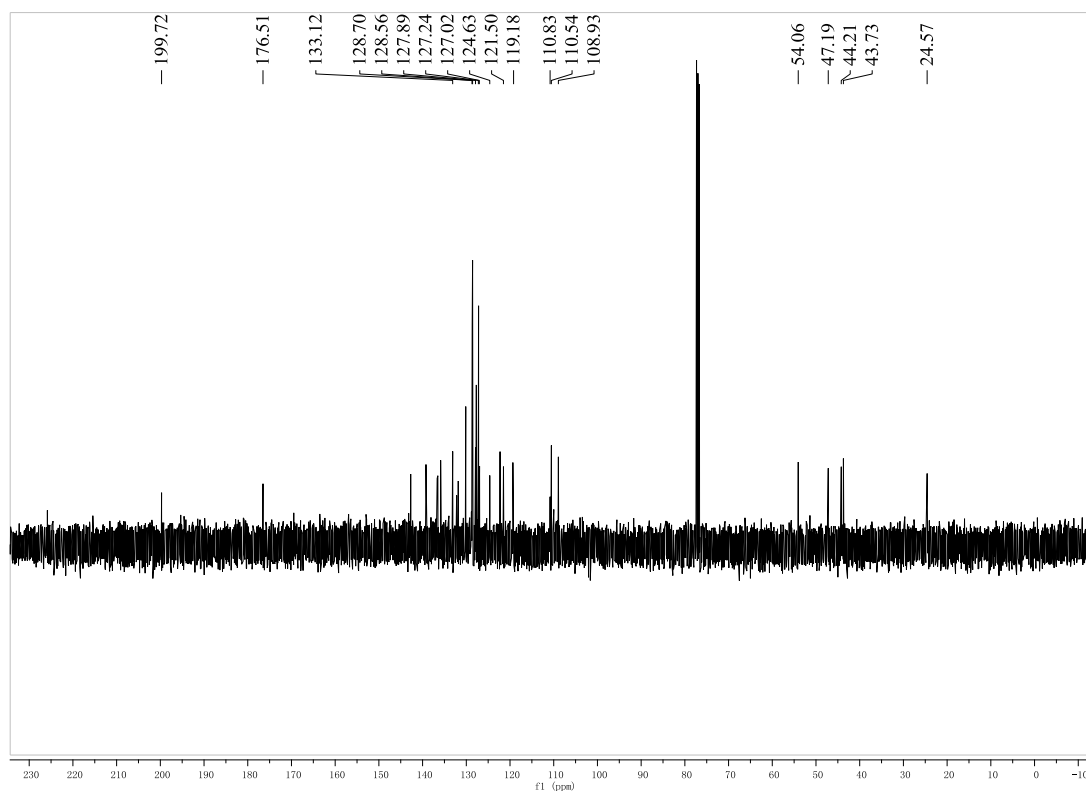




2-Benzoyl-1'-benzyl-4-phenyl-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1a'):

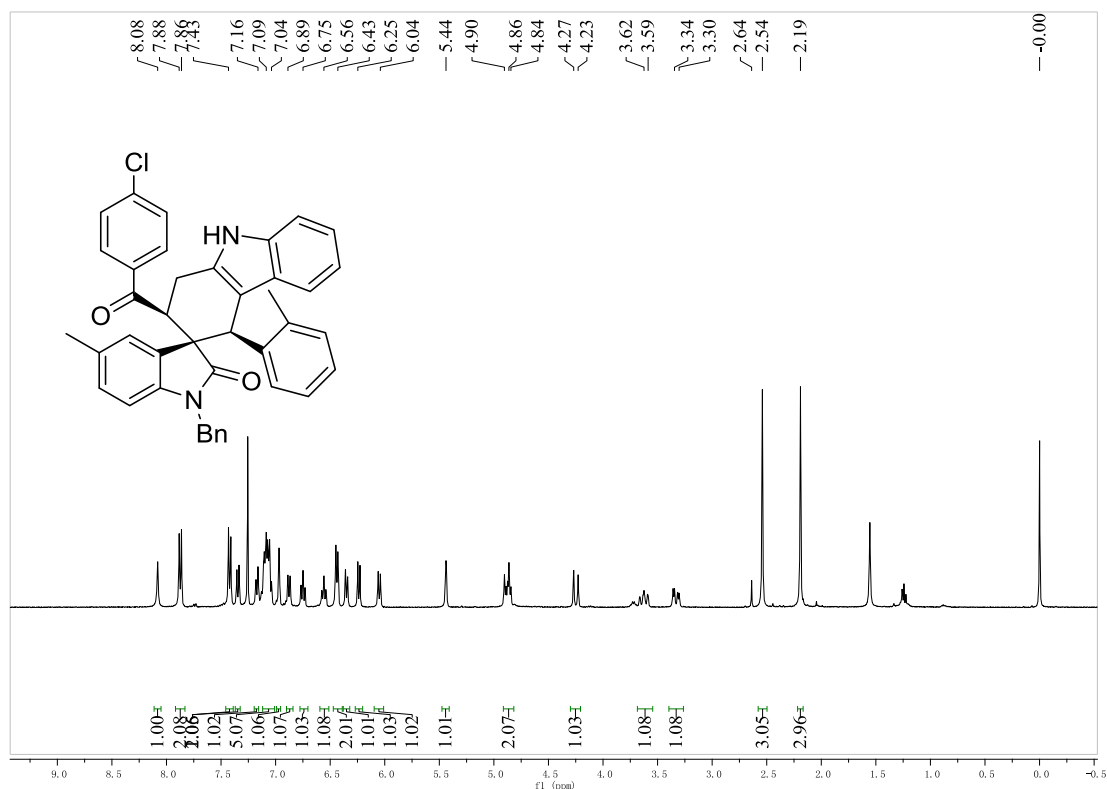
purple solid, 6%, m.p. 195-198 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.04 (s, 1H, NH), 7.79 (d, J = 7.6 Hz, 2H, ArH), 7.51 (t, J = 7.2 Hz, 1H, ArH), 7.39 (t, J = 7.2 Hz, 2H, ArH), 7.25-7.19 (m, 5H, ArH), 7.16-7.06 (m, 5H, ArH), 7.04-6.97 (m, 4H, ArH), 6.83 (d, J = 7.2 Hz, 2H, ArH), 6.75-6.73 (m, 1H, ArH), 6.62 (d, J = 7.6 Hz, 1H, ArH), 4.94 (d, J = 15.6 Hz, 1H, CH), 4.84 (s, 1H, CH), 4.56 (d, J = 15.6 Hz, 1H, CH), 4.55-4.53 (m, 1H, CH), 3.81-3.75 (m, 1H, CH), 3.07 (dd, J_1 = 16.8 Hz, J_2 = 7.6 Hz, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 199.7, 176.5, 142.7, 139.2, 136.6, 136.5, 135.8, 133.1, 132.2, 131.8, 130.1, 128.7, 128.6, 128.5, 128.5, 127.8, 127.7, 127.2, 127.1, 127.0, 126.9, 124.6, 122.2, 121.5, 119.3, 119.1, 110.8, 110.5, 108.9, 54.0, 47.1, 44.2, 43.7, 24.5; IR (KBr) ν : 3354, 3263, 3211, 3130, 3078, 2935, 2864, 2817, 2077, 1867, 1644, 1607, 1537, 1437, 1355, 1256, 1155, 1109, 997, 988, 845, 768 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{39}\text{H}_{30}\text{N}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 581.2199, found: 589.2187.

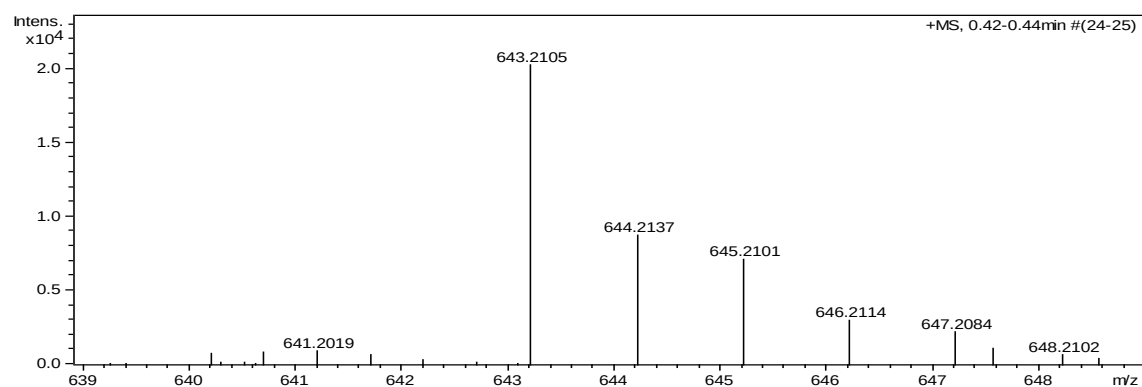
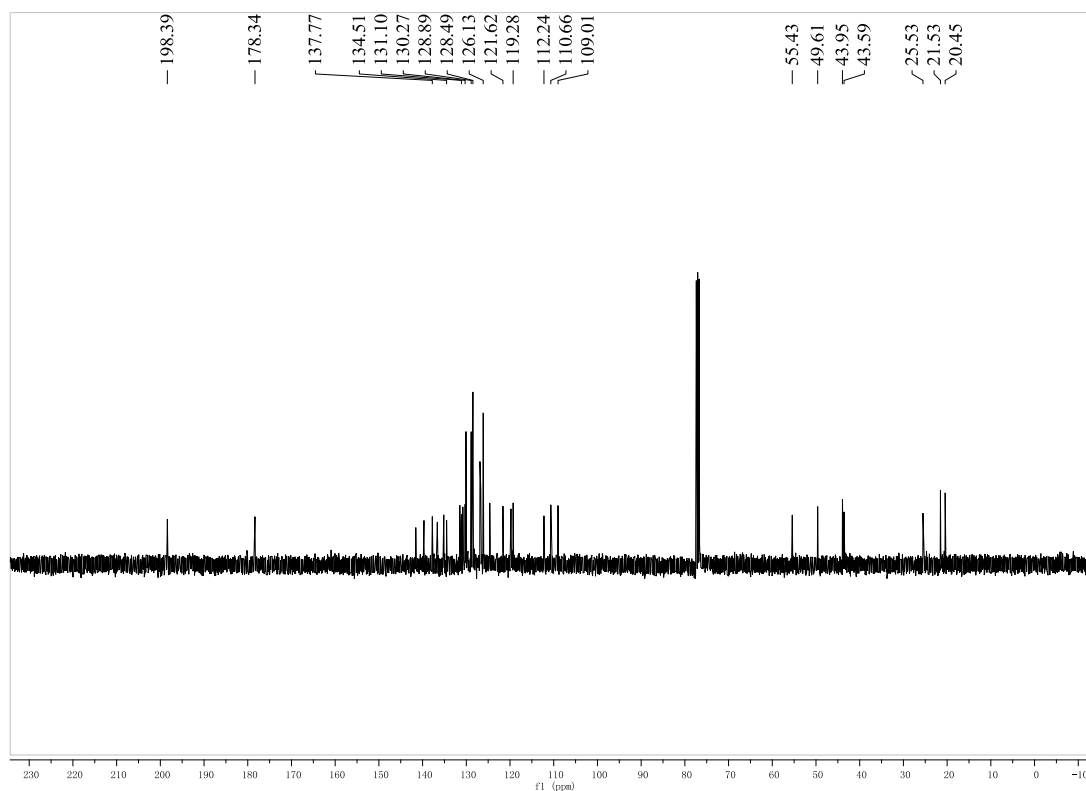




1'-Benzyl-2-(4-chlorobenzoyl)-5'-methyl-4-(*o*-tolyl)-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1b):

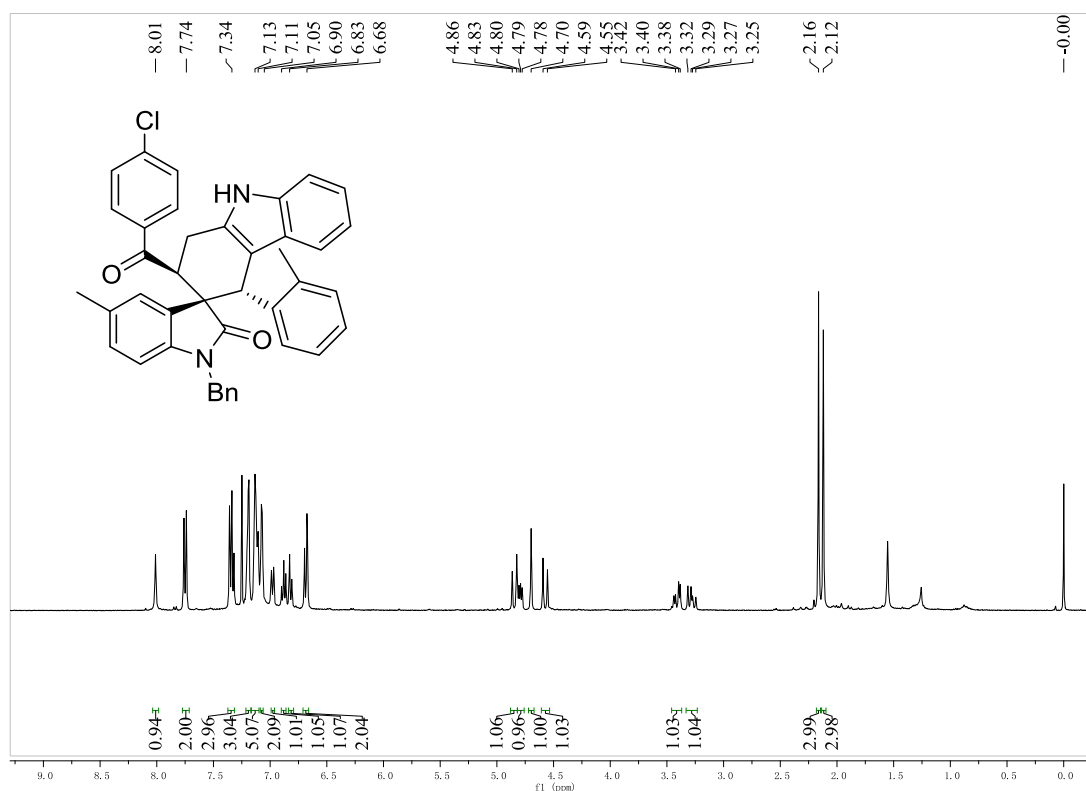
purple solid, 55%, m.p. 190-193 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.08 (s, 1H, NH), 7.87 (d, J = 8.4 Hz, 2H, ArH), 7.42 (d, J = 8.4 Hz, 2H, ArH), 7.34 (d, J = 8.0 Hz, 1H, ArH), 7.17 (d, J = 7.2 Hz, 1H, ArH), 7.10-7.04 (m, 5H, ArH), 6.99-6.97 (m, 1H, ArH), 6.87 (d, J = 8.0 Hz, 1H, ArH), 6.75 (t, J = 7.8 Hz, 1H, ArH), 6.56 (t, J = 7.2 Hz, 1H, ArH), 6.44 (d, J = 7.2 Hz, 2H, ArH), 6.35 (d, J = 8.0 Hz, 1H, ArH), 6.23 (d, J = 8.0 Hz, 1H, ArH), 6.05 (d, J = 8.0 Hz, 1H, ArH), 5.44 (s, 1H, CH), 4.90-4.84 (m, 2H, CH_2), 4.24 (d, J = 16.4 Hz, 1H, CH), 3.67-3.59 (m, 1H, CH), 3.32 (dd, J_1 = 16.8 Hz, J_2 = 5.2 Hz, 1H, CH), 2.54 (s, 3H, CH_3), 2.19 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 198.3, 178.3, 141.5, 139.6, 137.7, 136.6, 135.2, 135.1, 134.5, 131.4, 131.1, 130.7, 130.2, 130.0, 128.8, 128.5, 128.4, 126.8, 126.1, 124.6, 121.6, 119.7, 119.2, 112.2, 110.6, 109.0, 55.4, 49.6, 43.9, 43.5, 25.5, 21.5, 20.4; IR (KBr) ν : 3344, 3271, 3160, 3018, 2955, 2864, 2821, 1874, 1631, 1600, 1567, 1488, 1363, 1231, 1131, 1107, 955, 908, 841, 766 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{41}\text{H}_{33}\text{Cl}^{35}\text{N}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 643.2123, found: 643.2105. $\text{C}_{41}\text{H}_{33}\text{Cl}^{37}\text{N}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 645.2123, found: 645.2101.

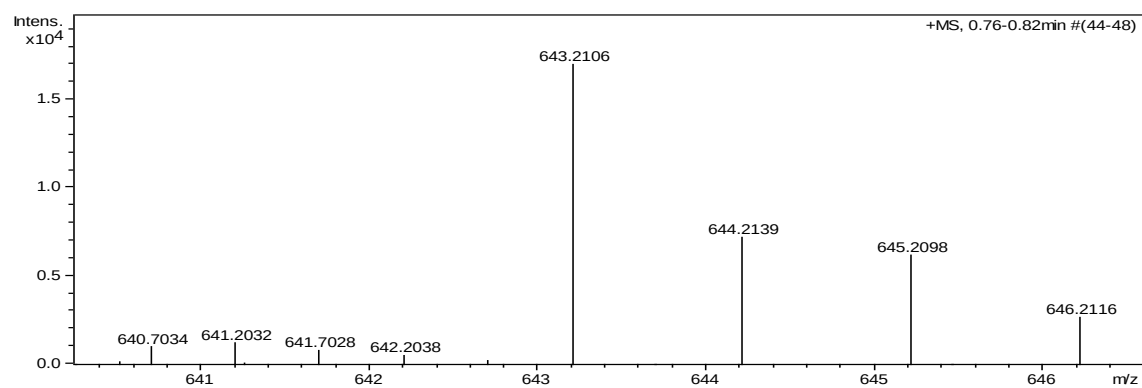
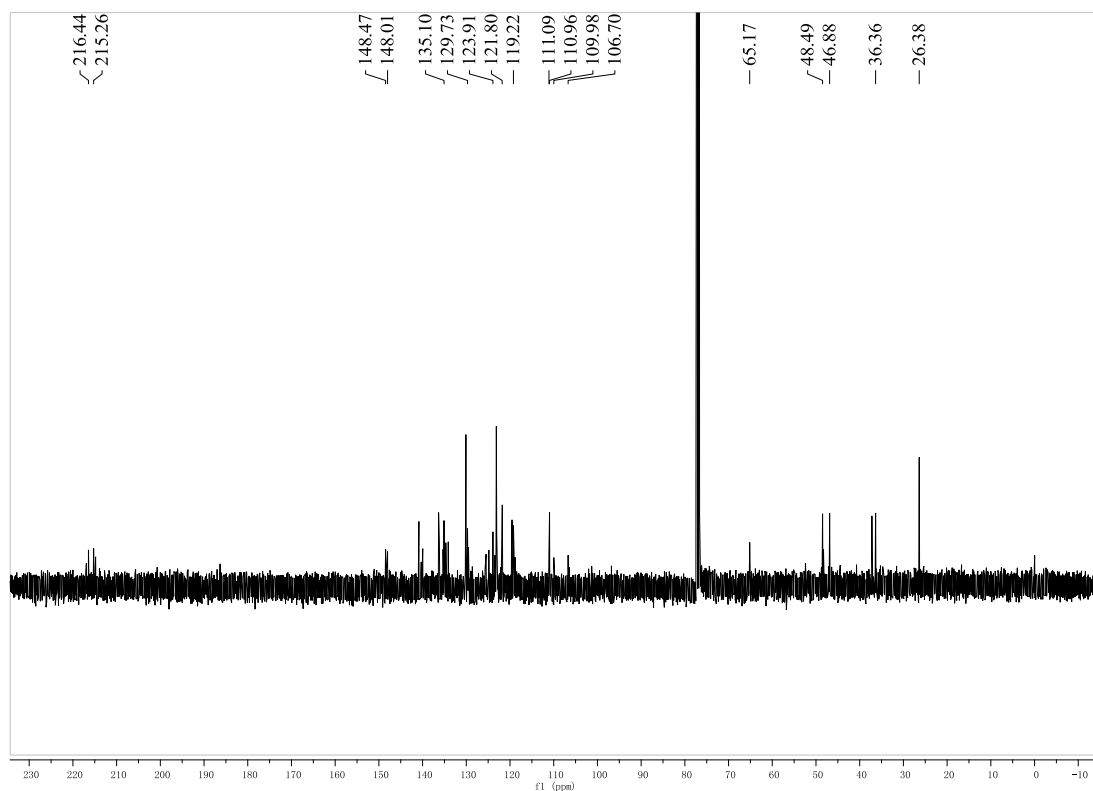




1'-Benzyl-2-(4-chlorobenzoyl)-5'-methyl-4-(*o*-tolyl)-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1b'):

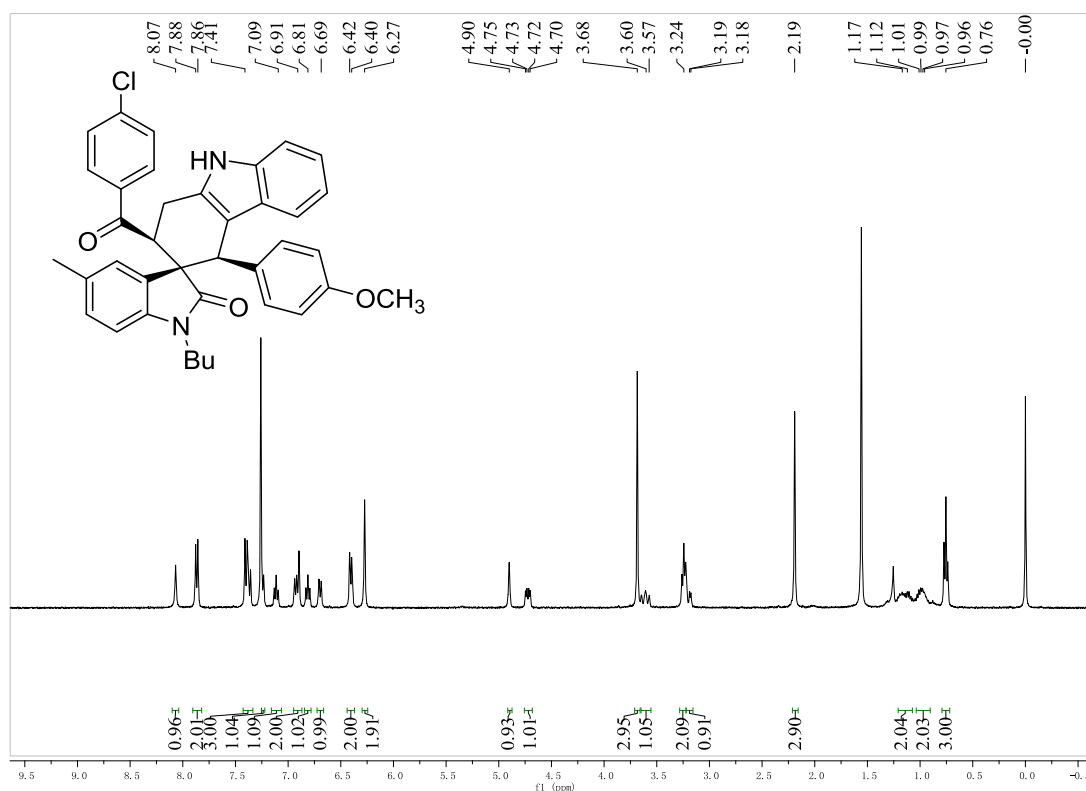
purple solid, 7%, m.p. 200-203 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.01 (s, 1H, NH), 7.75 (d, J = 8.8 Hz, 2H, ArH), 7.36-7.32 (m, 3H, ArH), 7.20-7.19 (m, 3H, ArH), 7.14-7.11 (m, 5H, ArH), 7.08-7.05 (m, 2H, ArH), 6.97 (d, J = 8.0 Hz, 1H, ArH), 6.88 (t, J = 7.2 Hz, 1H, ArH), 6.81 (d, J = 8.0 Hz, 1H, ArH), 6.68 (d, J = 8.4 Hz, 2H, ArH), 4.84 (d, J = 15.6 Hz, 1H, CH), 4.79 (t, J = 5.6 Hz, 1H, CH), 4.70 (s, 1H, CH), 4.57 (d, J = 15.6 Hz, 1H, CH), 3.41 (dd, J_1 = 16.8 Hz, J_2 = 5.6 Hz, 1H, CH), 3.28 (dd, J_1 = 16.8 Hz, J_2 = 11.6 Hz, 1H, CH), 2.16 (s, 3H, CH_3), 2.12 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 216.4, 215.2, 148.4, 148.0, 140.8, 139.9, 136.3, 136.1, 135.3, 135.0, 134.6, 134.1, 130.0, 129.7, 129.5, 125.4, 124.8, 123.9, 123.1, 123.0, 122.9, 121.8, 121.7, 119.6, 119.5, 119.2, 111.0, 110.9, 109.9, 106.7, 65.1, 48.4, 46.8, 37.2, 37.1, 36.3, 26.3; IR (KBr) ν : 3301, 3267, 3154, 3037, 2955, 2864, 2831, 2051, 1878, 1647, 1633, 1524, 1466, 1371, 1234, 1178, 1168, 944, 907, 862, 755 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{41}\text{H}_{33}\text{Cl}^{35}\text{N}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 643.2123, found: 643.2106. $\text{C}_{41}\text{H}_{33}\text{Cl}^{37}\text{N}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 644.2156, found: 644.2139.

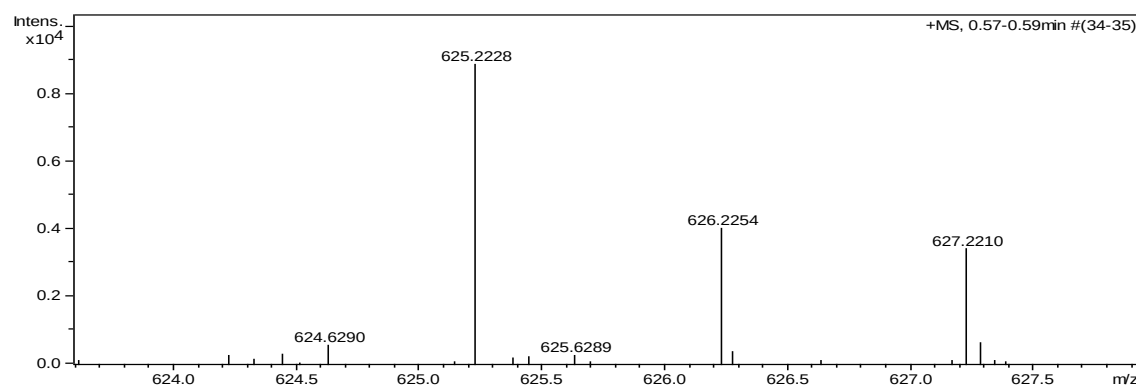
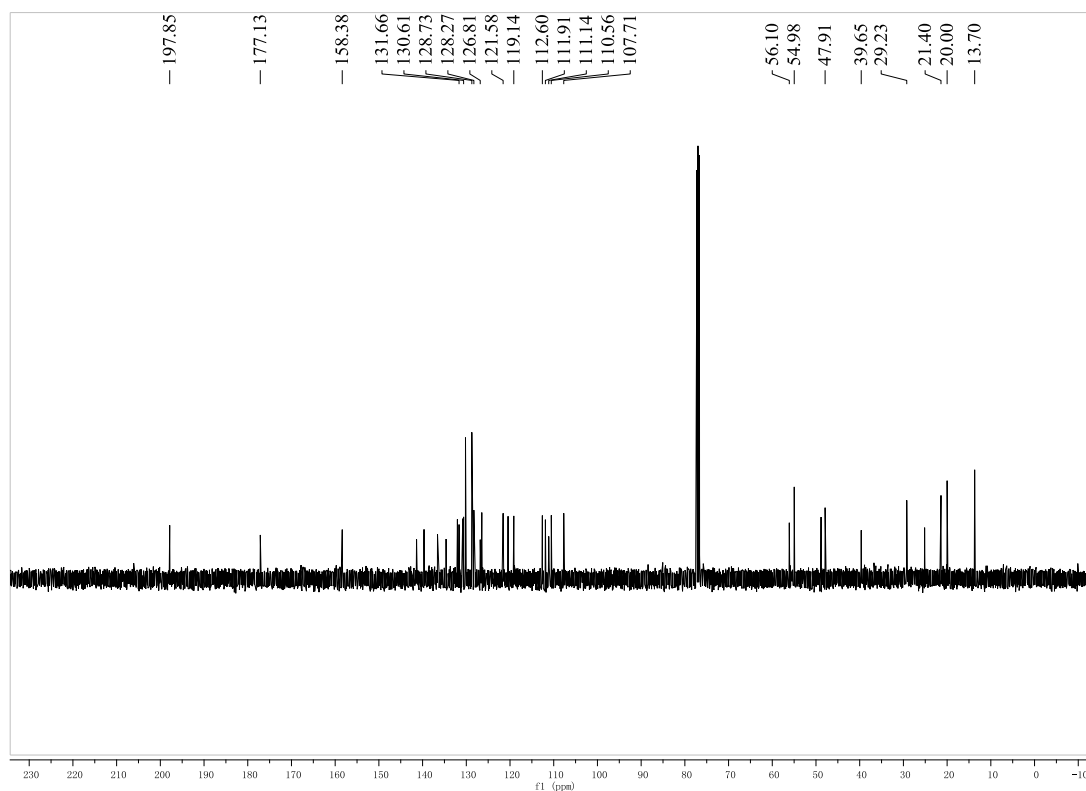




1'-Butyl-2-(4-chlorobenzoyl)-4-(4-methoxyphenyl)-5'-methyl-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1c):

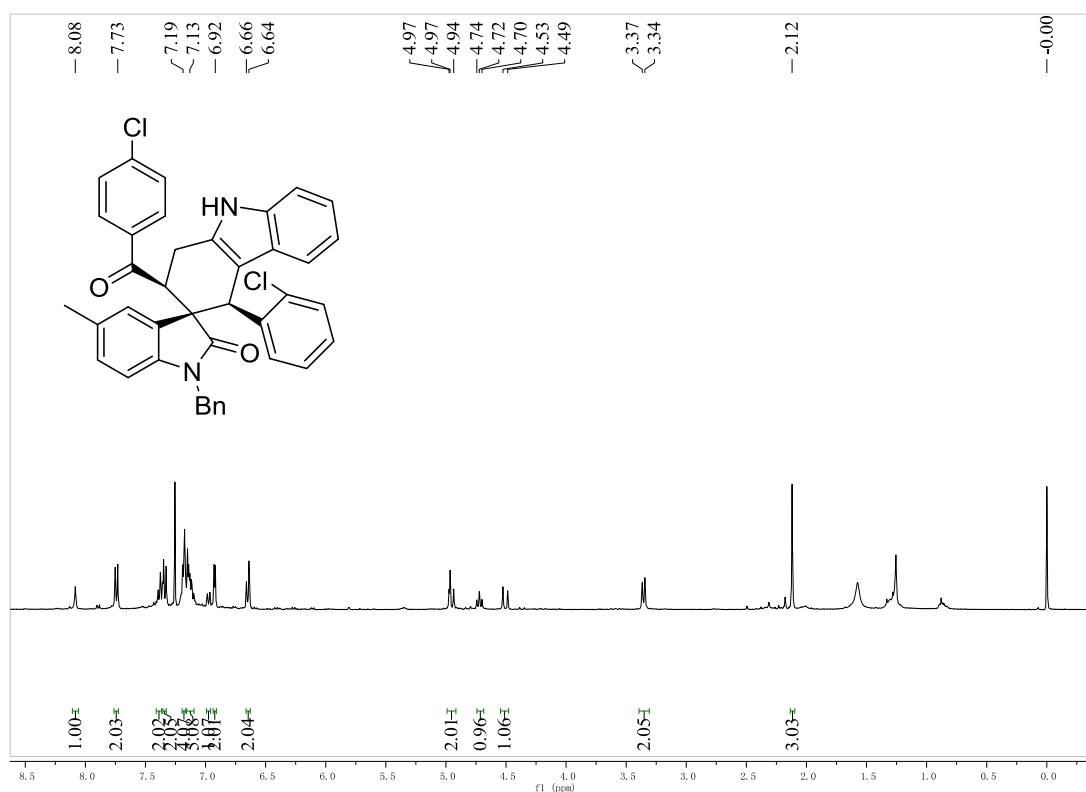
purple solid, 57%, m.p. 193-195 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.07 (s, 1H, NH), 7.86 (d, J = 8.4 Hz, 2H, ArH), 7.41-7.36 (m, 3H, ArH), 7.26-7.23 (m, 1H, ArH), 7.11 (t, J = 7.2 Hz, 1H, ArH), 6.94-6.90 (m, 2H, ArH), 6.81 (t, J = 7.2 Hz, 1H, ArH), 6.69 (d, J = 8.4 Hz, 1H, ArH), 6.40 (d, J = 7.6 Hz, 2H, ArH), 6.28-6.27 (m, 2H, ArH), 4.90 (s, 1H, CH), 4.72 (dd, J_1 = 12.4 Hz, J_2 = 5.2 Hz, 1H, CH), 3.69 (s, 3H, OCH_3), 3.65-3.57 (m, 1H, CH), 3.26-3.23 (m, 2H, CH_2), 3.18 (d, J = 5.2 Hz, 1H, CH), 2.19 (s, 3H, CH_3), 1.18-1.11 (m, 2H, CH_2), 1.01-0.96 (m, 2H, CH_2), 0.76 (t, J = 7.2 Hz, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 197.8, 177.1, 158.3, 141.3, 139.6, 136.5, 134.6, 132.0, 131.6, 130.8, 130.6, 130.1, 128.7, 128.5, 128.2, 128.1, 126.8, 126.4, 121.5, 120.4, 119.1, 112.6, 111.9, 111.1, 110.5, 107.7, 56.1, 54.9, 48.8, 47.9, 39.6, 29.2, 25.1, 21.4, 20.0, 13.7; IR(KBr) ν : 3362, 3301, 3256, 3144, 3067, 2963, 2811, 2750, 2143, 1831, 1654, 1611, 1542, 1466, 1355, 1241, 1100, 1054, 966, 931, 849, 772 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{38}\text{H}_{35}\text{Cl}^{35}\text{N}_2\text{O}_3$ ($[\text{M}+\text{Na}]^+$): 625.2228, found: 625.2228. $\text{C}_{38}\text{H}_{35}\text{Cl}^{37}\text{N}_2\text{O}_3$ ($[\text{M}+\text{Na}]^+$): 626.2262, found: 626.2254.

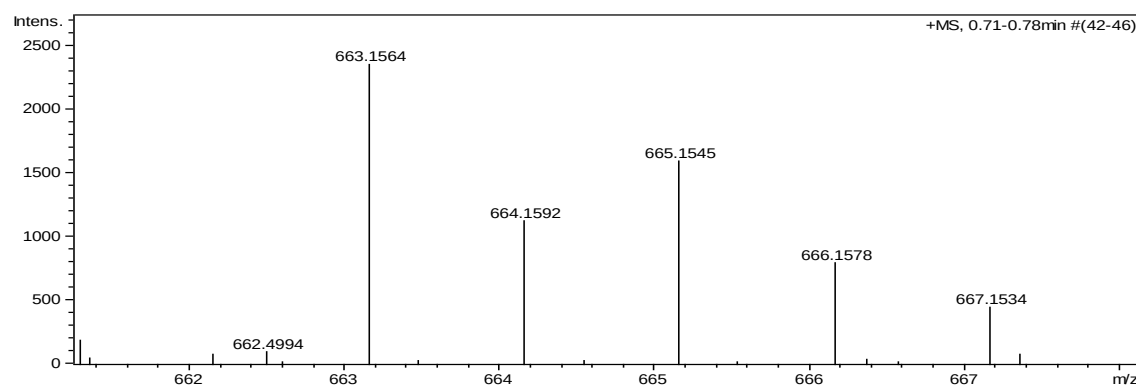
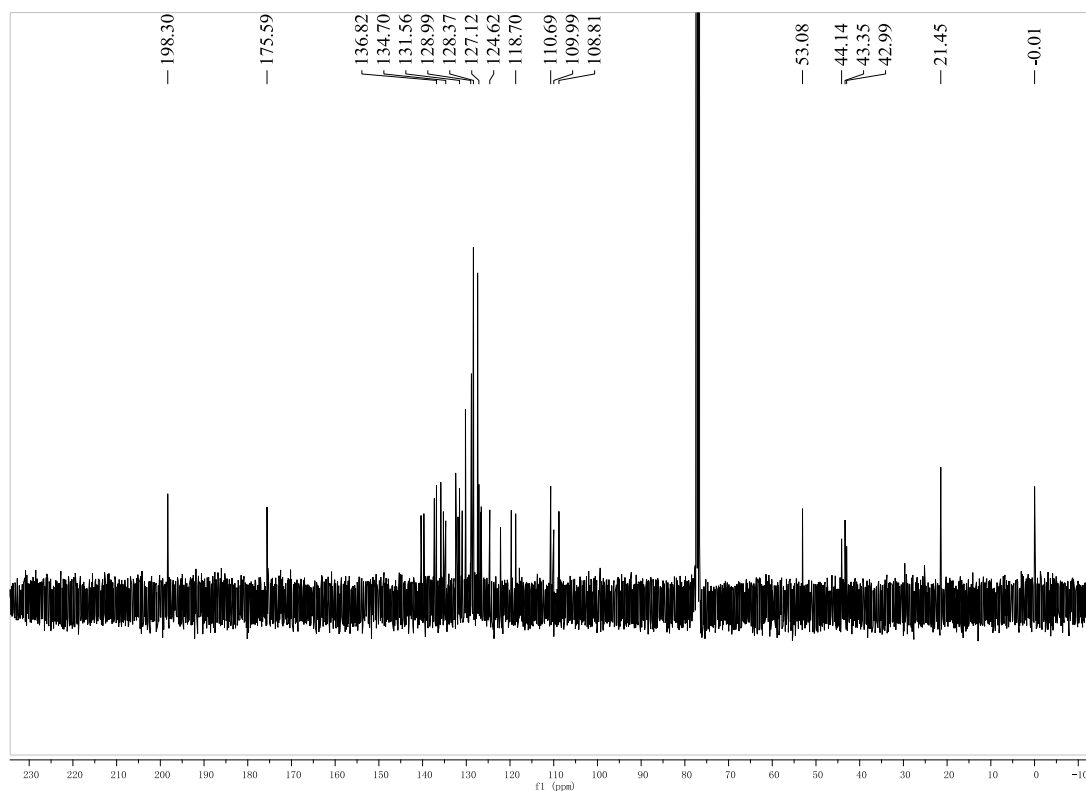




**1'-Benzyl-2-(4-chlorobenzoyl)-4-(2-chlorophenyl)-5'-methyl-1,2,4,9-tetrahydrospiro[carbazol
e-3,3'-indolin]-2'-one (1d):**

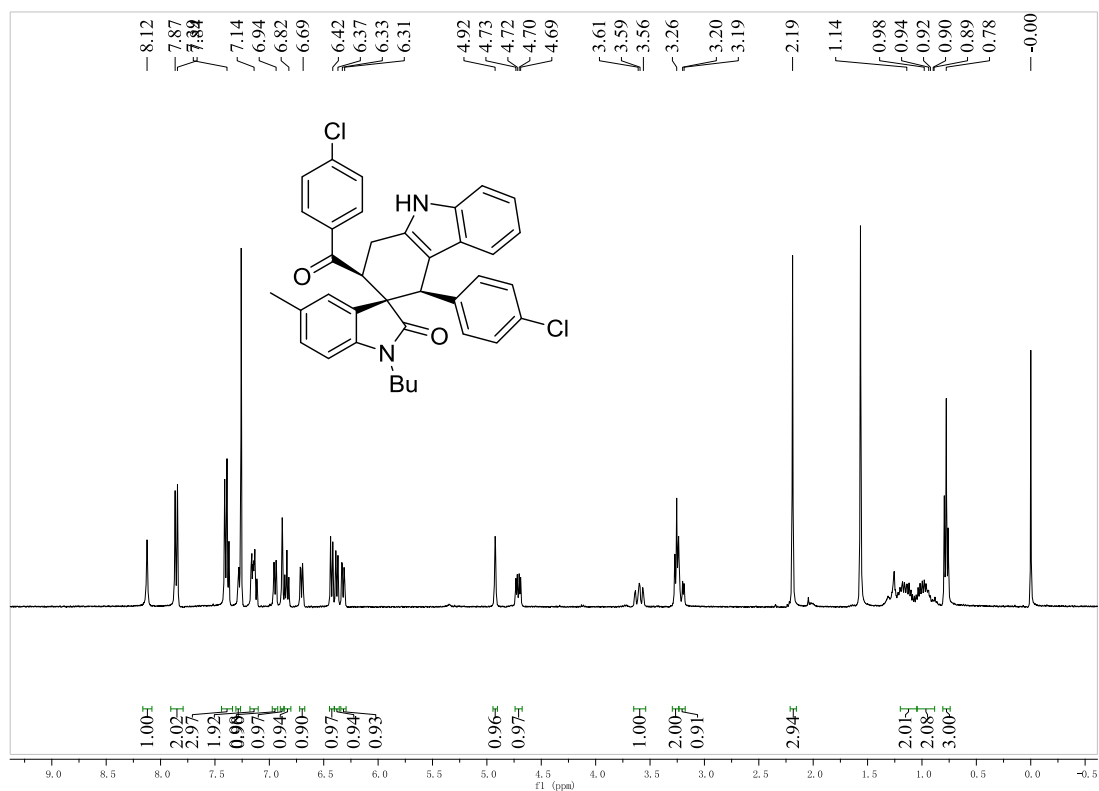
purple solid, 58%, m.p. 184-187 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.08 (s, 1H, NH), 7.73 (d, J = 8.4 Hz, 2H, ArH), 7.37 (d, J = 7.2 Hz, 2H, ArH), 7.35-7.32 (m, 2H, ArH), 7.18-7.16 (m, 4H, ArH), 7.14-7.11 (m, 5H, ArH), 6.96 (d, J = 8.0 Hz, 1H, ArH), 6.92 (d, J = 4.0 Hz, 2H, ArH), 6.64 (d, J = 8.0 Hz, 2H, ArH), 4.96 (s, 1H, CH), 4.94 (d, J = 15.6 Hz, 1H, CH), 4.71 (t, J = 8.4 Hz, 1H, CH), 4.58 (d, J = 16.0 Hz, 1H, CH), 3.34 (d, J = 8.4 Hz, 2H, CH_2), 2.11 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 198.3, 175.5, 140.3, 139.7, 137.3, 136.8, 135.8, 135.2, 134.7, 132.4, 131.8, 131.5, 130.9, 130.1, 128.9, 128.8, 128.4, 128.3, 128.3, 127.4, 127.1, 126.7, 126.5, 124.6, 122.1, 119.7, 118.7, 110.6, 109.9, 108.8, 53.0, 44.1, 43.3, 42.9, 21.4, 0.1 cm^{-1} ; IR (KBr) ν : 3341, 3305, 3248, 3103, 3054, 2954, 2866, 2760, 2149, 1860, 1653, 1667, 1553, 1470, 1344, 1249, 1150, 1140, 967, 933, 849, 761 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{40}\text{H}_{30}\text{Cl}_2^{35}\text{N}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 663.1577, found: 663.1564. $\text{C}_{40}\text{H}_{30}\text{Cl}_2^{37}\text{N}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 665.1547, found: 665.1545.

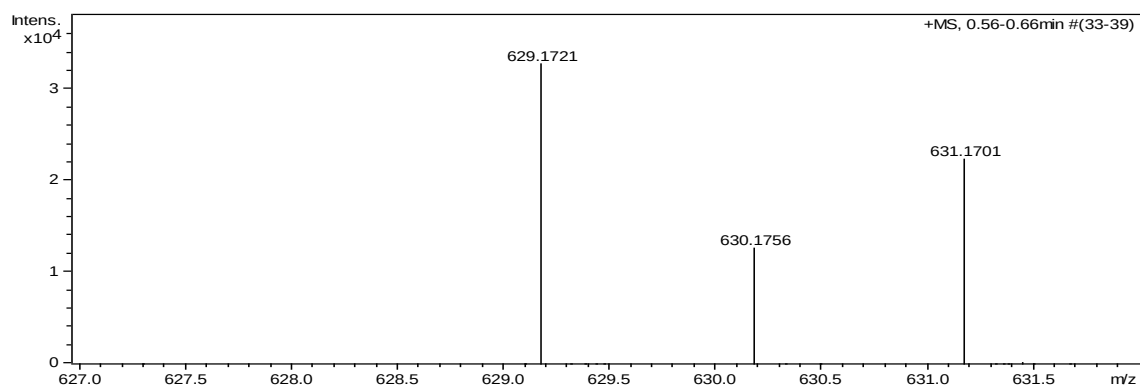
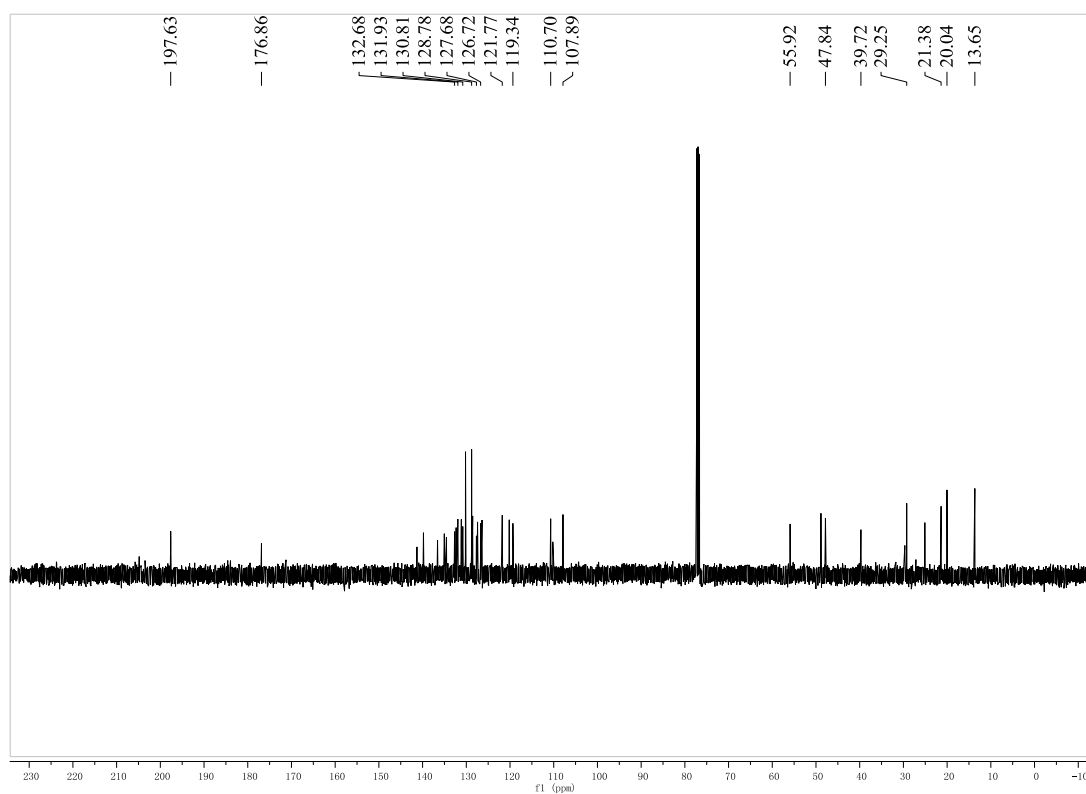




1'-Butyl-2-(4-chlorobenzoyl)-4-(4-chlorophenyl)-5'-methyl-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1e):

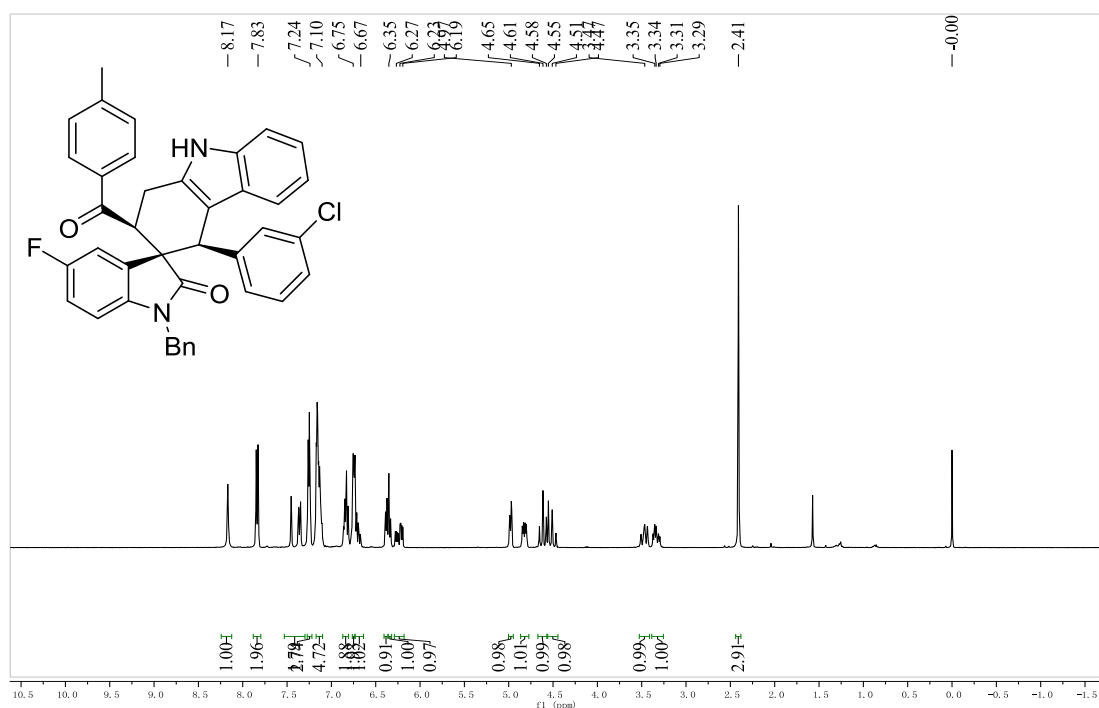
purple solid, 65%, m.p. 185-188 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.12 (s, 1H, NH), 7.85 (d, J = 8.4 Hz, 2H, ArH), 7.39 (t, J = 8.4 Hz, 3H, ArH), 7.29-7.28 (m, 1H, ArH), 7.17-7.12 (m, 2H, ArH), 6.94 (d, J = 7.6 Hz, 1H, ArH), 6.88-6.87 (m, 1H, ArH), 6.84 (t, J = 7.6 Hz, 1H, ArH), 6.70 (dd, J_1 = 8.4 Hz, J_2 = 2.0 Hz, 1H, ArH), 6.42 (d, J = 8.0 Hz, 1H, ArH), 6.37 (d, J = 8.0 Hz, 1H, ArH), 6.32 (dd, J_1 = 8.4 Hz, J_2 = 2.0 Hz, 1H, ArH), 4.93 (s, 1H, CH), 4.71 (dd, J_1 = 12.4 Hz, J_2 = 5.2 Hz, 1H, ArH), 3.64-3.56 (m, 1H, CH), 3.26 (q, J = 7.2 Hz, 2H, CH_2), 3.23-3.19 (m, 1H, CH), 2.19 (s, 3H, CH_3), 1.16-1.10 (m, 2H, CH_2), 1.02-0.89 (m, 2H, CH_2), 0.78 (t, J = 7.2 Hz, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 197.6, 176.8, 141.2, 139.8, 136.5, 135.0, 134.5, 132.6, 132.3, 131.9, 131.1, 130.8, 130.1, 128.7, 128.5, 127.6, 127.4, 126.7, 126.4, 121.7, 120.1, 119.3, 110.6, 107.8, 55.9, 48.8, 47.8, 39.7, 29.2, 25.0, 21.3, 20.0, 13.6; IR (KBr) ν : 3307, 3278, 3148, 3033, 2971, 2863, 2801, 2133, 1862, 1655, 1638, 1564, 1467, 1358, 1231, 1146, 1100, 948, 903, 843, 788 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{37}\text{H}_{32}\text{Cl}_2^{35}\text{N}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 629.1733, found: 629.1733. $\text{C}_{37}\text{H}_{32}\text{Cl}_2^{37}\text{N}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 631.1704, found: 631.1701.

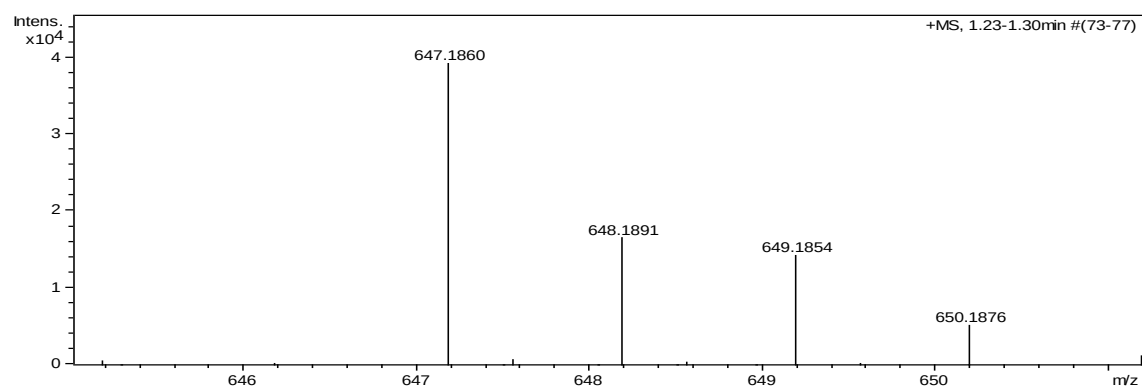
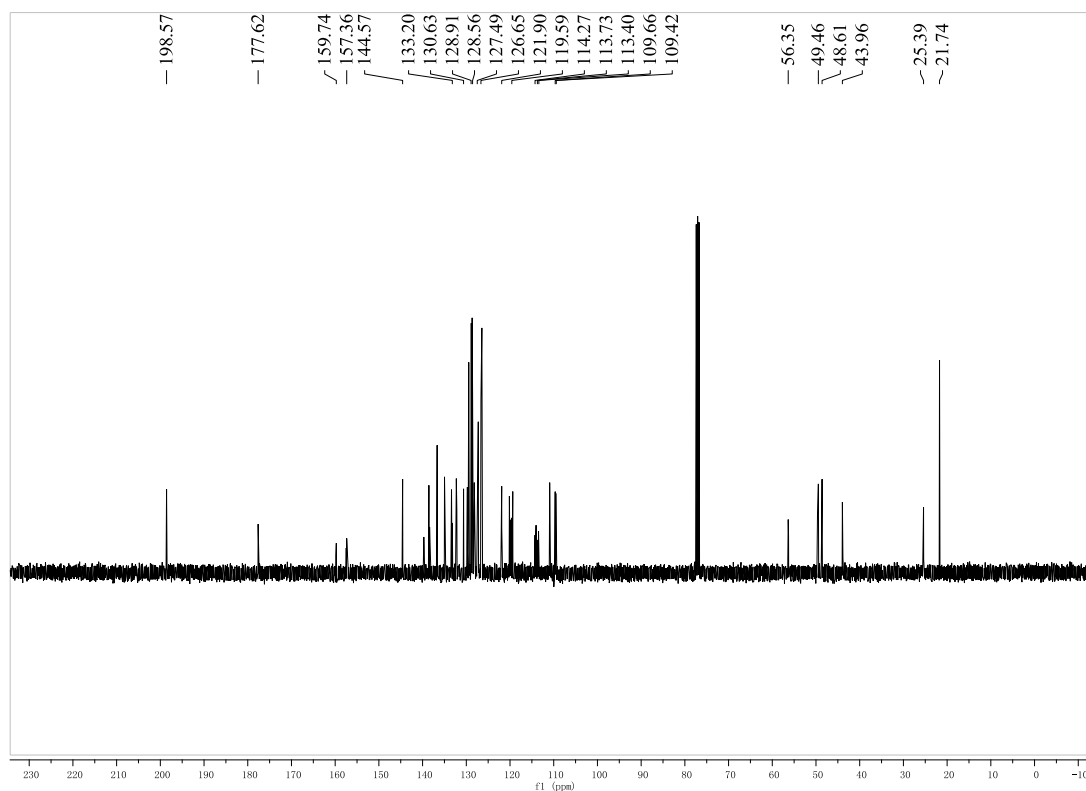




1'-Benzyl-4-(3-chlorophenyl)-5'-fluoro-2-(4-methylbenzoyl)-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1f):

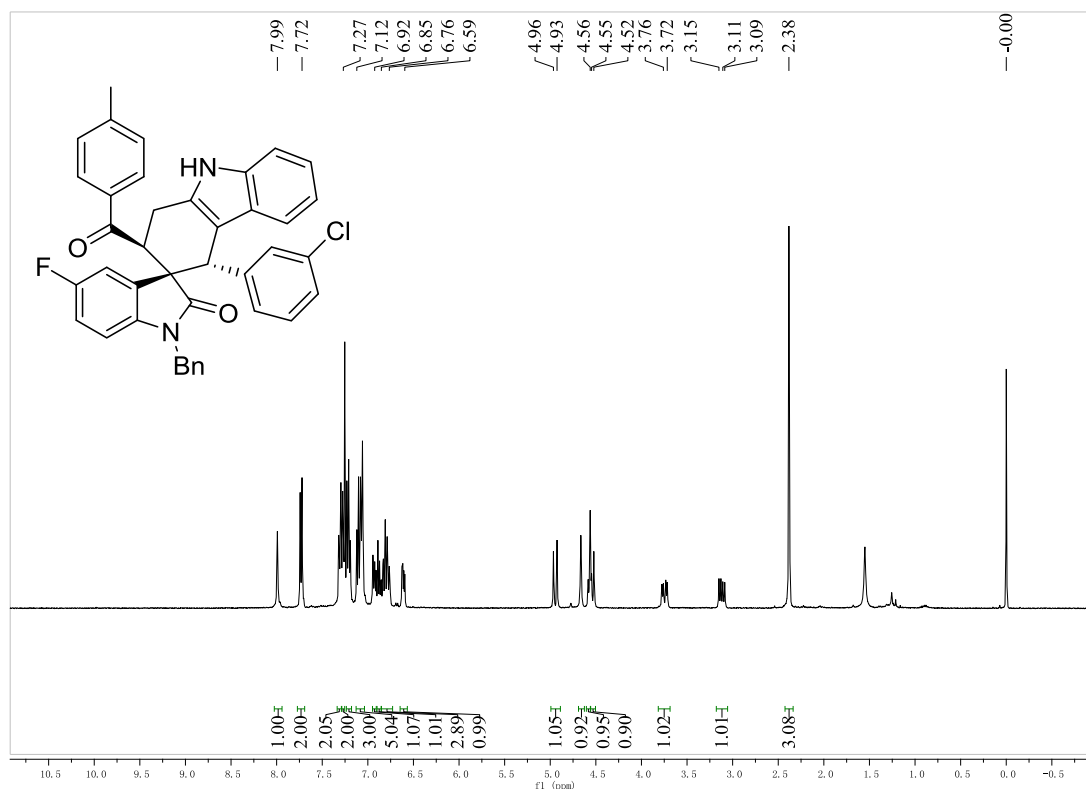
purple solid, 55%, m.p. 195-198 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.17 (s, 1H, NH), 7.83 (d, J = 8.0 Hz, 2H, ArH), 7.45-7.35 (m, 2H, ArH), 7.26-7.24 (m, 3H, ArH), 7.17-7.11 (m, 5H, ArH), 6.86-6.81 (m, 2H, ArH), 6.77-6.73 (m, 2H, ArH), 6.71-6.67 (m, 1H, ArH), 6.38 (d, J = 7.2 Hz, 1H, ArH), 6.34 (d, J = 7.6 Hz, 1H, ArH), 6.28-6.20 (m, 1H, ArH), 4.98 (d, J = 7.2 Hz, 1H, CH), 4.82 (dd, J_1 = 12.0 Hz, J_2 = 5.6 Hz, 1H, CH), 4.61 (t, J = 16.0 Hz, 1H, CH), 4.51 (t, J = 16.0 Hz, 1H, CH), 3.47 (t, J = 16.0 Hz, 1H, CH), 3.37-3.30 (m, 1H, CH), 2.41 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 198.5, 177.6, 159.7 (d, J = 11.0 Hz), 157.3 (d, J = 10.9 Hz), 144.5 (d, J = 2.6 Hz), 139.7 (d, J = 11.7 Hz), 138.4 (d, J = 16.2 Hz), 136.6, 134.9, 133.3, 133.2, 133.1, 132.2 (d, J = 11.8 Hz), 130.6, 129.7, 129.4, 128.9, 128.8 (d, J = 10.2 Hz), 128.5, 127.4 (d, J = 9.8 Hz), 127.2, 126.6, 126.4 (d, J = 9.2 Hz), 121.9 (d, J = 22.0 Hz), 119.9 (d, J = 21.8 Hz), 119.3 (d, J = 21.8 Hz), 114.3 (d, J = 11.5 Hz), 114.1 (d, J = 11.2 Hz), 113.7 (d, J = 8.3 Hz), 113.4 (d, J = 7.8 Hz), 110.9, 109.6 (d, J = 5.9 Hz), 109.5 (d, J = 10.6 Hz), 56.3, 49.4, 48.6, 43.9, 25.3, 21.7; IR (KBr) ν : 3299, 3217, 3166, 3031, 2973, 2855, 2814, 2168, 1859, 1643, 1600, 1578, 1463, 1354, 1280, 1167, 1139, 983, 955, 883, 755 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{40}\text{H}_{30}\text{Cl}^{35}\text{FN}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 647.1872, found: 647.1860. $\text{C}_{40}\text{H}_{30}\text{Cl}^{37}\text{FN}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 648.1906, found: 648.1891.

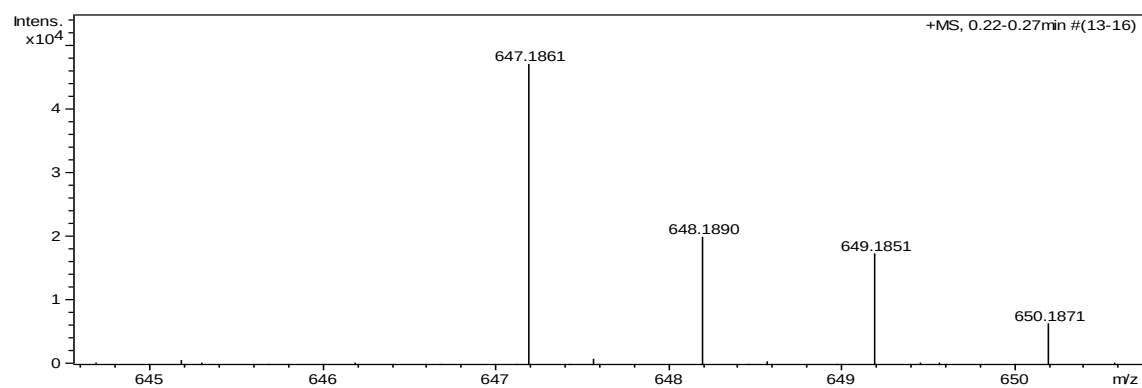
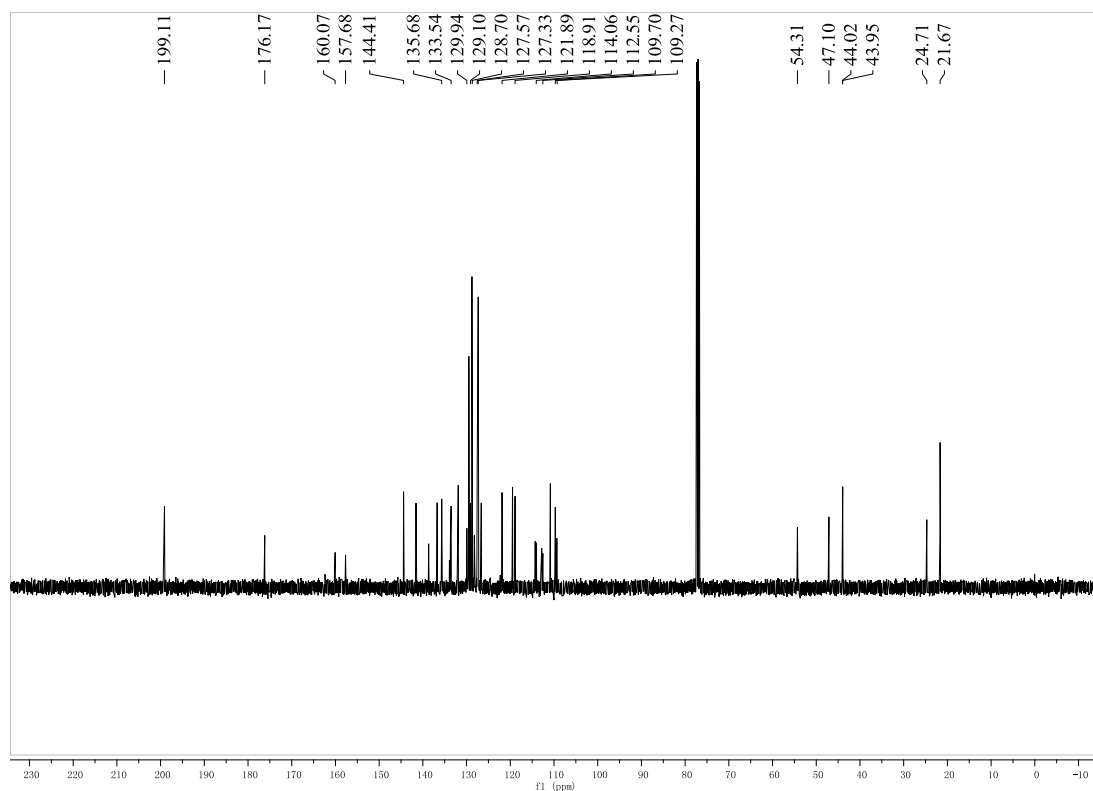




1'-Benzyl-4-(3-chlorophenyl)-5'-fluoro-2-(4-methylbenzoyl)-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1f'):

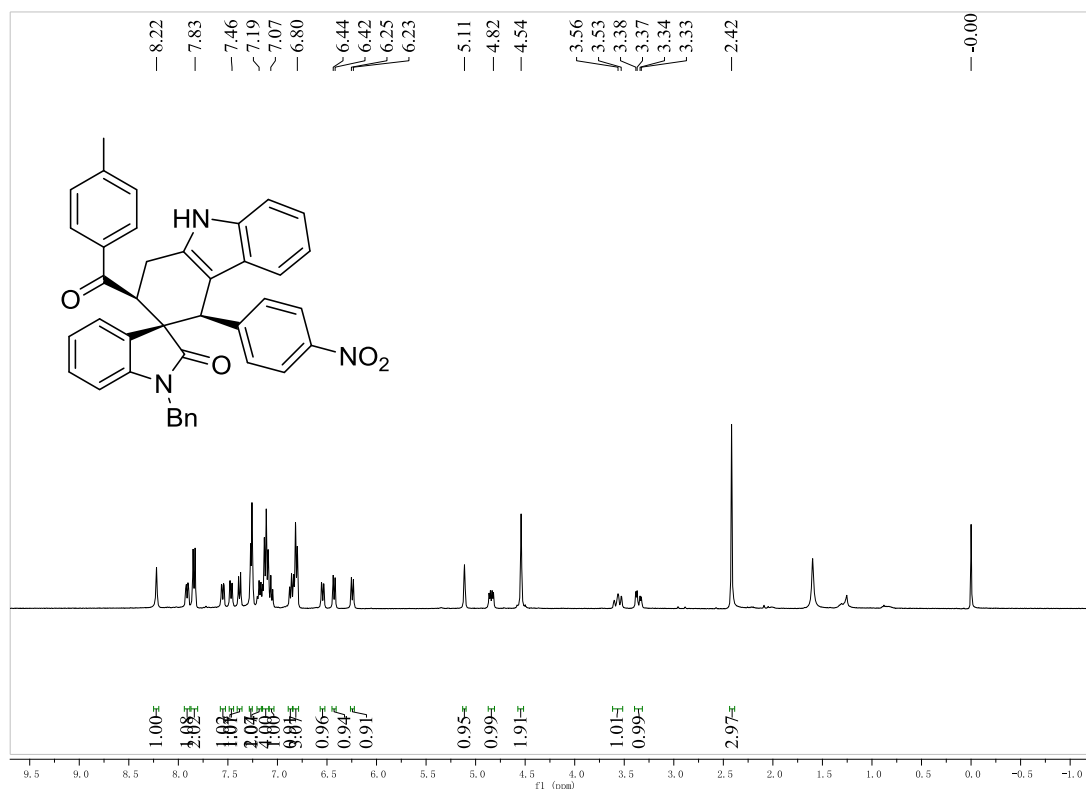
purple solid, 5%, m.p. 212-214 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.99 (s, 1H, NH), 7.73 (d, J = 8.0 Hz, 2H, ArH), 7.32-7.30 (m, 2H, ArH), 7.28-7.25 (m, 2H, ArH), 7.21 (t, J = 8.0 Hz, 3H, ArH), 7.12-7.06 (m, 5H, ArH), 6.93 (t, J = 8.0 Hz, 1H, ArH), 6.89-6.86 (m, 1H, ArH), 6.85-6.76 (m, 3H, ArH), 6.61 (dd, J_1 = 8.4 Hz, J_2 = 4.4 Hz, 1H, ArH), 4.94 (d, J = 15.6 Hz, 1H, CH), 4.67 (s, 1H, CH), 4.57 (d, J = 8.8 Hz, 1H, CH), 4.53 (d, J = 10.0 Hz, 1H, CH_2), 3.75 (dd, J_1 = 16.8 Hz, J_2 = 6.4 Hz, 1H, CH), 3.11 (dd, J_1 = 16.8 Hz, J_2 = 8.4 Hz, 1H, CH), 2.38 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 199.1, 176.1, 160.0, 157.6, 144.4, 141.5, 138.6, 138.6, 136.7, 135.6, 133.7, 133.5, 131.9, 129.9, 129.4, 129.0, 128.7, 128.6, 128.2, 127.5, 127.4, 127.3, 126.6, 121.8, 119.4, 118.9, 114.2 (d, J = 23.4 Hz), 112.8 (d, J = 25.9 Hz), 110.8, 109.6, 109.3 (d, J = 8.1 Hz), 54.3, 47.1, 44.0, 43.9, 24.7, 21.6; IR (KBr) ν : 3330, 3300, 3259, 3146, 3083, 2958, 2878, 2842, 2174, 1863, 1625, 1617, 1542, 1438, 1334, 1255, 1157, 1119, 983, 917, 862, 781 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{40}\text{H}_{30}\text{Cl}^{35}\text{FN}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 647.1872, found: 647.1861. $\text{C}_{40}\text{H}_{30}\text{Cl}^{37}\text{FN}_2\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 648.1906, found: 648.1890.

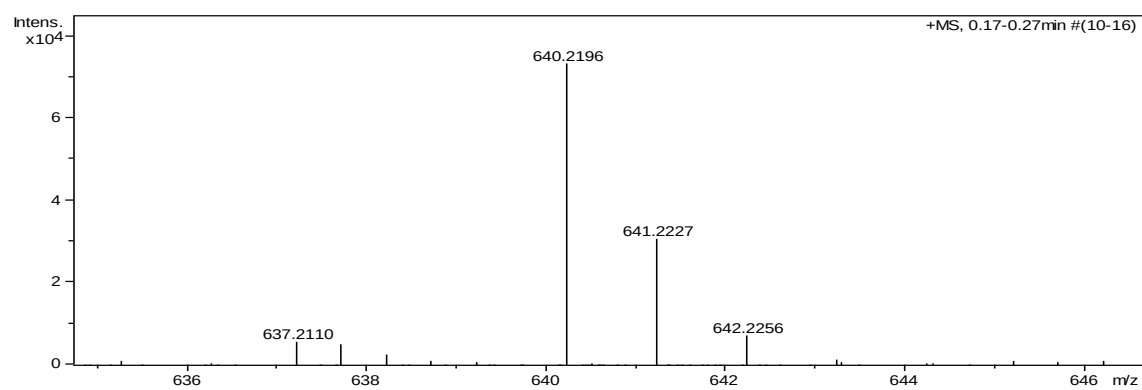
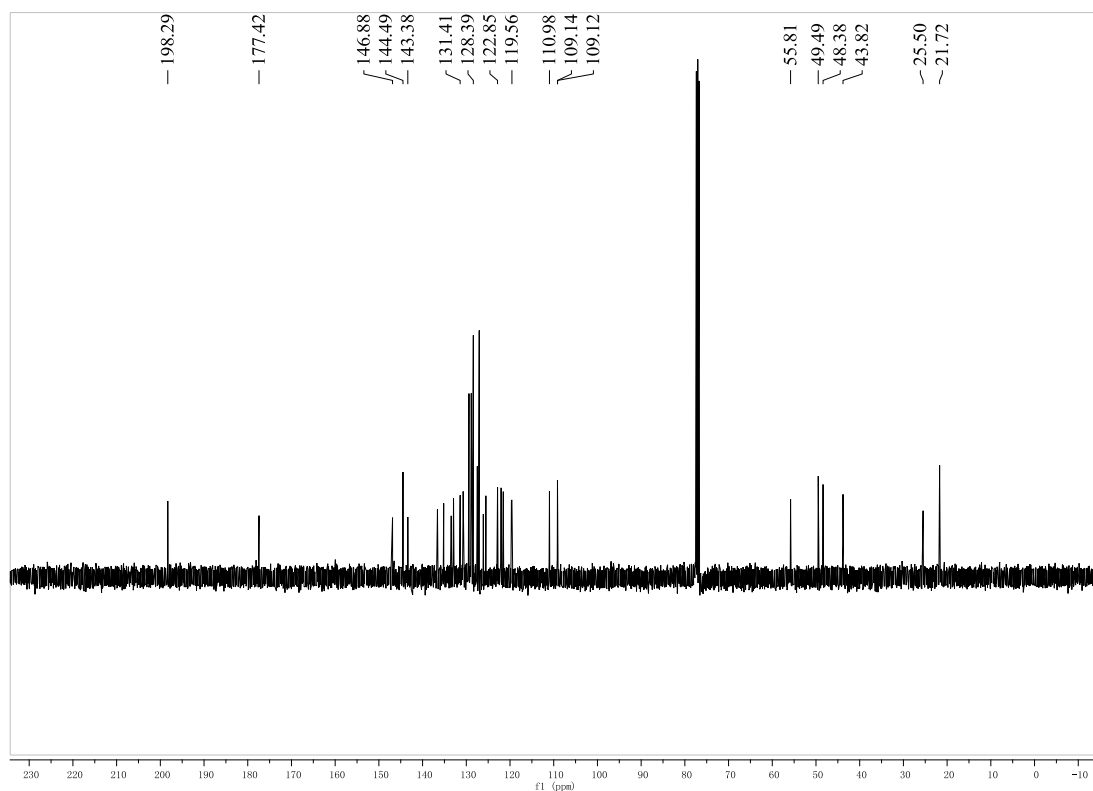




**1'-Benzyl-2-(4-methylbenzoyl)-4-(4-nitrophenyl)-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indol
in]-2'-one (1g):**

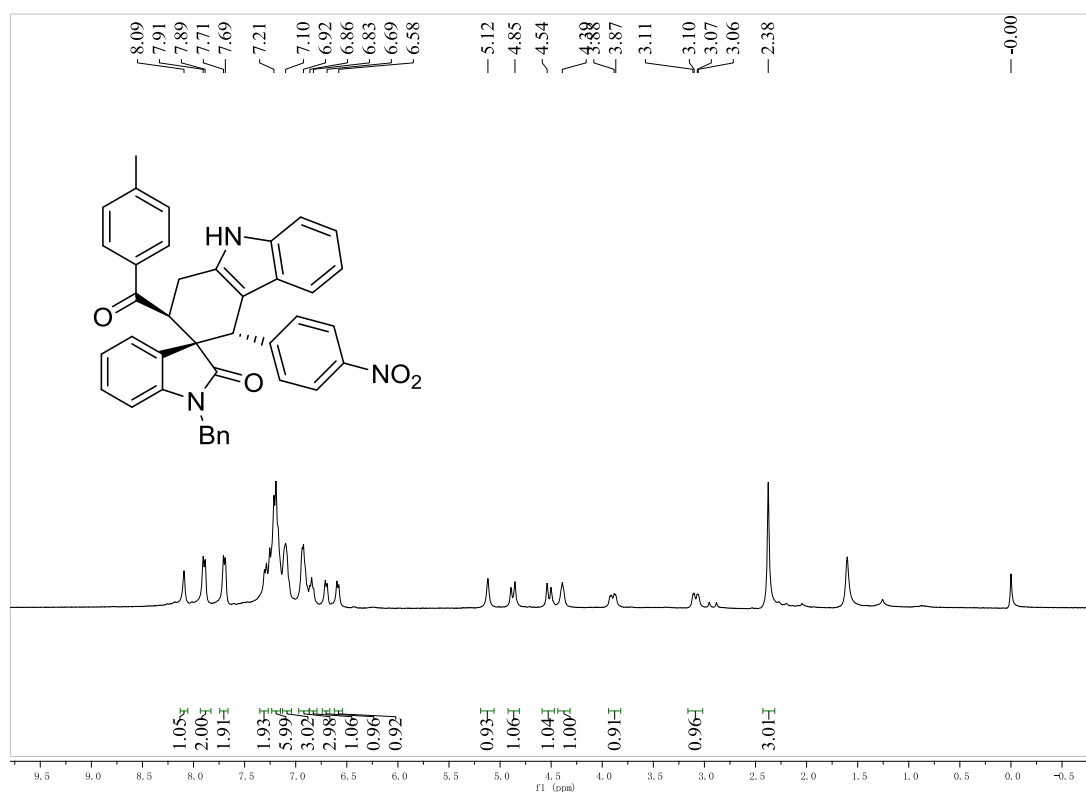
purple solid, 54%, m.p. 198-201 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.22 (s, 1H, NH), 7.90 (dd, $J_1 = 8.4$ Hz, $J_2 = 2.4$ Hz, 1H, ArH), 7.84 (d, $J = 8.0$ Hz, 2H, ArH), 7.55 (dd, $J_1 = 8.4$ Hz, $J_2 = 2.4$ Hz, 1H, ArH), 7.46 (d, $J = 8.0$ Hz, 1H, ArH), 7.38 (d, $J = 8.0$ Hz, 1H, ArH), 7.27-7.26 (m, 2H, ArH), 7.19 (t, $J = 7.2$ Hz, 1H, ArH), 7.13-7.09 (m, 4H, ArH), 7.05 (d, $J = 7.6$ Hz, 1H, ArH), 6.86 (d, $J = 7.6$ Hz, 1H, ArH), 6.82 (t, $J = 7.6$ Hz, 3H, ArH), 6.54 (d, $J = 8.8$ Hz, 1H, ArH), 6.42 (d, $J = 5.6$ Hz, 1H, ArH), 6.24 (d, $J = 8.0$ Hz, 1H, ArH), 5.11 (s, 1H, CH), 4.84 (dd, $J_1 = 12.4$ Hz, $J_2 = 5.6$ Hz, 1H, CH), 4.55-4.54 (m, 2H, CH_2), 3.60-3.53 (m, 1H, CH), 3.36 (dd, $J_1 = 16.8$ Hz, $J_2 = 5.6$ Hz, 1H, CH), 2.42 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 198.2, 177.4, 146.8, 144.4, 143.3, 136.5, 135.1, 133.4, 132.9, 131.4, 130.7, 129.3, 128.8, 128.4, 128.3, 127.4, 127.0, 126.1, 125.5, 122.8, 122.0, 121.9, 121.5, 119.6, 119.5, 110.9, 109.1, 109.1, 55.8, 49.4, 48.3, 43.8, 25.5, 21.7; IR(KBr) ν : 3300, 3270, 3217, 3148, 3043, 2967, 2831, 2800, 2169, 1855, 1637, 1600, 1549, 1434, 1338, 1267, 1138, 1171, 963, 955, 863, 754 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{40}\text{H}_{31}\text{N}_3\text{O}_4$ ($[\text{M}+\text{Na}]^+$): 640.2207, found: 640.2196.

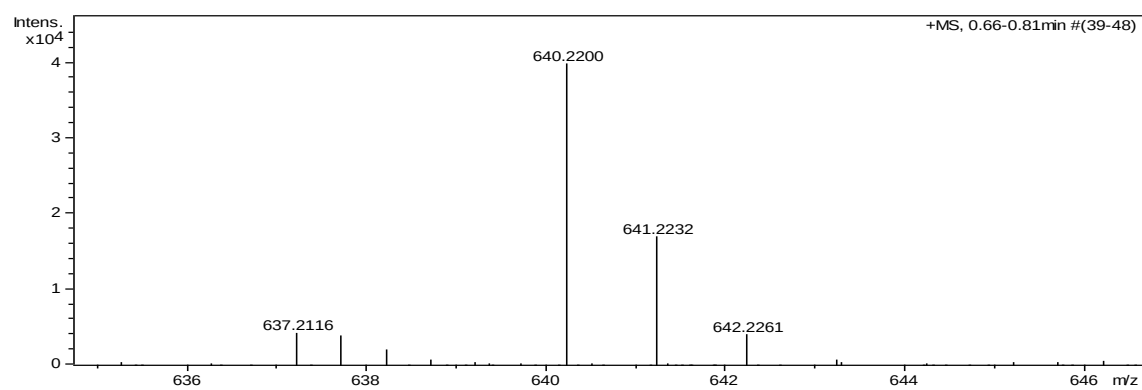
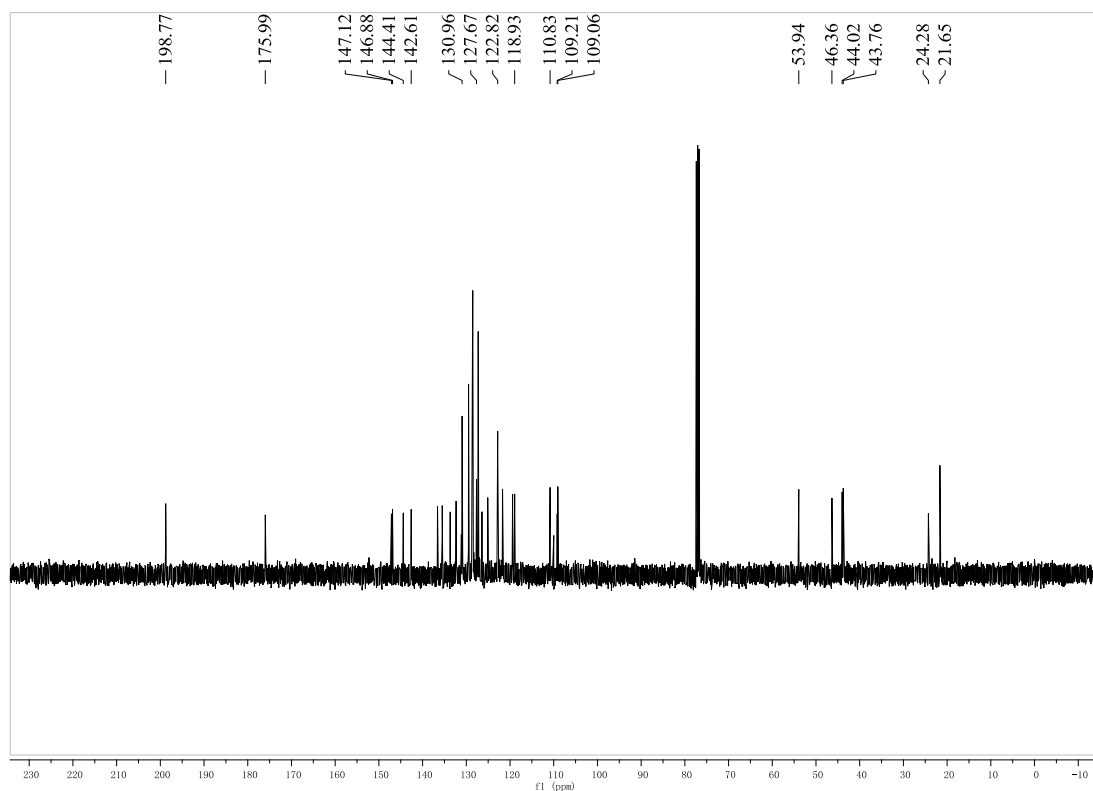




1'-Benzyl-2-(4-methylbenzoyl)-4-(4-nitrophenyl)-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1g'):

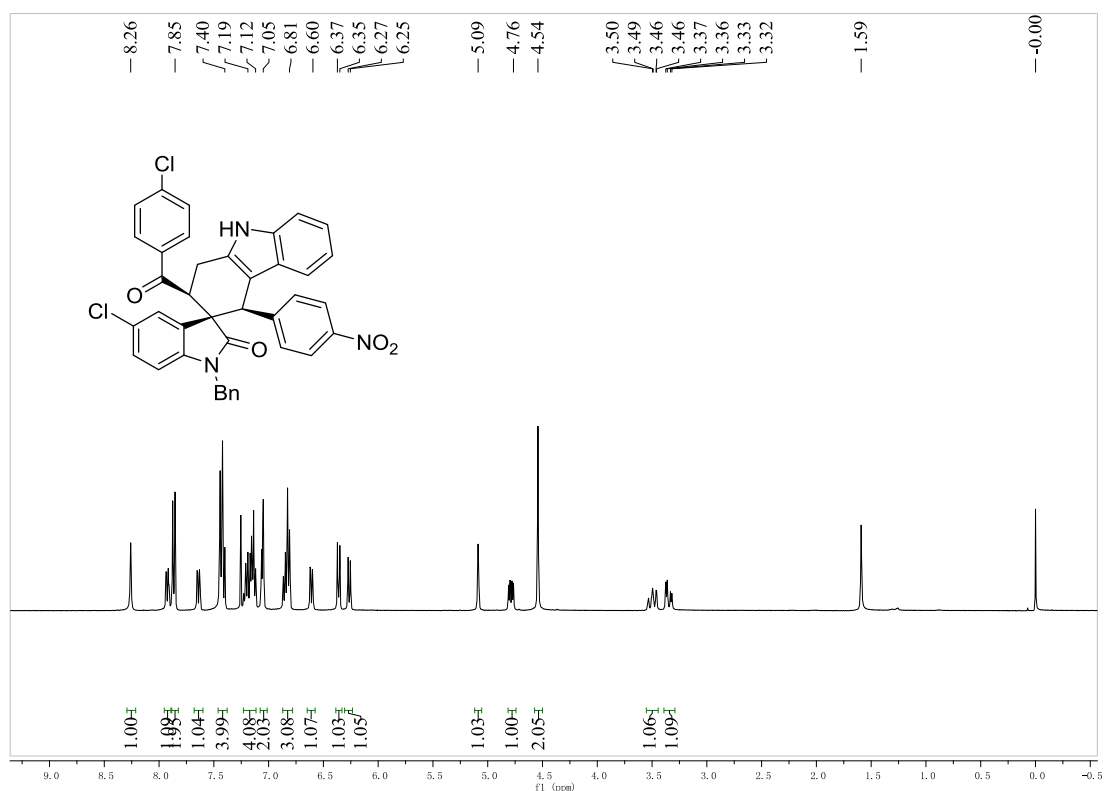
purple solid, 9%, m.p. 211-213 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.09 (s, 1H, NH), 7.89 (d, $J = 7.6$ Hz, 2H, ArH), 7.69 (d, $J = 7.2$ Hz, 2H, ArH), 7.29 (d, $J = 7.6$ Hz, 2H, ArH), 7.21-7.14 (m, 6H, ArH), 7.13-7.09 (m, 3H, ArH), 6.94-6.89 (m, 3H, ArH), 6.84 (t, $J = 7.2$ Hz, 1H, ArH), 6.70 (t, $J = 7.2$ Hz, 1H, ArH), 6.58 (d, $J = 7.2$ Hz, 1H, ArH), 5.12 (s, 1H, CH), 4.87 (d, $J = 15.6$ Hz, 1H, CH), 4.52 (d, $J = 15.6$ Hz, 1H, CH), 4.39 (s, 1H, CH), 3.92-3.86 (m, 1H, CH), 3.11-3.06 (m, 1H, CH), 2.38 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 198.7, 175.9, 147.1, 146.8, 144.4, 142.6, 136.5, 135.5, 133.6, 132.3, 130.9, 129.4, 128.6, 128.5, 127.6, 127.2, 126.4, 125.0, 122.8, 122.7, 121.7, 119.4, 118.9, 110.8, 109.2, 109.0, 53.9, 46.3, 44.0, 43.7, 24.2, 21.6; IR (KBr) ν : 3331, 3300, 3267, 3175, 3067, 2938, 2866, 2817, 2167, 1855, 1632, 1618, 1544, 1467, 1378, 1263, 1154, 1117, 963, 944, 869, 771 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{40}\text{H}_{31}\text{N}_3\text{O}_4$ ($[\text{M}+\text{Na}]^+$): 640.2207, found: 640.2200.

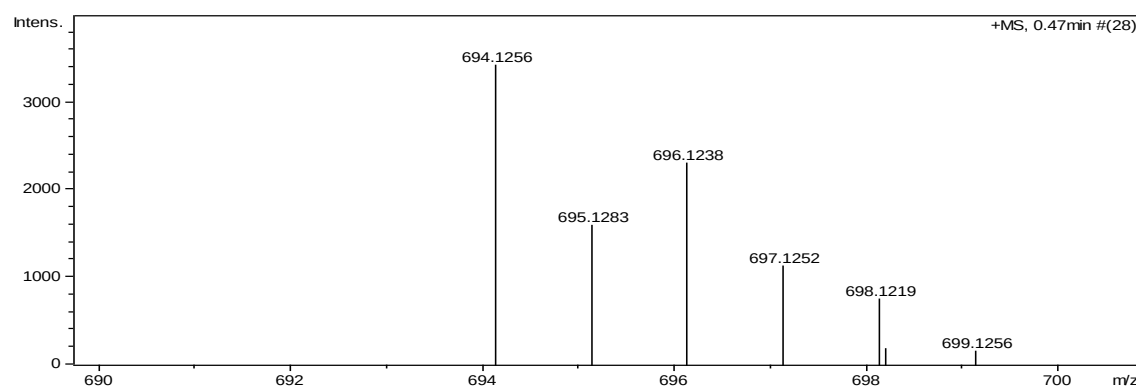
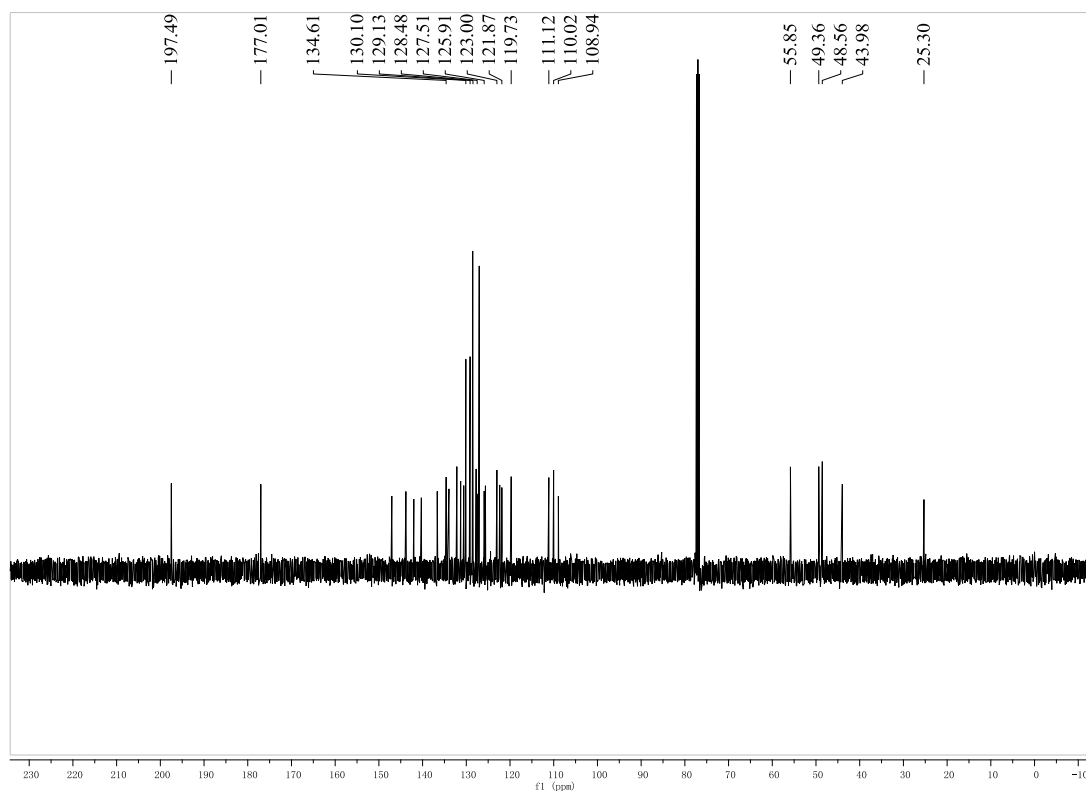




1'-Benzyl-5'-chloro-2-(4-chlorobenzoyl)-4-(4-nitrophenyl)-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1h):

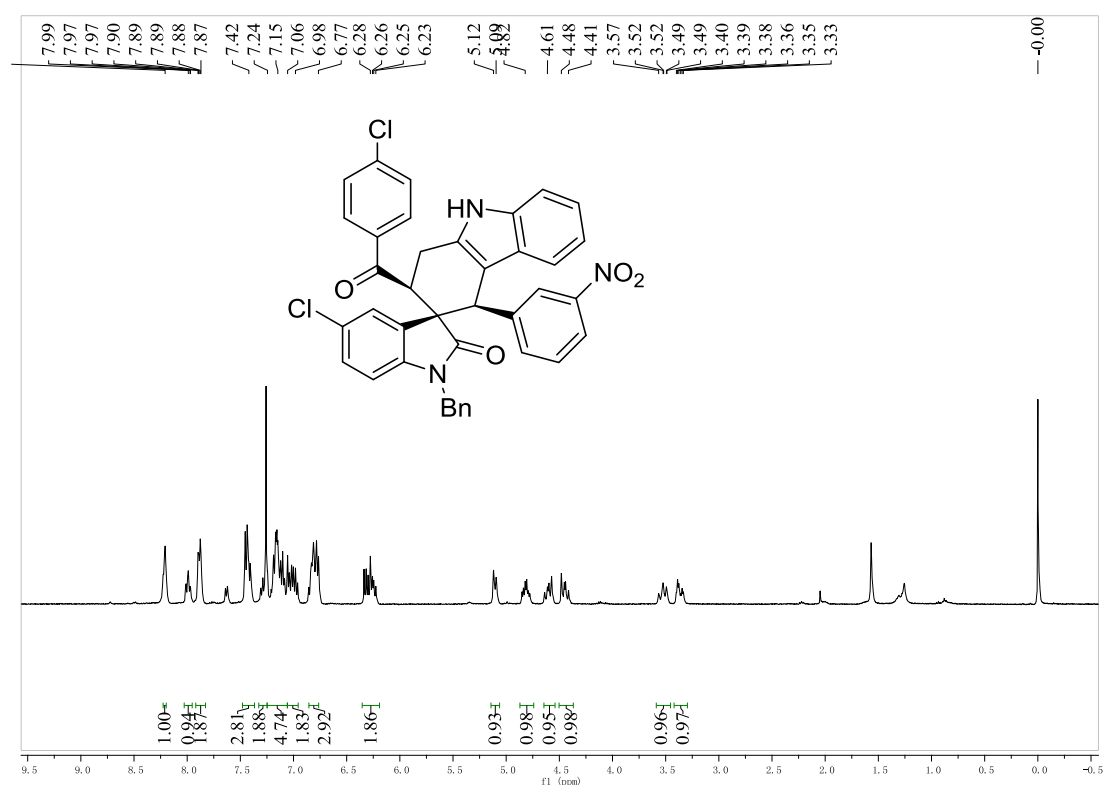
purple solid, 57%, m.p. 196-199 °C; ¹H NMR (400 MHz, CDCl₃) δ: 8.26 (s, 1H, NH), 7.92 (dd, *J*₁ = 8.4 Hz, *J*₂ = 2.4 Hz, 1H, ArH), 7.86 (d, *J* = 8.8 Hz, 2H, ArH), 7.64 (dd, *J*₁ = 8.4 Hz, *J*₂ = 2.4 Hz, 1H, ArH), 7.44-7.40 (m, 4H, ArH), 7.23-7.12 (m, 4H, ArH), 7.07-7.05 (m, 2H, ArH), 6.87-6.81 (m, 3H, ArH), 6.61 (dd, *J*₁ = 8.8 Hz, *J*₂ = 1.6 Hz, 1H, ArH), 6.36 (d, *J* = 8.8 Hz, 1H, ArH), 6.26 (d, *J* = 8.0 Hz, 1H, ArH), 5.09 (s, 1H, CH), 4.78 (dd, *J*₁ = 12.4 Hz, *J*₂ = 5.6 Hz, 1H, CH), 4.54-4.53 (m, 2H, CH₂), 3.54-3.46 (m, 1H, CH), 3.34 (dd, *J*₁ = 16.8 Hz, *J*₂ = 5.6 Hz, 1H, CH); ¹³C NMR (400 MHz, CDCl₃) δ: 197.4, 177.0, 147.0, 143.8, 142.0, 140.2, 136.6, 134.6, 133.9, 132.1, 131.2, 130.5, 130.1, 129.1, 129.1, 128.5, 128.4, 127.7, 127.5, 127.0, 125.9, 125.6, 123.0, 122.3, 121.8, 119.8, 119.7, 111.1, 110.0, 108.9, 55.8, 49.3, 48.5, 43.9, 25.3; IR (KBr) ν: 3354, 3310, 3267, 3149, 3066, 2955, 2871, 2846, 2176, 1848, 1655, 1600, 1531, 1414, 1317, 1248, 1167, 1130, 992, 917, 867, 768 cm⁻¹; MS (*m/z*): HRMS (ESI) Calcd. for C₃₉H₂₇Cl₂³⁵N₃O₄ ([M+Na]⁺): 694.1271, found: 694.1256. C₃₉H₂₇Cl₂³⁷N₃O₄ ([M+Na]⁺): 696.1241, found: 696.1238.

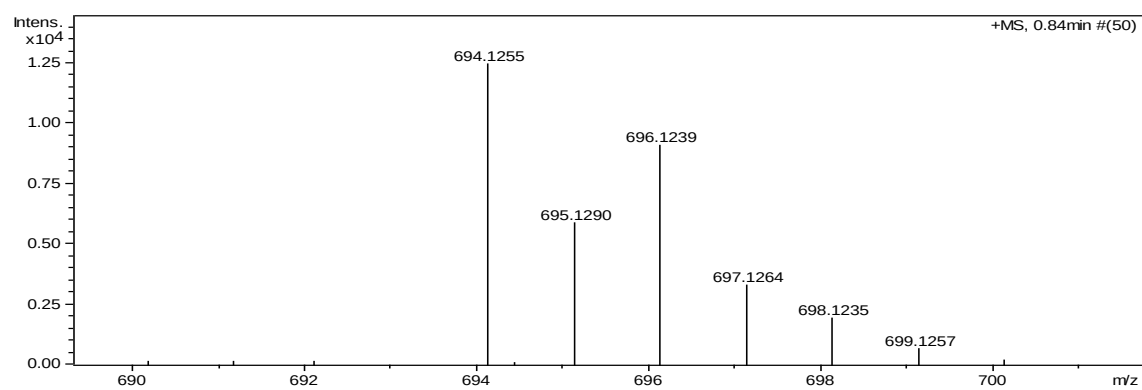
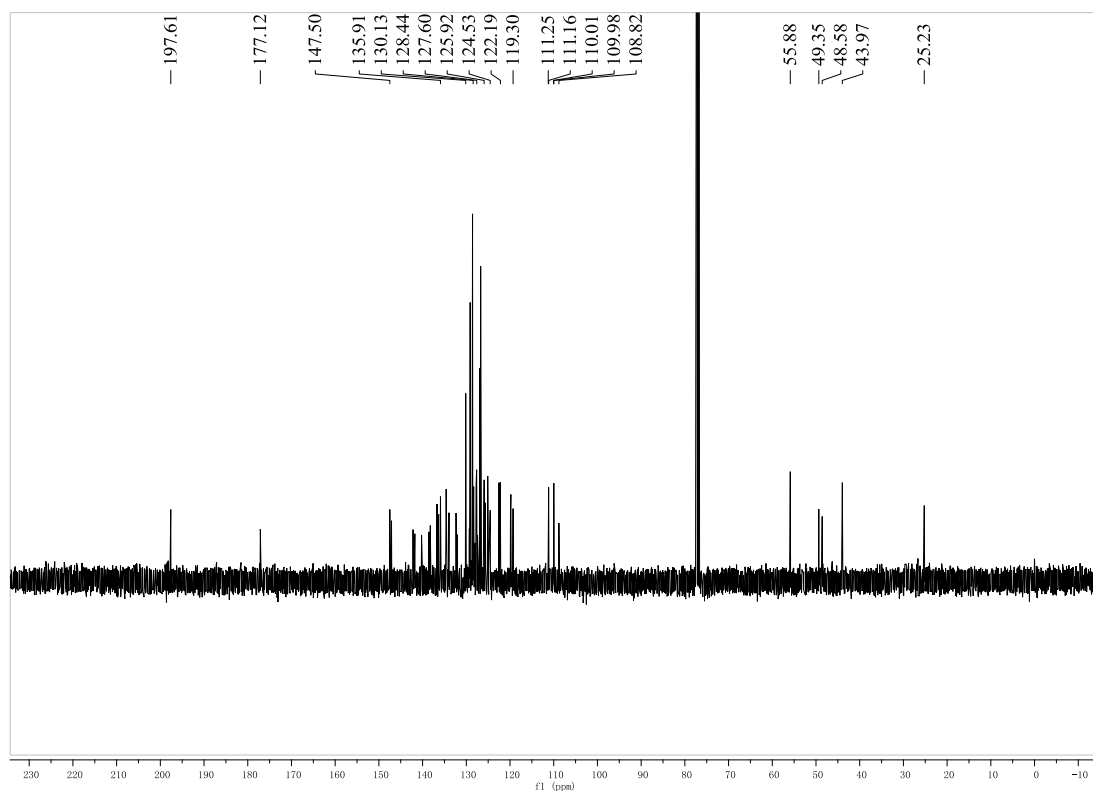




1'-Benzyl-5'-chloro-2-(4-chlorobenzoyl)-4-(3-nitrophenyl)-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1i):

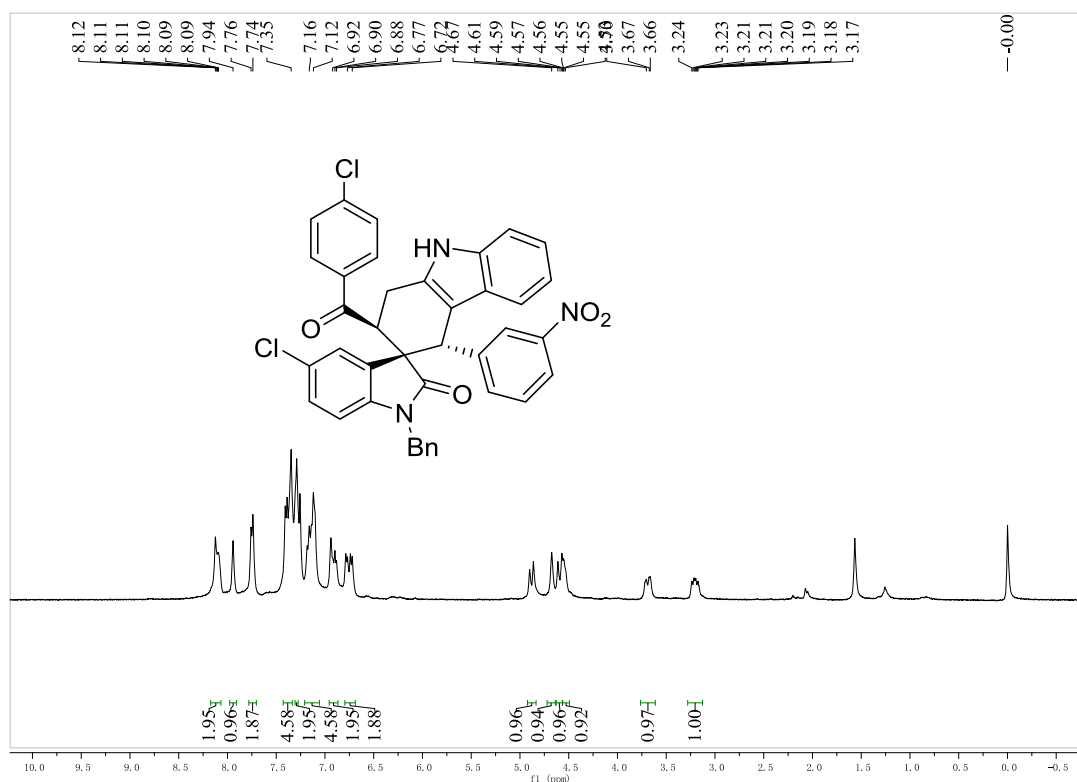
purple solid, 49%, m.p. 201-204 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.21 (s, 1H, NH), 7.99 (t, J = 8.0 Hz, 1H, ArH), 7.90-7.87 (m, 2H, ArH), 7.46-7.40 (m, 3H, ArH), 7.31-7.26 (m, 2H, ArH), 7.24-7.06 (m, 5H, ArH), 7.05-6.96 (m, 2H, ArH), 6.86-6.77 (m, 3H, ArH), 6.34-6.23 (m, 2H, ArH), 5.10 (d, J = 9.6 Hz, 1H, CH), 4.85-4.78 (m, 1H, CH), 4.64-4.57 (m, 1H, CH), 4.48-4.41 (m, 1H, CH), 3.57-3.49 (m, 1H, CH), 3.40-3.33 (m, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 197.6, 177.1, 147.5, 147.1, 142.1, 141.7, 140.2, 138.5, 136.7, 136.3, 135.9, 134.6, 133.9, 132.3, 130.1, 129.1, 128.6, 128.5, 128.4, 128.3, 127.7, 127.6, 127.6, 127.4, 126.9, 126.7, 125.9, 125.7, 125.6, 125.0, 124.5, 122.5, 122.3, 122.2, 122.1, 119.8, 119.8, 119.6, 119.2, 111.2, 111.1, 110.0, 109.9, 108.8, 55.8, 49.3, 48.5, 43.9, 25.2; IR (KBr) ν : 3355, 3238, 3146, 3067, 2955, 2848, 2168, 1848, 1637, 1617, 1555, 1447, 1362, 1276, 1155, 1168, 966, 903, 855, 769 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{39}\text{H}_{27}\text{Cl}_2^{35}\text{N}_3\text{O}_4$ ($[\text{M}+\text{Na}]^+$): 694.1271, found: 694.1255. $\text{C}_{39}\text{H}_{27}\text{Cl}_2^{37}\text{N}_3\text{O}_4$ ($[\text{M}+\text{Na}]^+$): 696.1241, found: 696.1239.

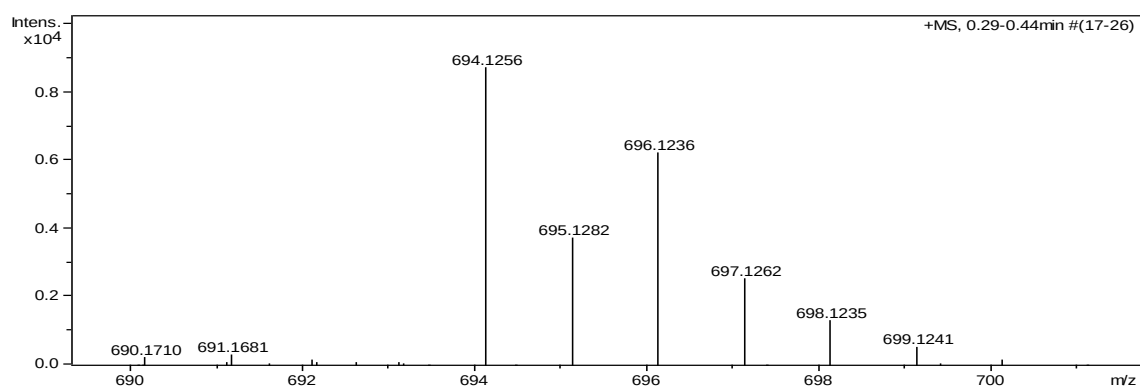
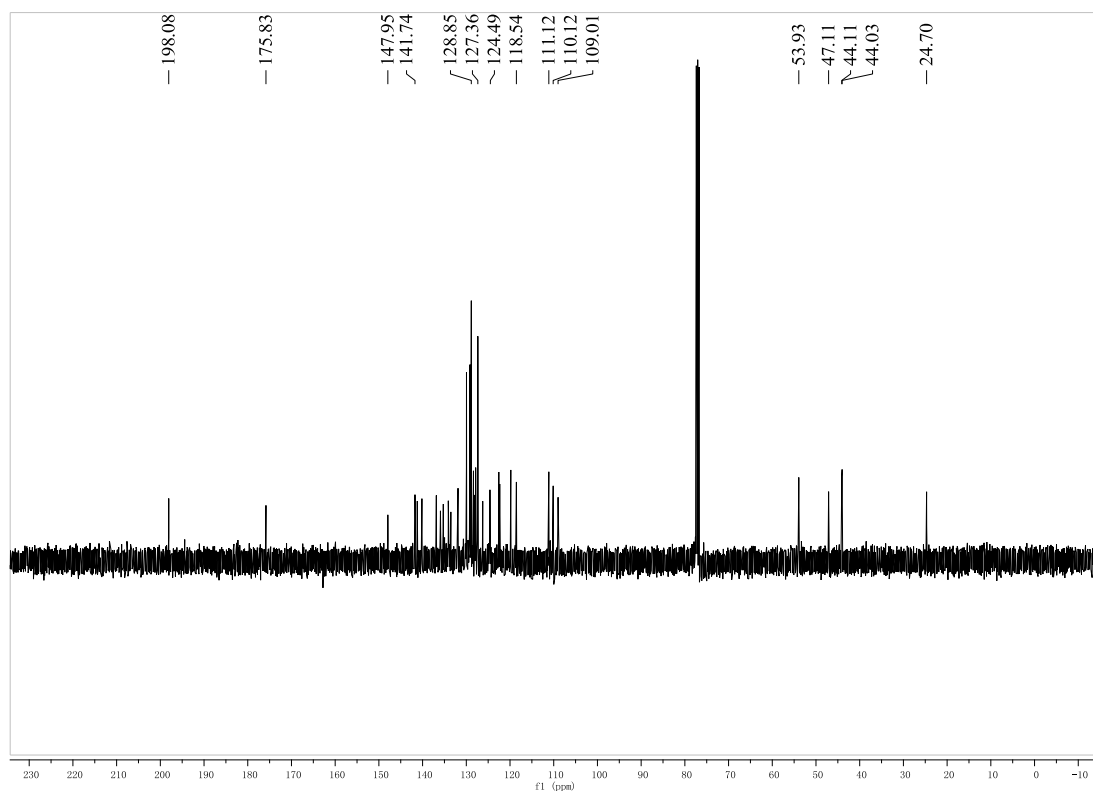




1'-Benzyl-5'-chloro-2-(4-chlorobenzoyl)-4-(3-nitrophenyl)-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1i'):

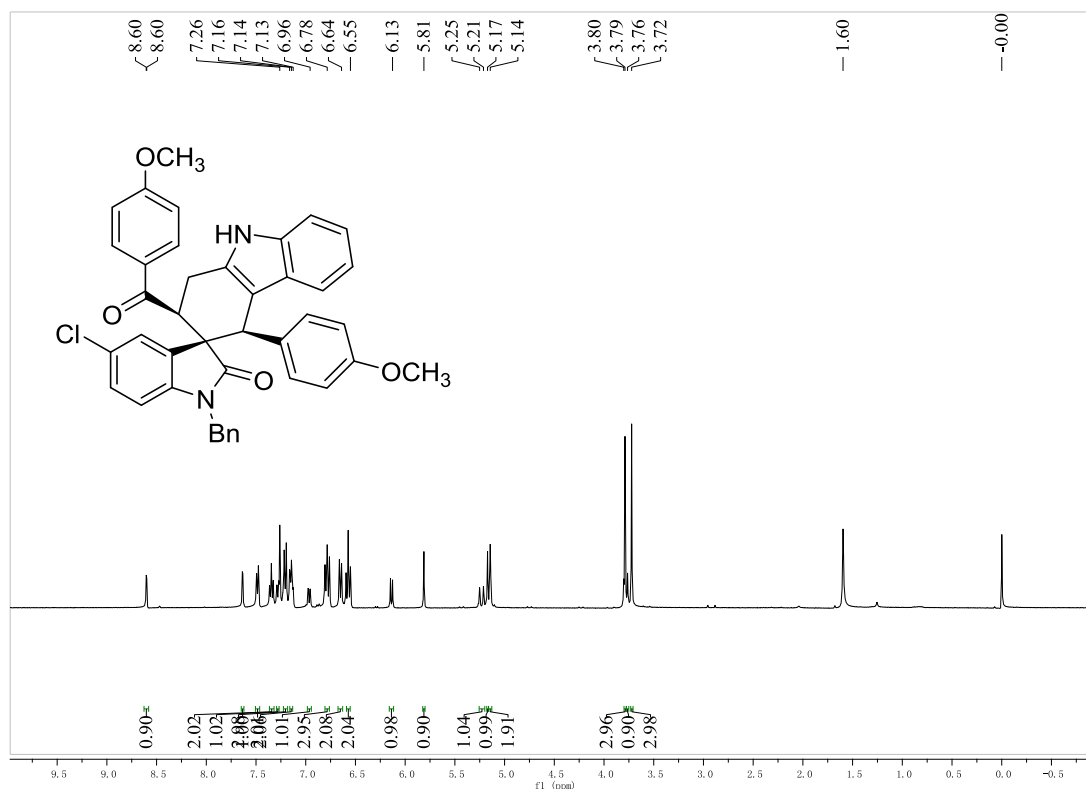
purple solid, 6%, m.p. 216-219 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.12-8.09 (m, 2H, NH), 7.94 (s, 1H, NH), 7.75 (d, $J = 7.6$ Hz, 2H, ArH), 7.41-7.35 (m, 5H, ArH), 7.29-7.26 (m, 2H, ArH), 7.18-7.12 (m, 5H, ArH), 6.94-6.88 (m, 2H, ArH), 6.75 (dd, $J_1 = 19.2$ Hz, $J_2 = 7.6$ Hz, 2H, ArH), 4.88 (d, $J = 15.2$ Hz, 1H, CH), 4.68 (s, 1H, CH), 4.59 (d, $J = 15.2$ Hz, 1H, CH), 4.56-4.53 (m, 1H, CH), 3.72-3.66 (m, 1H, CH), 3.24-3.17 (m, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 198.0, 175.8, 147.9, 141.7, 141.2, 140.1, 136.8, 135.9, 135.2, 134.1, 133.5, 131.8, 129.9, 129.1, 128.8, 128.3, 128.0, 127.8, 127.3, 126.2, 124.5, 124.5, 124.4, 122.5, 122.3, 119.8, 118.5, 111.1, 110.1, 109.0, 53.9, 47.1, 44.1, 44.0, 24.6 cm^{-1} ; IR (KBr) ν : 3341, 3278, 3166, 3048, 2966, 2849, 2158, 1871, 1671, 1643, 1522, 1458, 1331, 1268, 1148, 1137, 954, 913, 832, 767 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{39}\text{H}_{27}\text{Cl}_2^{35}\text{N}_3\text{O}_4$ ($[\text{M}+\text{Na}]^+$): 694.1271, found: 694.1256. $\text{C}_{39}\text{H}_{27}\text{Cl}_2^{37}\text{N}_3\text{O}_4$ ($[\text{M}+\text{Na}]^+$): 696.1241, found: 696.1236.

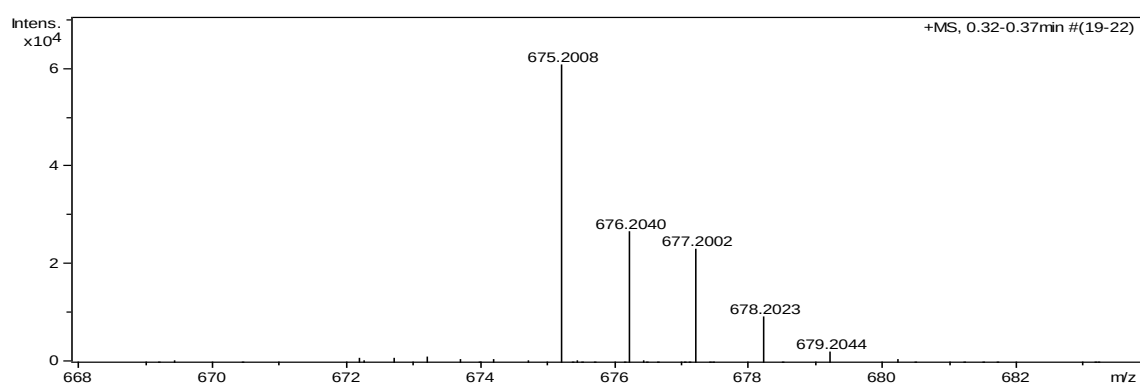
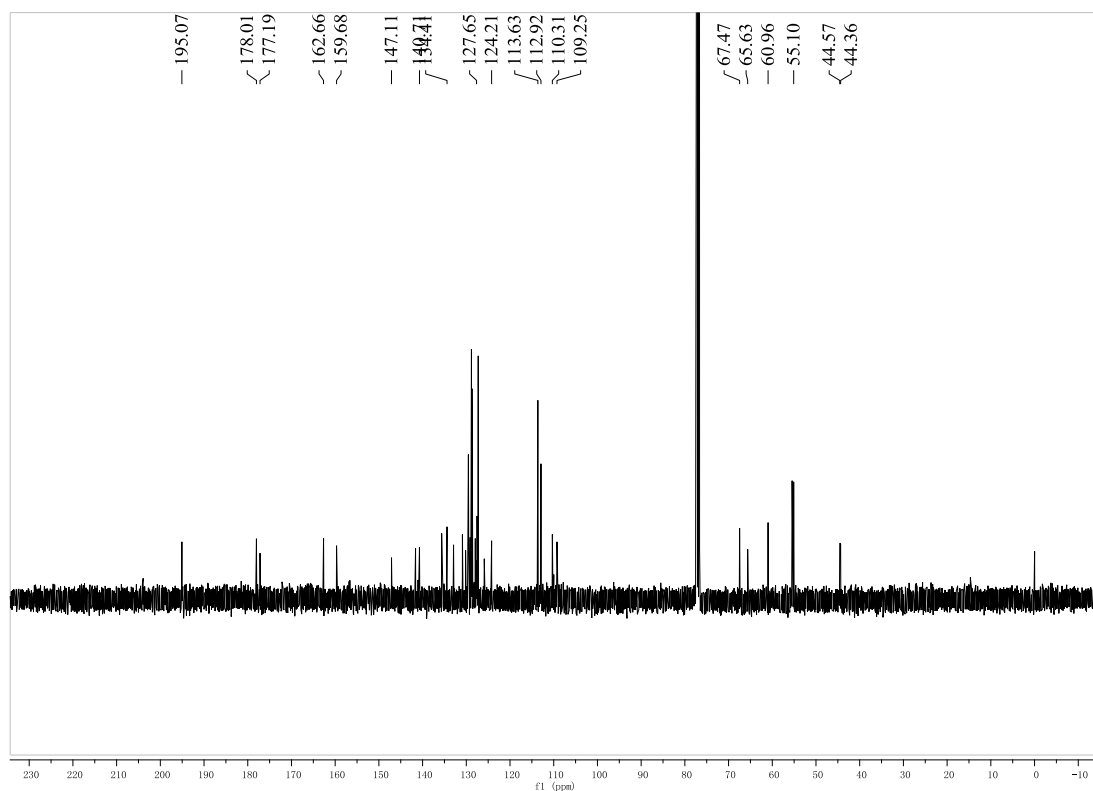




1'-Benzyl-5'-chloro-2-(4-methoxybenzoyl)-4-(4-methoxyphenyl)-1,2,4,9-tetrahydrospiro[carbazole-3,3'-indolin]-2'-one (1j):

purple solid, 65%, m.p. 189-192 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.60 (d, $J = 2.0$ Hz, 1H, NH), 7.63 (d, $J = 2.0$ Hz, 1H, ArH), 7.48 (d, $J = 7.6$ Hz, 2H, ArH), 7.35 (t, $J = 7.6$ Hz, 2H, ArH), 7.28 (d, $J = 7.6$ Hz, 1H, ArH), 7.20 (d, $J = 8.4$ Hz, 3H, ArH), 7.16-7.13 (m, 2H, ArH), 6.96 (dd, $J_1 = 8.4$ Hz, $J_2 = 2.0$ Hz, 1H, ArH), 6.78 (d, $J = 8.8$ Hz, 3H, ArH), 6.65 (d, $J = 8.8$ Hz, 2H, ArH), 6.57 (t, $J = 8.8$ Hz, 2H, ArH), 6.13 (d, $J = 8.4$ Hz, 1H, ArH), 5.81 (s, 1H, CH), 5.23 (d, $J = 15.6$ Hz, 1H, CH), 5.17-5.16 (m, 1H, CH), 5.16-5.14 (m, 2H, CH_2), 3.79 (d, $J = 15.6$ Hz, 1H, CH), 3.78 (s, 3H, OCH_3), 3.72 (s, 3H, OCH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 195.0, 177.6 (d, $J = 8.0$ Hz), 162.6, 159.6, 147.1, 141.6, 140.7, 135.6, 134.4, 132.9, 130.8, 130.1, 129.5, 129.4, 129.1, 128.8, 128.8, 128.7, 128.5, 127.9, 127.6, 127.5, 127.5, 127.2, 127.2, 125.8, 124.2, 113.6, 112.9, 110.3, 109.2, 67.4, 65.6, 60.9, 55.4, 55.1, 44.5, 44.3; IR (KBr) ν : 3300, 3284, 3180, 3078, 2967, 2849, 2831, 2156, 1833, 1619, 1600, 1531, 1429, 1333, 1219, 1117, 1100, 955, 948, 860, 769 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{41}\text{H}_{33}\text{Cl}^{35}\text{N}_2\text{O}_4$ ($[\text{M}+\text{Na}]^+$): 675.2021, found: 675.2008. $\text{C}_{41}\text{H}_{33}\text{Cl}^{37}\text{N}_2\text{O}_4$ ($[\text{M}+\text{Na}]^+$): 676.2055, found: 676.2040.

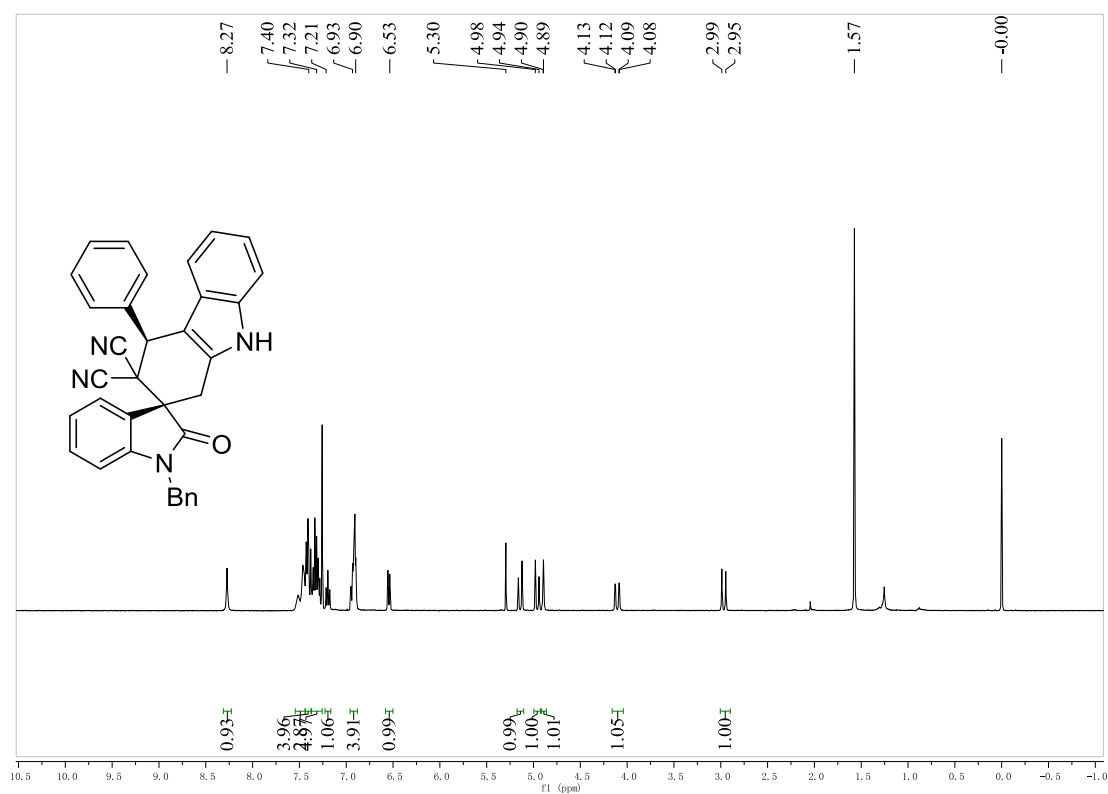


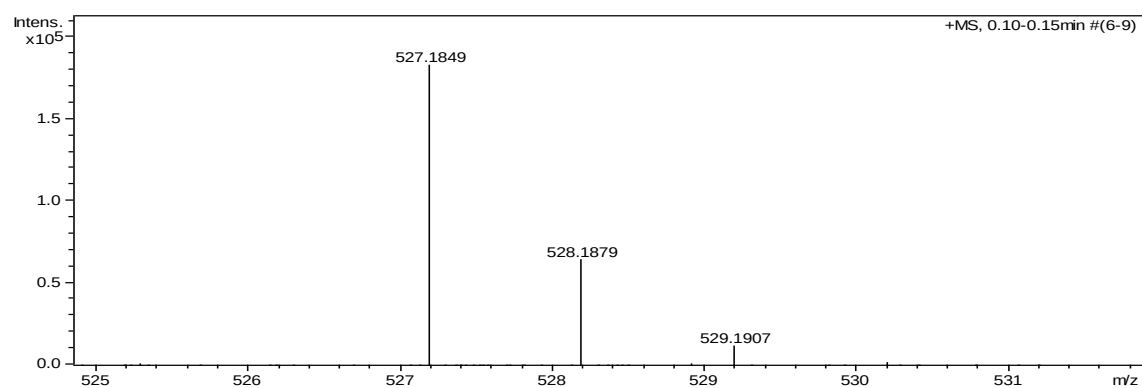
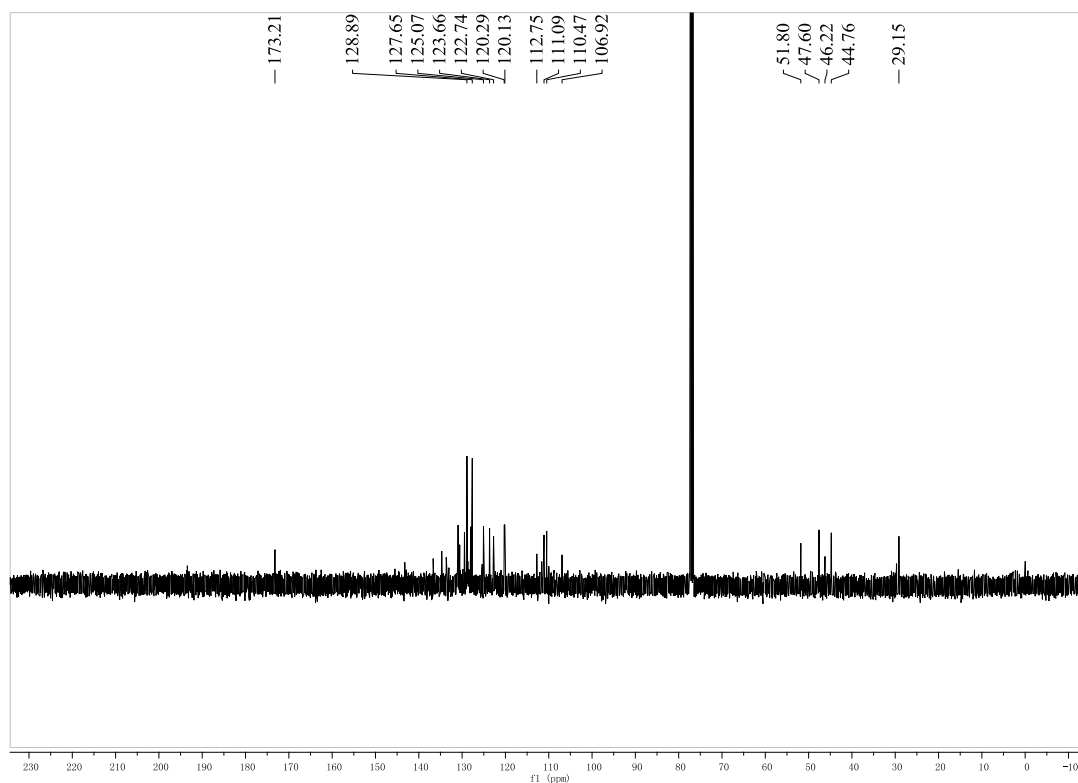


1'-Benzyl-2'-oxo-4-phenyl-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1*H*)-dicarbonitrile

(2a):

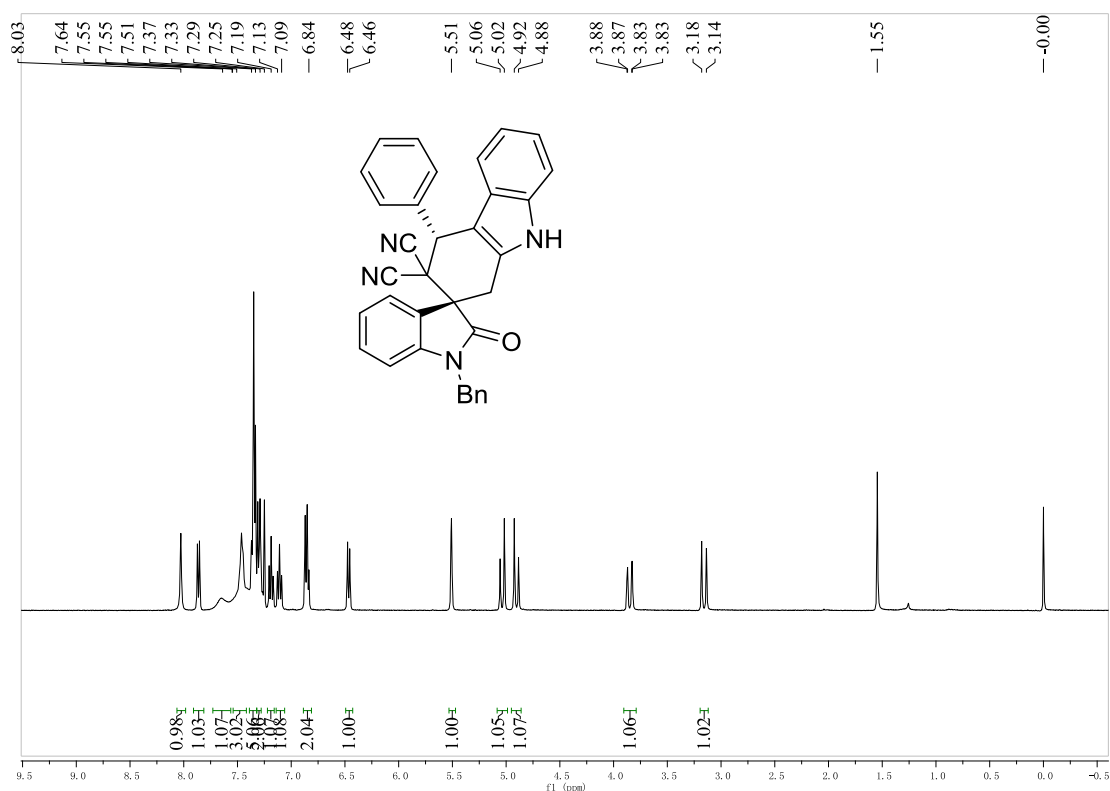
white solid, 51%, m.p. 201-204 °C; ¹H NMR (400 MHz, CDCl₃) δ: 8.27 (s, 1H, NH), 7.52-7.46 (m, 3H, ArH), 7.43-7.38 (m, 4H, ArH), 7.35-7.28 (m, 5H, ArH), 7.20 (t, *J* = 7.6 Hz, 1H, CH), 6.95-6.90 (m, 4H, ArH), 6.54 (d, *J* = 8.0 Hz, 1H, CH), 5.14 (d, *J* = 15.6 Hz, 1H, CH), 4.96 (d, *J* = 15.6 Hz, 1H, CH), 4.89 (s, 1H, CH), 4.10 (dd, *J*₁ = 16.4 Hz, *J*₂ = 2.4 Hz, 1H, CH), 2.96 (d, *J* = 16.4 Hz, 1H, CH); ¹³C NMR (400 MHz, CDCl₃) δ: 173.2, 136.6, 134.7, 133.6, 130.9, 129.4, 128.8, 128.0, 127.6, 125.0, 123.6, 122.7, 120.2, 120.1, 112.7, 111.0, 110.4, 106.9, 51.8, 47.6, 46.2, 44.7, 29.1; IR (KBr) ν: 3355, 3207, 3117, 3048, 2963, 2831, 2167, 1871, 1641, 1633, 1554, 1431, 1370, 1240, 1131, 1100, 972, 961, 881, 764 cm⁻¹; MS (*m/z*): HRMS (ESI) Calcd. for C₃₄H₂₄N₄O ([M+Na]⁺): 527.1842, found: 527.1849.

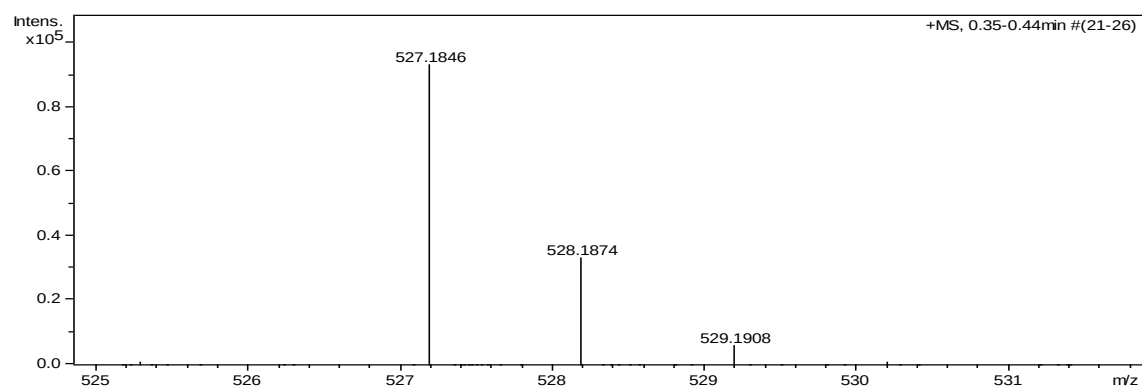
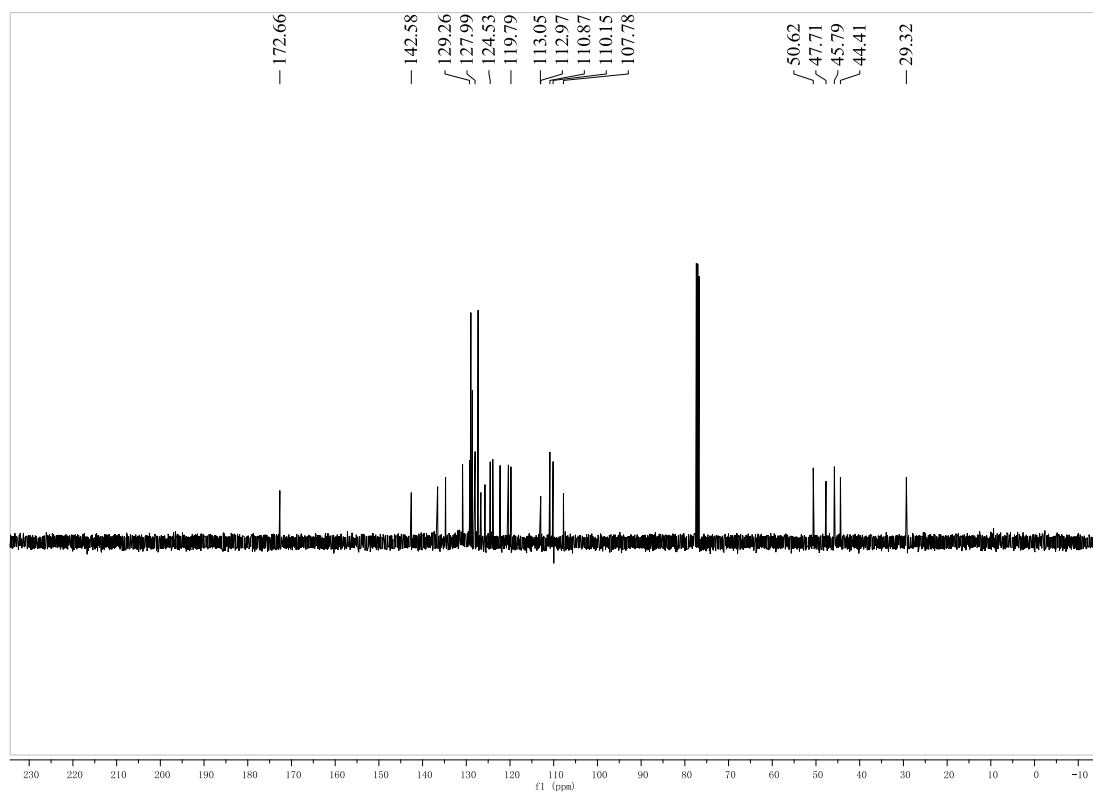




1'-Benzyl-2'-oxo-4-phenyl-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1*H*)-dicarbonitrile (2a')

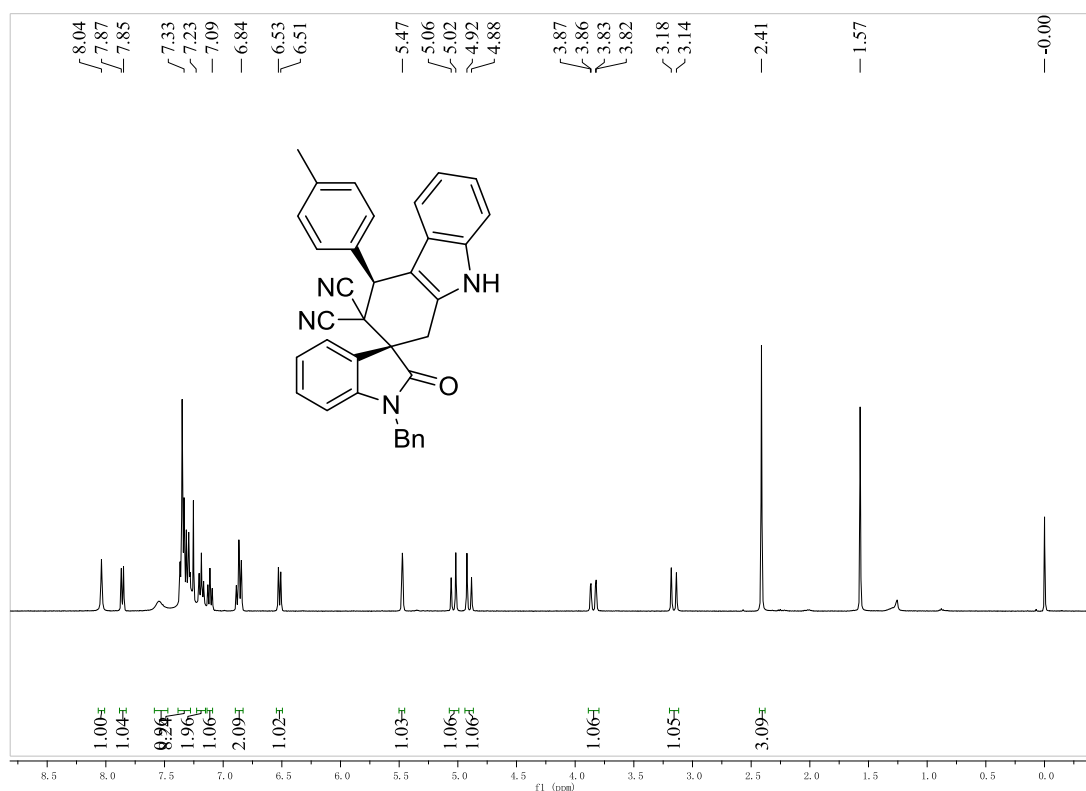
white solid, 7%, m.p. 213-215 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.03 (s, 1H, NH), 7.86 (d, J = 7.6 Hz, 1H, ArH), 7.66-7.63 (m, 1H, ArH), 7.55-7.46 (m, 3H, ArH), 7.37-7.33 (m, 5H, ArH), 7.31-7.28 (m, 2H, ArH), 7.19 (t, J = 7.6 Hz, 1H, ArH), 7.11 (t, J = 7.6 Hz, 1H, ArH), 6.86 (d, J = 7.6 Hz, 2H, ArH), 6.46 (d, J = 8.0 Hz, 1H, ArH), 5.51 (s, 1H, CH), 5.03 (d, J = 15.6 Hz, 1H, CH), 4.90 (d, J = 15.6 Hz, 1H, CH), 3.85 (dd, J_1 = 17.6 Hz, J_2 = 2.0 Hz, 1H, CH), 3.15 (d, J = 17.6 Hz, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 172.6, 142.5, 136.5, 134.7, 134.6, 130.8, 129.2, 129.1, 128.9, 128.6, 127.9, 127.3, 126.6, 125.7, 124.5, 123.9, 122.2, 120.3, 119.7, 113.0, 112.9, 110.8, 110.1, 107.7, 50.6, 47.7, 45.7, 44.4, 29.3; IR (KBr) ν : 3300, 3267, 3178, 3043, 2965, 2848, 2164, 1853, 1666, 1607, 1541, 1456, 1355, 1251, 1167, 1121, 969, 955, 870, 768 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{34}\text{H}_{24}\text{N}_4\text{O}$ ($[\text{M}+\text{Na}]^+$): 527.1842, found: 527.1846.

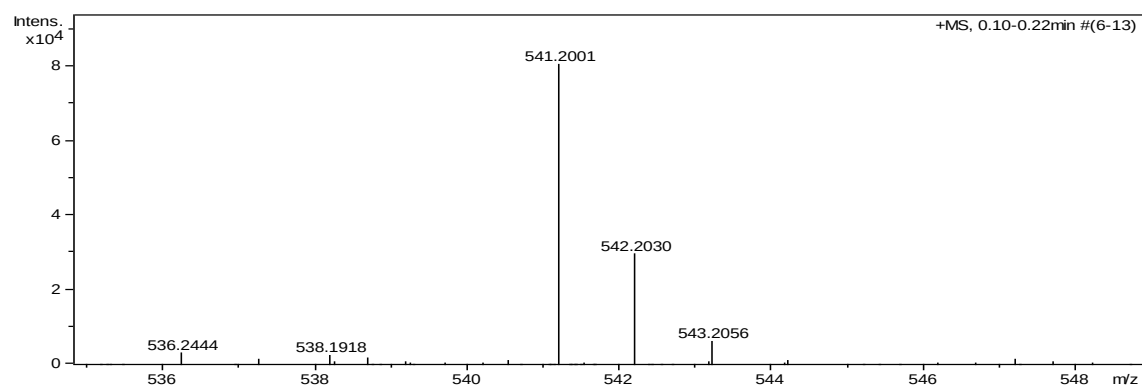
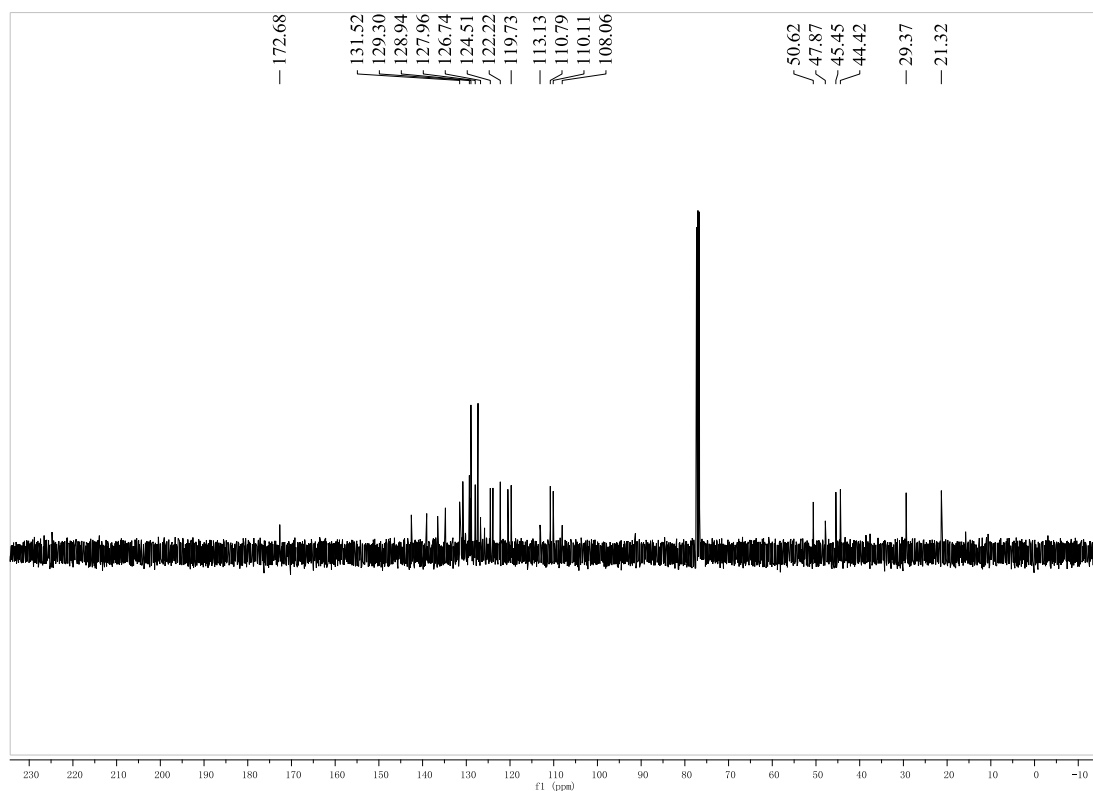




**1'-Benzyl-2'-oxo-4-(*p*-tolyl)-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1*H*)-dicarbonitrile
(2b):**

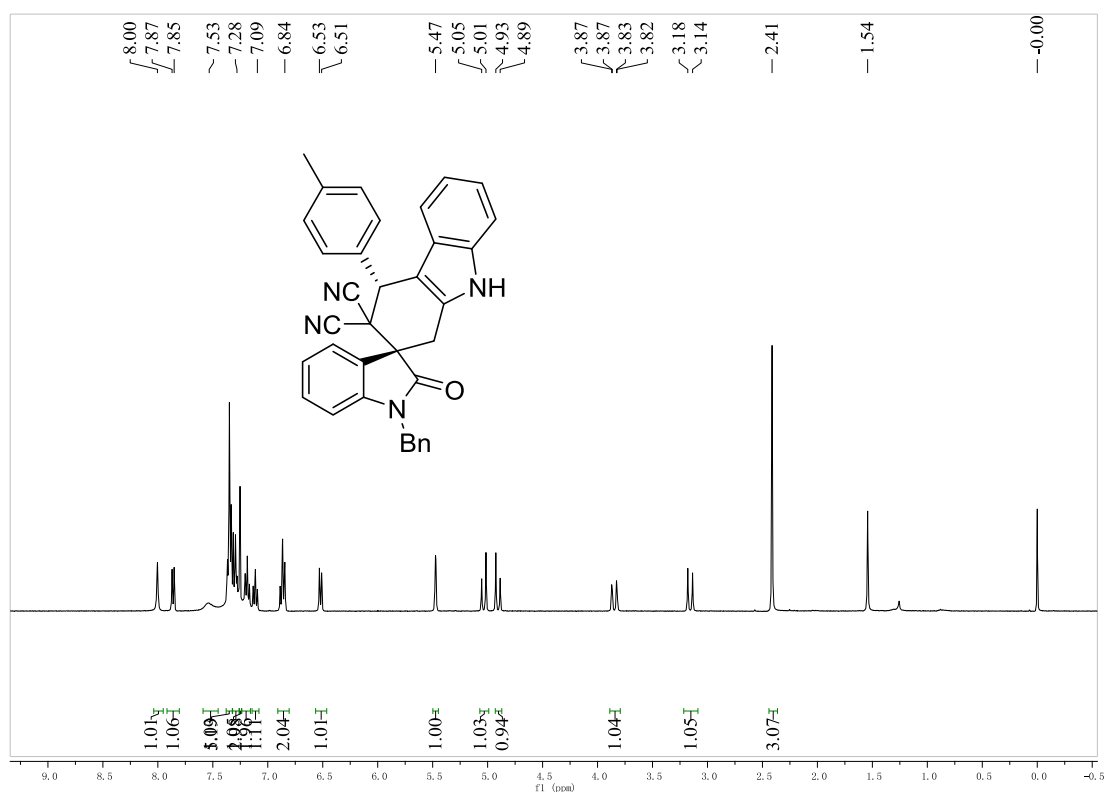
white solid, 62%, m.p. 195-198 °C; ¹H NMR (400 MHz, CDCl₃) δ: 8.04 (s, 1H, NH), 7.85 (d, *J* = 7.6 Hz, 1H, ArH), 7.55-7.52 (m, 1H, ArH), 7.37-7.27 (m, 8H, ArH), 7.19 (t, *J* = 7.6 Hz, 2H, ArH), 7.11 (t, *J* = 7.2 Hz, 1H, ArH), 6.87 (t, *J* = 8.4 Hz, 2H, ArH), 6.51 (d, *J* = 8.0 Hz, 1H, ArH), 5.47 (s, 1H, CH), 5.03 (d, *J* = 15.6 Hz, 1H, CH), 4.90 (d, *J* = 15.6 Hz, 1H, CH), 3.84 (dd, *J*₁ = 17.6 Hz, *J*₂ = 2.0 Hz, 1H, CH), 3.15 (d, *J* = 17.6 Hz, 1H, CH), 2.41 (s, 3H, CH₃); ¹³C NMR (400 MHz, CDCl₃) δ: 172.6, 142.5, 139.0, 136.5, 134.7, 131.5, 130.7, 129.3, 129.0, 128.9, 128.8, 127.9, 127.3, 126.7, 125.8, 124.5, 123.9, 122.2, 120.4, 119.7, 113.1, 110.7, 110.1, 108.0, 50.6, 47.8, 45.4, 44.4, 29.3, 21.3; IR (KBr) ν: 3367, 3310, 3245, 3087, 2956, 2833, 2155, 1860, 1671, 1655, 1543, 1445, 1361, 1267, 1158, 1141, 955, 901, 868, 783 cm⁻¹; MS (*m/z*): HRMS (ESI) Calcd. for C₃₅H₁₆N₄O ([M+Na]⁺): 541.1999, found: 541.2001.

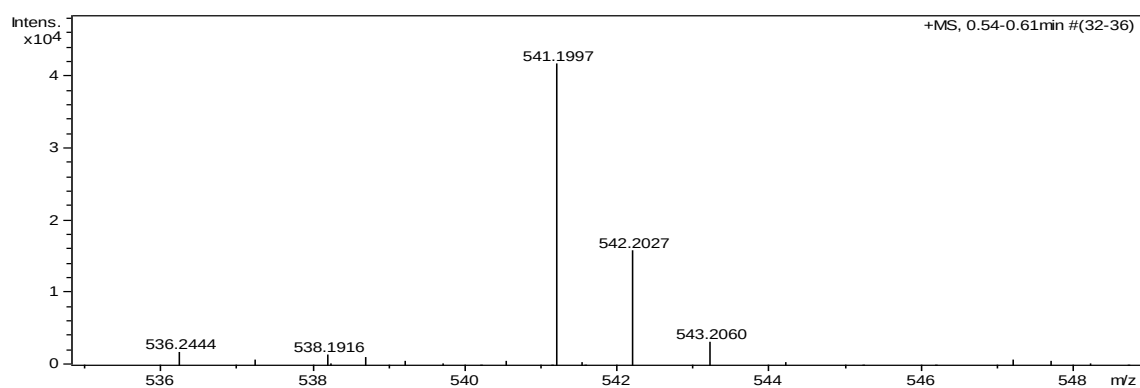
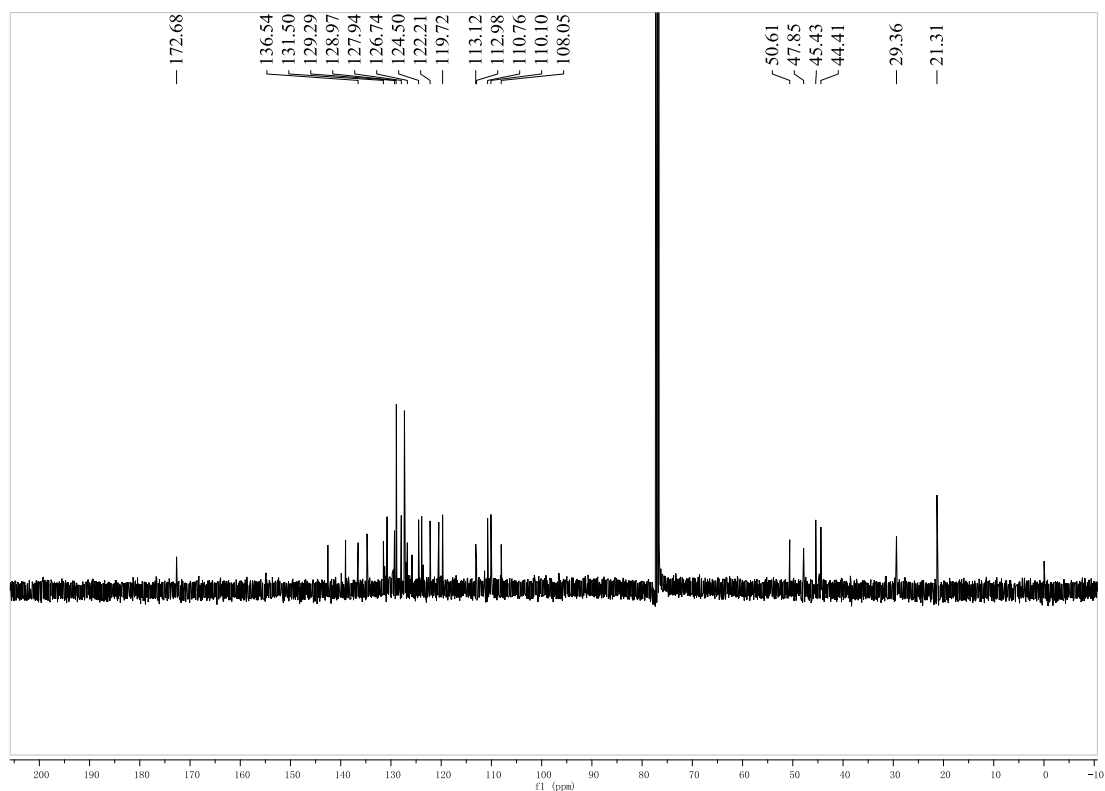




1'-Benzyl-2'-oxo-4-(*p*-tolyl)-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1*H*)-dicarbonitrile (2b'):

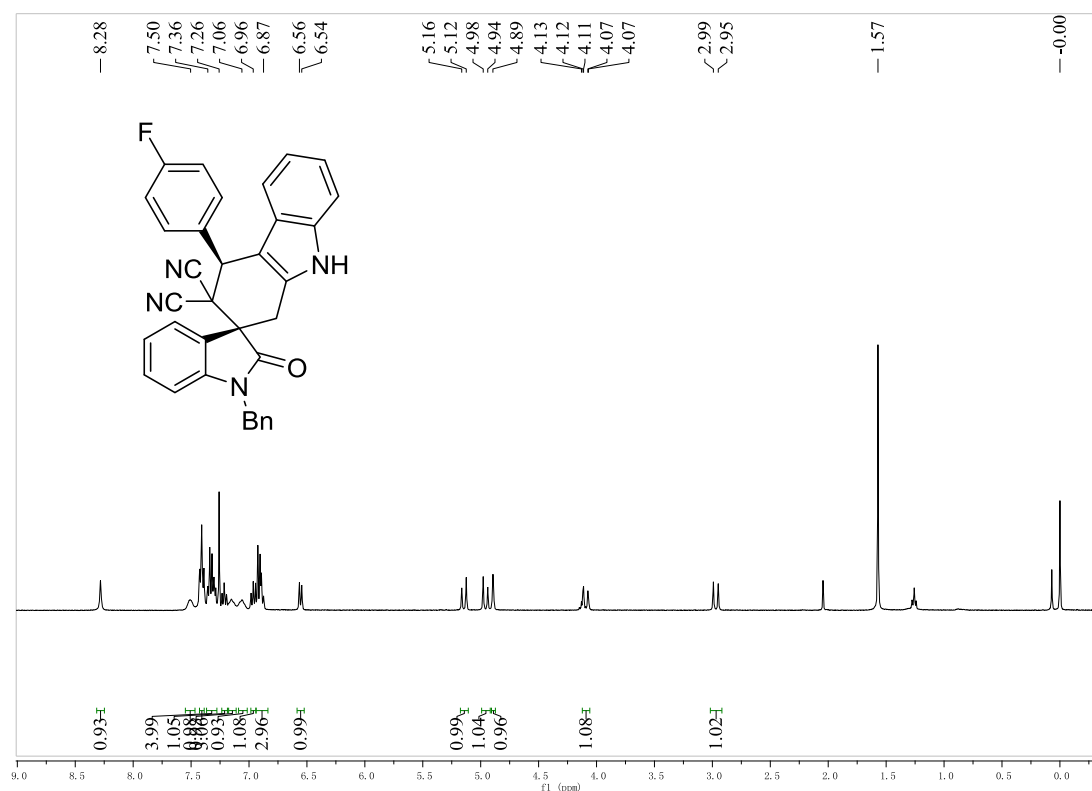
white solid, 10%, m.p. 205-208 °C; ¹H NMR (400 MHz, CDCl₃) δ: 8.00 (s, 1H, NH), 7.86 (d, *J* = 7.6 Hz, 1H, ArH), 7.55-7.52 (m, 1H, ArH), 7.37-7.33 (m, 5H, ArH), 7.31-7.27 (m, 3H, ArH), 7.27-7.25 (m, 1H, ArH), 7.19 (t, *J* = 7.6 Hz, 2H, ArH), 7.11 (t, *J* = 8.0 Hz, 1H, ArH), 6.87 (t, *J* = 8.8 Hz, 2H, ArH), 6.52 (d, *J* = 8.0 Hz, 1H, ArH), 5.47 (s, 1H, CH), 5.03 (d, *J* = 15.6 Hz, 1H, CH), 4.90 (d, *J* = 16.0 Hz, 1H, CH), 3.84 (dd, *J*₁ = 17.6 Hz, *J*₂ = 2.0 Hz, 1H, CH), 3.15 (d, *J* = 17.6 Hz, 1H, CH), 2.41 (s, 3H, CH₃); ¹³C NMR (400 MHz, CDCl₃) δ: 172.6, 142.5, 139.0, 136.5, 134.7, 131.5, 130.7, 129.2, 129.0, 128.9, 128.9, 127.9, 127.3, 126.7, 125.8, 124.4, 123.8, 122.2, 120.4, 119.7, 113.1, 112.9, 110.7, 110.1, 108.0, 50.6, 47.8, 45.4, 44.4, 29.3, 21.3; IR (KBr) ν: 3343, 3245, 3171, 3067, 2955, 2848, 2161, 1843, 1643, 1618, 1565, 1455, 1348, 1231, 1155, 1131, 984, 922, 862, 755 cm⁻¹; MS (*m/z*): HRMS (ESI) Calcd. for C₃₅H₁₆N₄O ([M+Na]⁺): 541.1999, found: 541.1997.

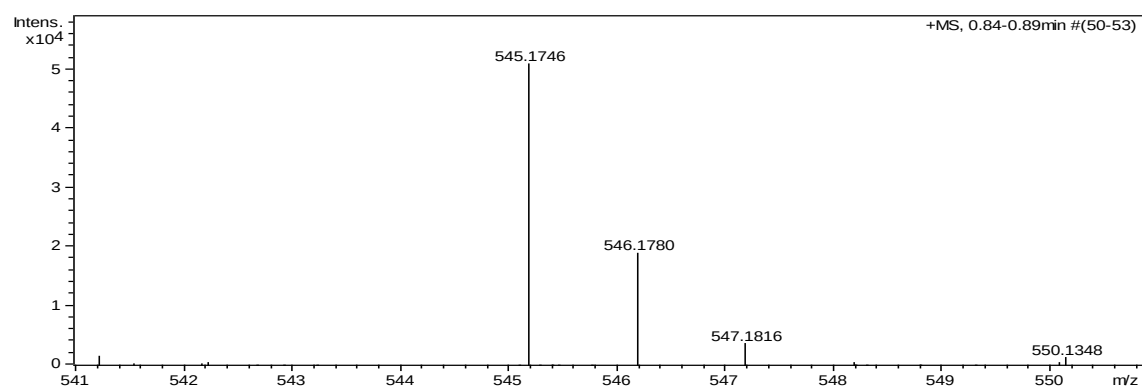
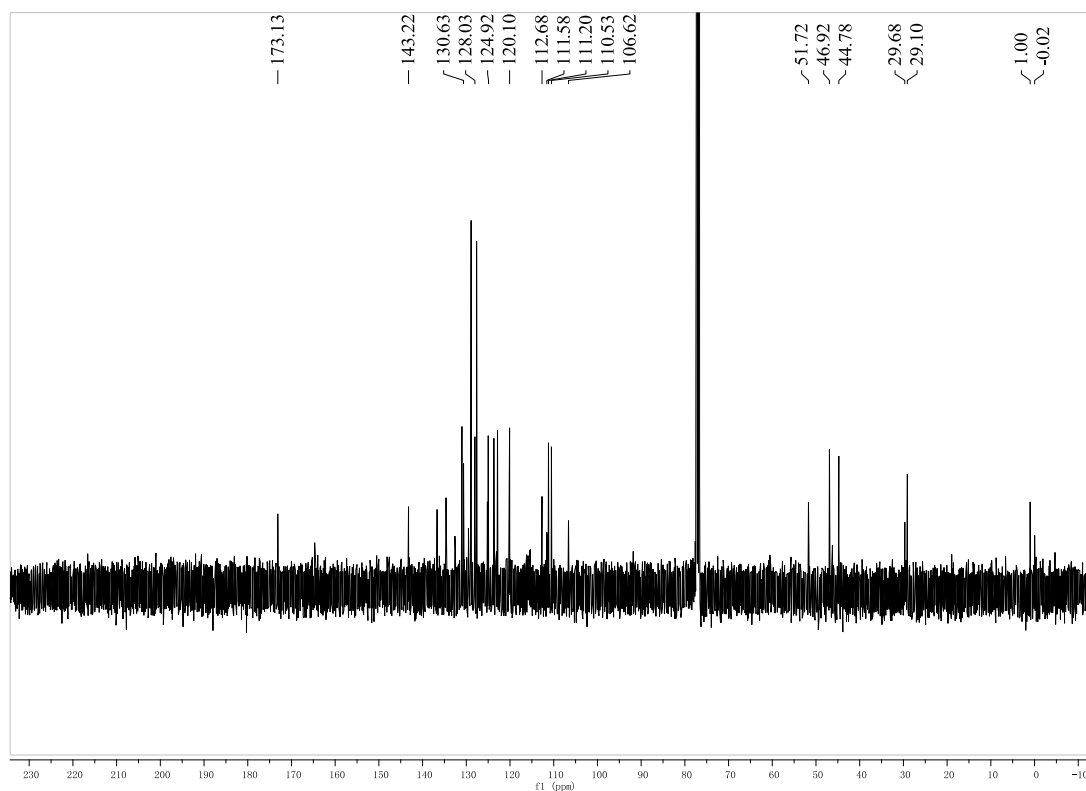




1'-Benzyl-4-(4-fluorophenyl)-2'-oxo-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1H)-dicarbonitrile (2c):

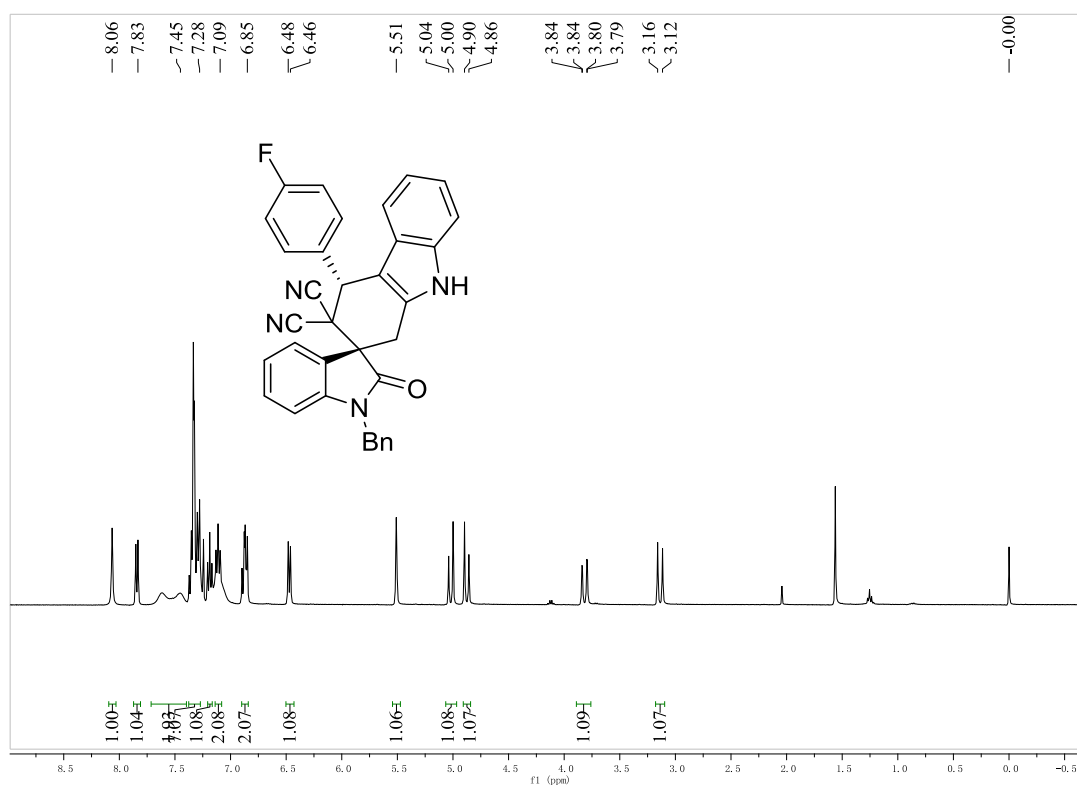
white solid, 56%, m.p. 201-204 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.28 (s, 1H, NH), 7.52-7.50 (m, 1H, ArH), 7.41 (t, $J = 7.6$ Hz, 3H, ArH), 7.36-7.29 (m, 4H, ArH), 7.22 (t, $J = 8.0$ Hz, 1H, ArH), 7.15-7.14 (m, 1H, ArH), 7.06-7.04 (m, 1H, ArH), 6.96 (t, $J = 8.0$ Hz, 1H, ArH), 6.92-6.88 (m, 3H, ArH), 6.55 (d, $J = 8.0$ Hz, 1H, ArH), 5.14 (d, $J = 11.6$ Hz, 1H, CH), 4.95 (d, $J = 11.6$ Hz, 1H, CH), 4.90 (s, 1H, CH), 4.09 (dd, $J_1 = 16.8$ Hz, $J_2 = 2.0$ Hz, 1H, CH), 2.96 (d, $J = 16.8$ Hz, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 173.1, 143.2, 136.6, 134.6, 131.0, 130.6, 129.5, 129.4, 128.8, 128.0, 127.6, 125.1, 124.9, 123.6, 122.8, 120.2, 120.0, 112.6, 111.5, 111.2, 110.5, 106.6, 51.7, 46.9, 44.7, 29.6 (d, $J = 5.0$ Hz), 1.0 (d, $J = 4.0$ Hz); IR (KBr) ν : 3451, 3133, 3078, 3001, 2961, 1978, 1755, 1648, 1567, 1519, 1466, 1367, 1314, 1255, 1131, 970, 880, 783 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{34}\text{H}_{23}\text{FN}_4\text{O}$ ($[\text{M}+\text{Na}]^+$): 545.1748, found: 545.1746.

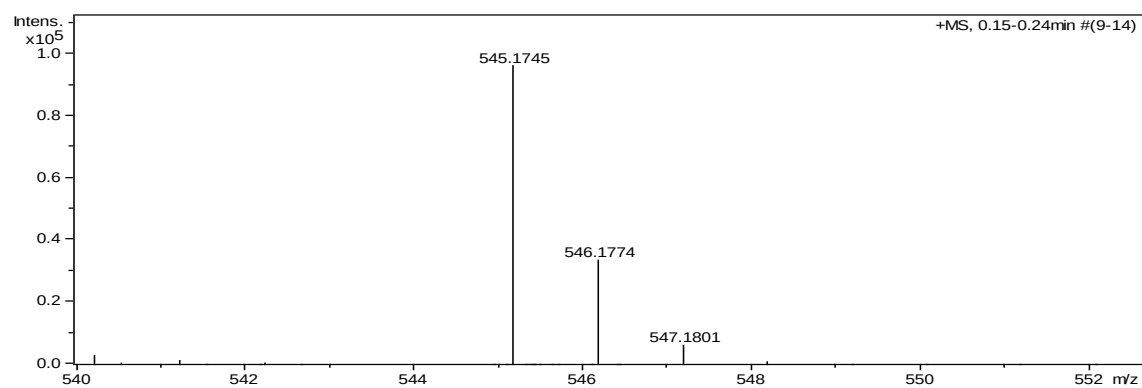
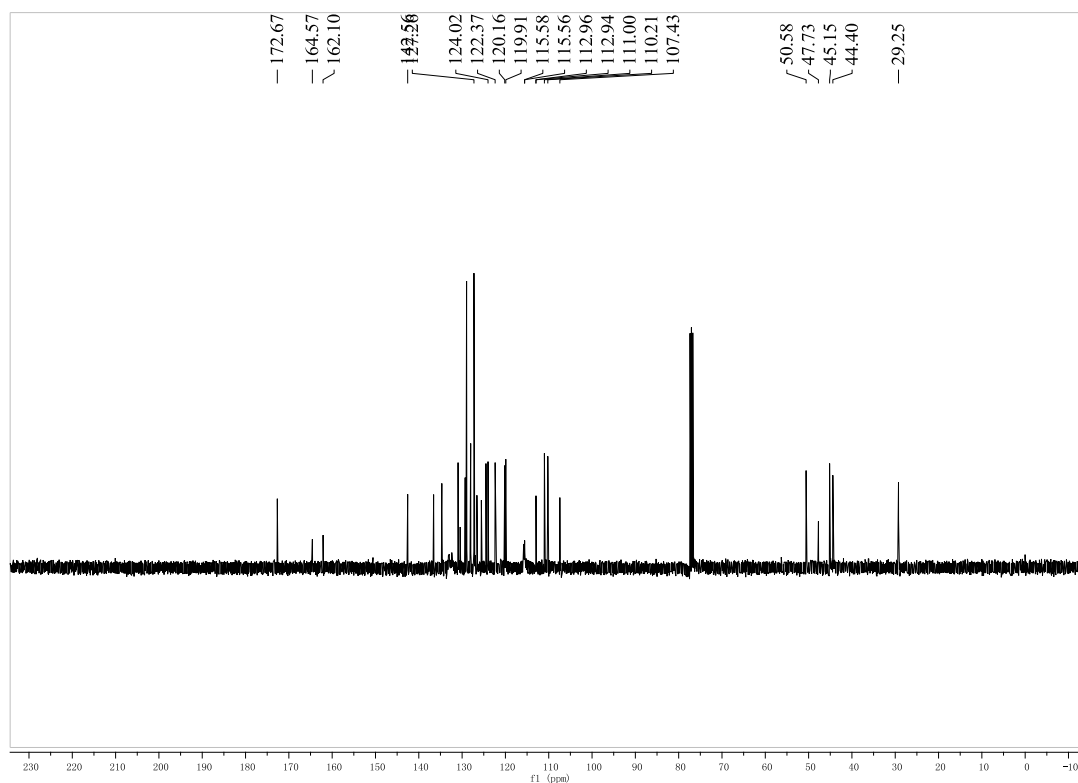




1'-Benzyl-4-(4-fluorophenyl)-2'-oxo-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1H)-dicyanonitrile (2c'):

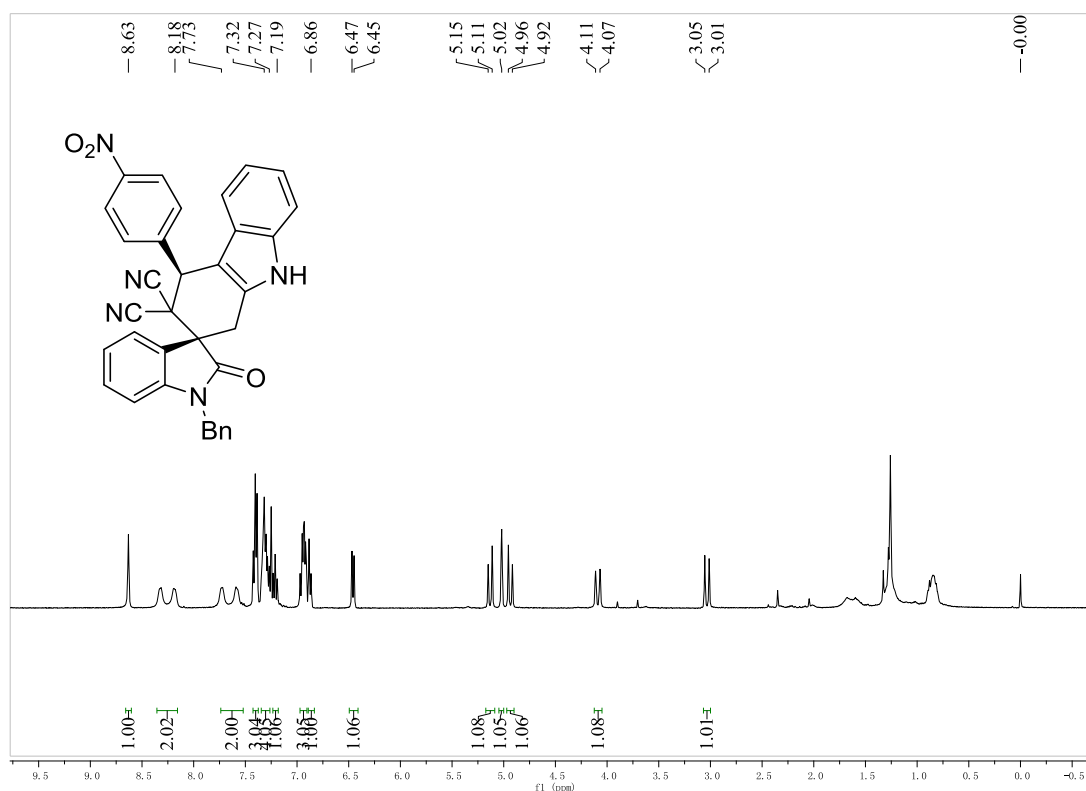
white solid, 8%, m.p. 215-218 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.07 (s, 1H, NH), 7.84 (d, J = 7.6 Hz, 1H, ArH), 7.62-7.45 (m, 2H, ArH), 7.37-7.28 (m, 7H, ArH), 7.19 (t, J = 7.6 Hz, 1H, ArH), 7.11 (t, J = 7.6 Hz, 2H, ArH), 6.90-6.85 (m, 2H, ArH), 6.47 (d, J = 8.0 Hz, 1H, ArH), 5.51 (s, 1H, CH), 5.01 (d, J = 16.0 Hz, 1H, CH), 4.87 (d, J = 16.0 Hz, 1H, CH), 3.81 (dd, J_1 = 17.6 Hz, J_2 = 2.0 Hz, 1H, CH), 3.14 (d, J = 17.6 Hz, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 172.6, 164.5, 162.1, 142.5, 136.5, 134.6, 130.9, 130.4, 129.2, 128.9, 128.0, 127.2, 126.5, 125.5, 124.4, 124.0, 122.3, 120.1, 119.9, 115.5, 115.5, 112.9, 112.9, 111.0, 110.2, 107.4, 50.5, 47.7, 45.1, 44.4, 29.2; IR (KBr) ν : 3417, 3078, 2978, 1843, 1765, 1678, 1617, 1583, 1457, 1355, 1317, 1248, 1131, 970, 861, 788 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{34}\text{H}_{23}\text{FN}_4\text{O}$ ($[\text{M}+\text{Na}]^+$): 545.1748, found: 545.1745.

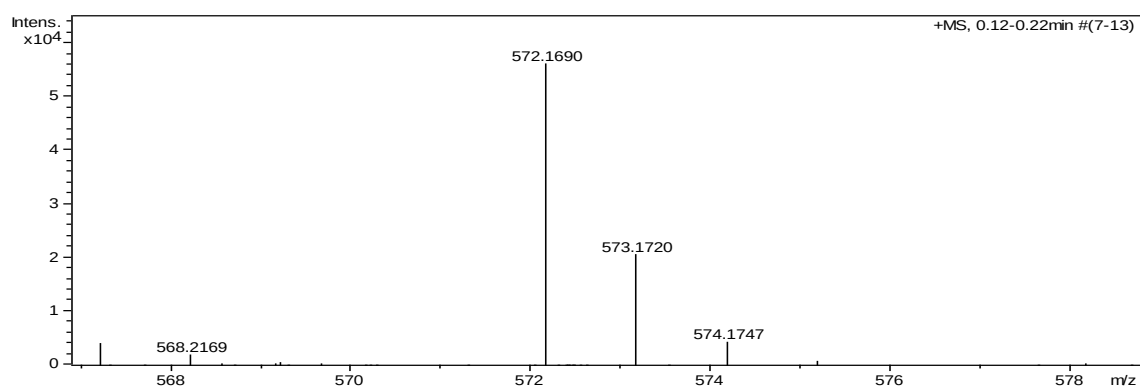
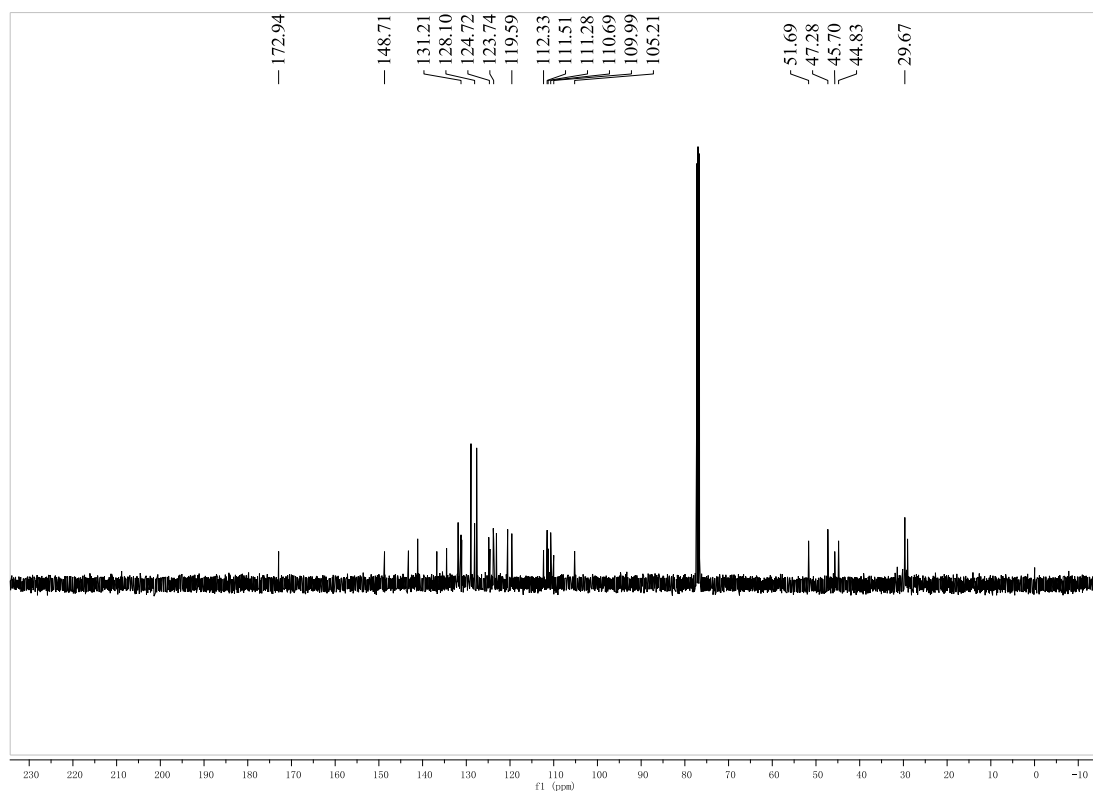




**1'-Benzyl-4-(4-nitrophenyl)-2'-oxo-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1H)-dicarbo
nitrile (2d):**

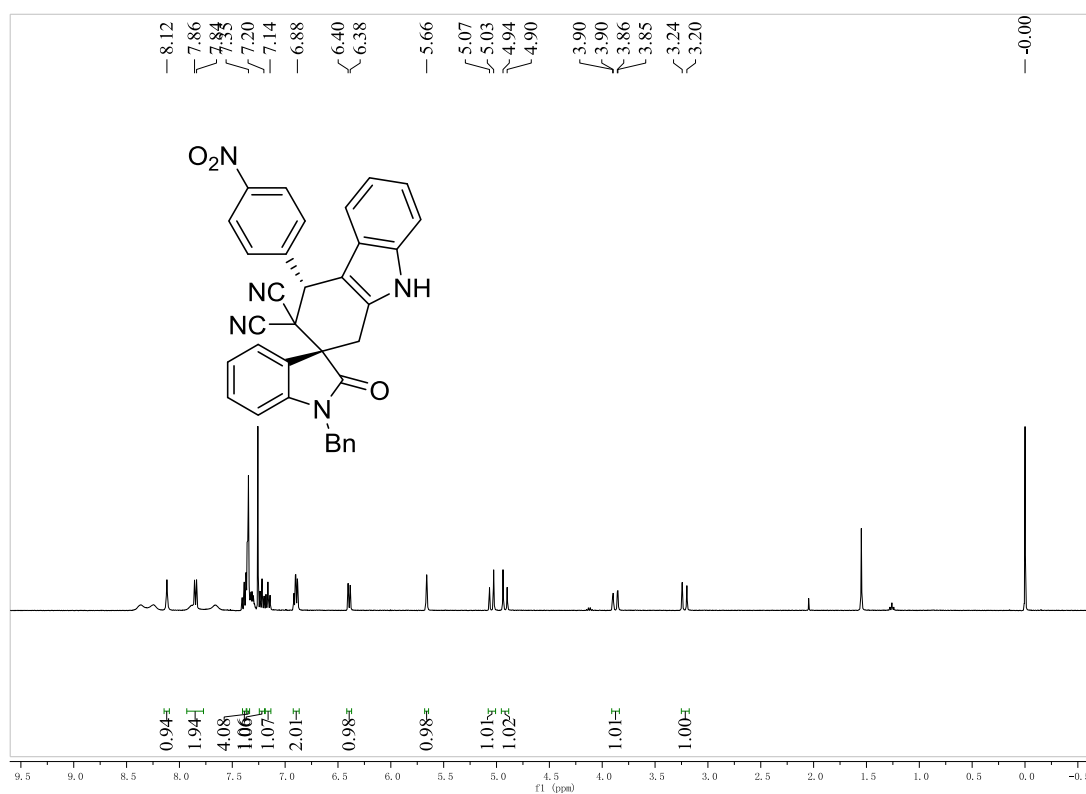
White solid, 56%, m.p. 191-194 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.63 (s, 1H, NH), 8.25 (dd, $J_1 = 54.0$ Hz, $J_2 = 5.2$ Hz, 2H, ArH), 7.64 (dd, $J_1 = 54.0$ Hz, $J_2 = 4.0$ Hz, 2H, ArH), 7.42-7.38 (m, 3H, ArH), 7.32-7.27 (m, 4H, ArH), 7.21 (t, $J = 8.0$ Hz, 1H, ArH), 6.97-6.92 (m, 3H, ArH), 6.87 (d, $J = 8.0$ Hz, 1H, ArH), 6.45 (d, $J = 8.0$ Hz, 1H, ArH), 5.13 (d, $J = 15.6$ Hz, 1H, CH), 5.02 (s, 1H, CH), 4.93 (d, $J = 15.6$ Hz, 1H, CH), 4.08 (d, $J = 17.2$ Hz, 1H, CH), 3.04 (d, $J = 16.8$ Hz, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 172.9, 148.7, 143.2, 141.1, 136.7, 134.5, 131.8, 131.2, 131.0, 128.9, 128.0, 127.6, 124.8, 124.7, 124.5, 123.8, 123.7, 123.1, 120.5, 119.5, 112.3, 111.5, 111.2, 110.6, 109.9, 105.2, 51.6, 47.2, 45.7, 44.8, 29.6; IR (KBr) ν : 3356, 3300, 3249, 3154, 3048, 2963, 1847, 1763, 1632, 1607, 1548, 1467, 1332, 1310, 1249, 1137, 959, 843 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{32}\text{H}_{29}\text{NO}_4$ ($[\text{M}+\text{Na}]^+$): 572.1693, found: 572.1690.

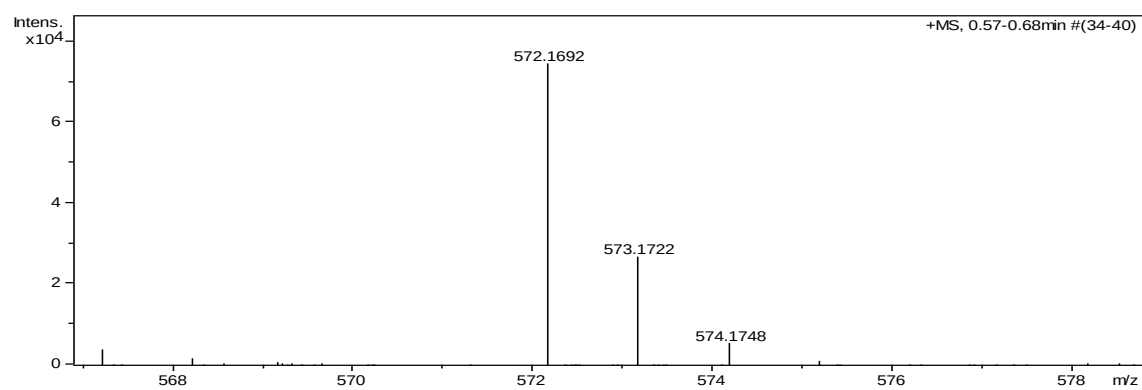
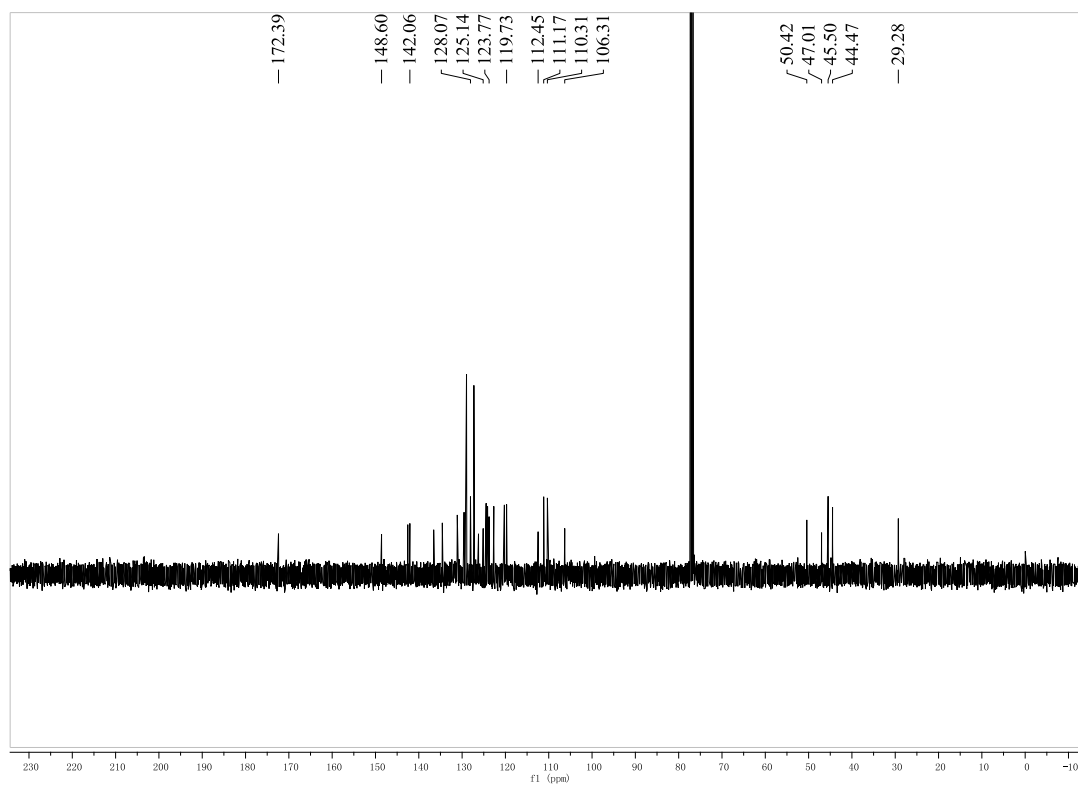




1'-Benzyl-4-(4-nitrophenyl)-2'-oxo-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1H)-dicarbonitrile (2d')

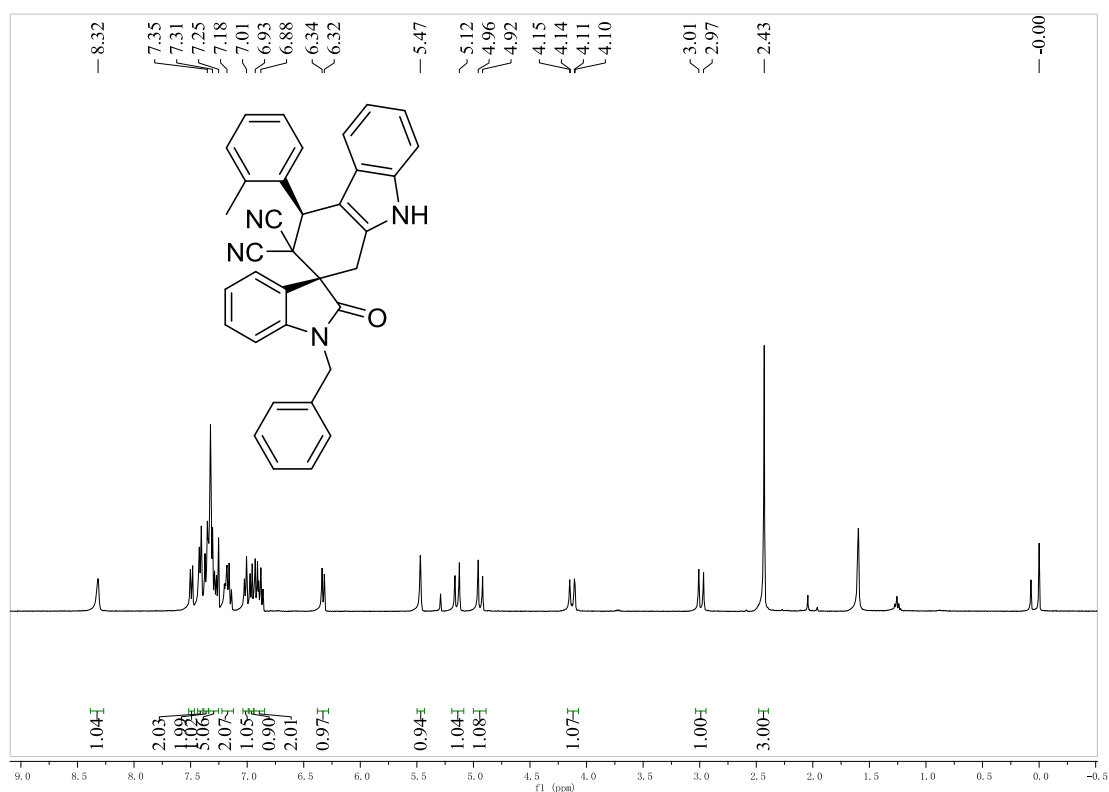
White solid, 7%, m.p. 200-203 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.12 (s, 1H, NH), 7.84 (d, J = 7.6 Hz, 2H, ArH), 7.41-7.37 (m, 1H, ArH), 7.36-7.35 (m, 4H, ArH), 7.16 (t, J = 7.2 Hz, 1H, ArH), 6.92-6.88 (m, 2H, ArH), 6.39 (d, J = 8.0 Hz, 1H, ArH), 5.66 (s, 1H, CH), 5.04 (d, J = 15.6 Hz, 1H, CH), 4.92 (d, J = 15.6 Hz, 1H, CH), 3.87 (dd, J_1 = 17.6 Hz, J_2 = 2.0 Hz, 1H, CH), 3.22 (d, J = 17.6 Hz, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 172.3, 148.5, 142.5, 142.0, 136.5, 134.5, 131.1, 129.5, 128.9, 128.0, 127.2, 126.2, 125.1, 124.4, 124.1, 123.7, 122.6, 120.2, 119.7, 112.4, 111.1, 110.3, 106.3, 50.4, 47.0, 45.5, 44.4, 29.2; IR (KBr) ν : 3418, 3240, 3179, 3054, 3004, 2976, 1857, 1767, 1648, 1638, 1564, 1433, 1378, 1334, 1258, 1165, 944, 831, 767 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{32}\text{H}_{29}\text{NO}_4$ ($[\text{M}+\text{Na}]^+$): 572.1693, found: 572.1692.

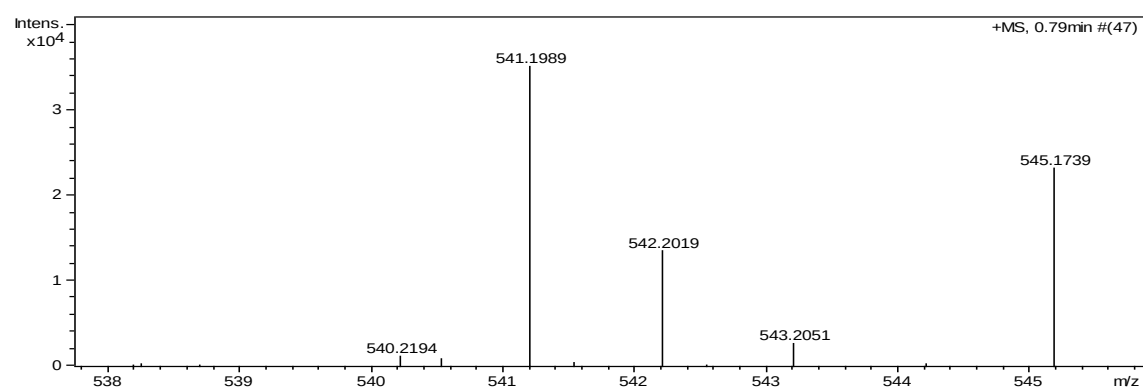
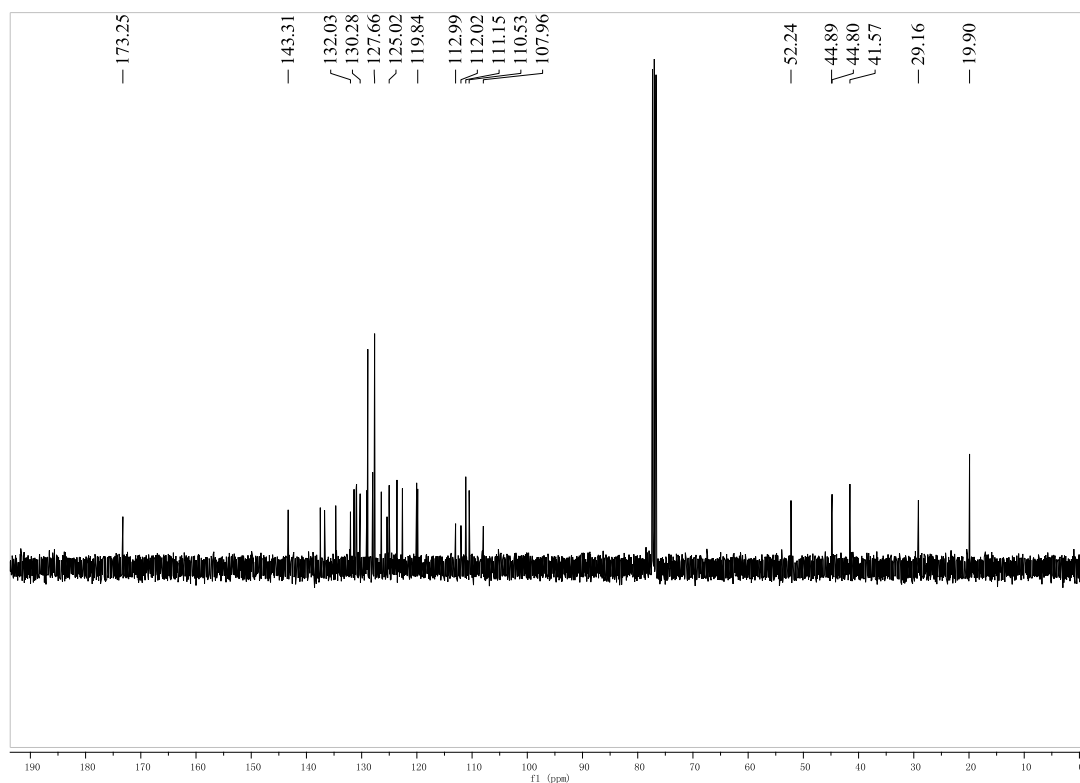




3-((2-Azanylidene)-3-methyl)-1'-benzyl-2'-oxo-4-(o-tolyl)-1,3,4,9-tetrahydrospiro[carbazole-2,3'-indoline]-3-carbonitrile (2e):

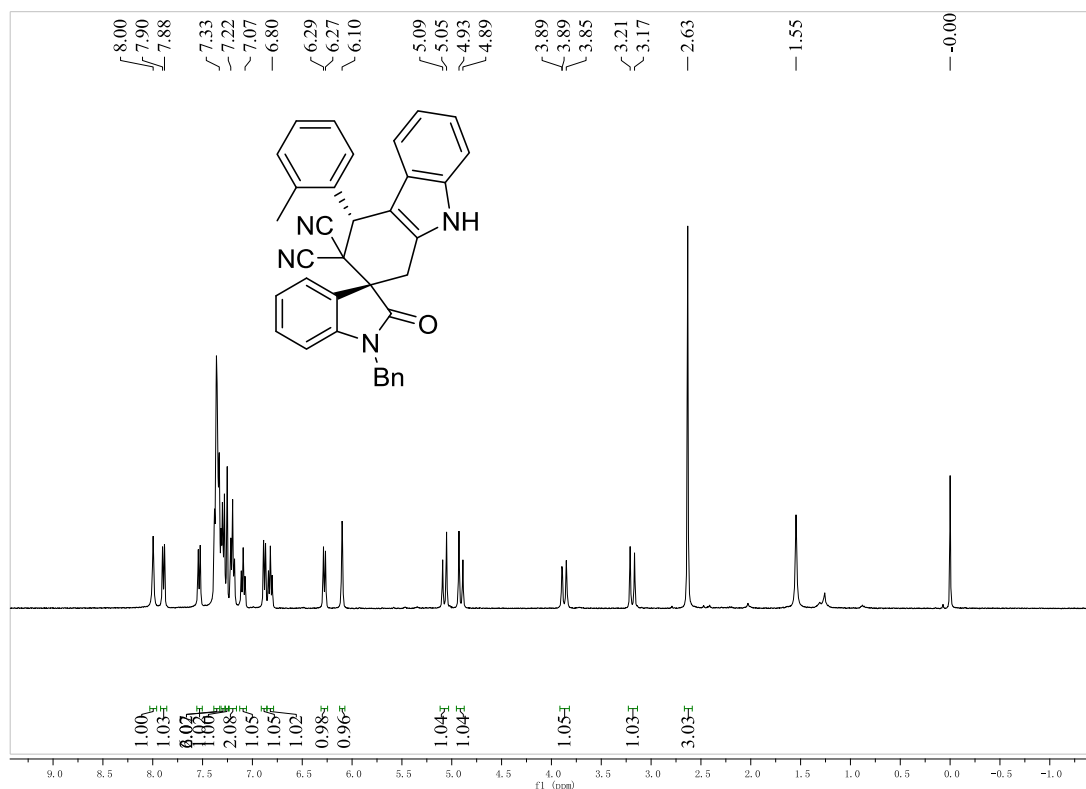
white solid, 57%, m.p. 187-190 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.32 (s, 1H, NH), 7.49 (d, J = 8.0 Hz, 1H, ArH), 7.41 (d, J = 6.8 Hz, 2H, ArH), 7.37-7.34 (m, 2H, ArH), 7.33-7.25 (m, 5H, ArH), 7.20-7.16 (m, 2H, ArH), 7.01 (d, J = 7.2 Hz, 1H, ArH), 6.96 (d, J = 7.6 Hz, 1H, ArH), 6.93-6.88 (m, 2H, ArH), 6.32 (d, J = 8.0 Hz, 1H, ArH), 5.47 (s, 1H, CH), 5.14 (d, J = 15.6 Hz, 1H, CH), 4.93 (d, J = 15.6 Hz, 1H, CH), 4.12 (dd, J_1 = 16.8 Hz, J_2 = 2.0 Hz, 1H, CH), 2.98 (d, J = 16.8 Hz, 1H, CH), 2.43 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 173.2, 143.3, 137.4, 136.7, 134.6, 132.0, 131.3, 130.9, 130.9, 130.2, 129.0, 128.8, 128.0, 127.6, 126.4, 125.4, 125.1, 125.0, 123.6, 122.6, 120.0, 119.8, 112.9, 112.0, 111.1, 110.5, 107.9, 52.2, 44.8, 44.8, 41.5, 29.1, 19.9; IR (KBr) ν : 3300, 3248, 3167, 2932, 2847, 2166, 1841, 1638, 1628, 1531, 1466, 1354, 1218, 1168, 1131, 955, 834, 789 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{35}\text{H}_{16}\text{N}_4\text{O}$ ($[\text{M}+\text{Na}]^+$): 541.1999, found: 541.1989.

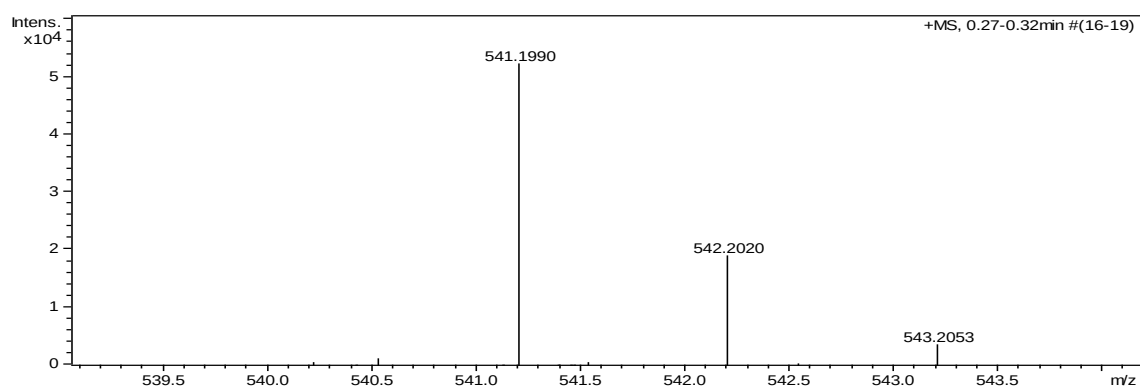
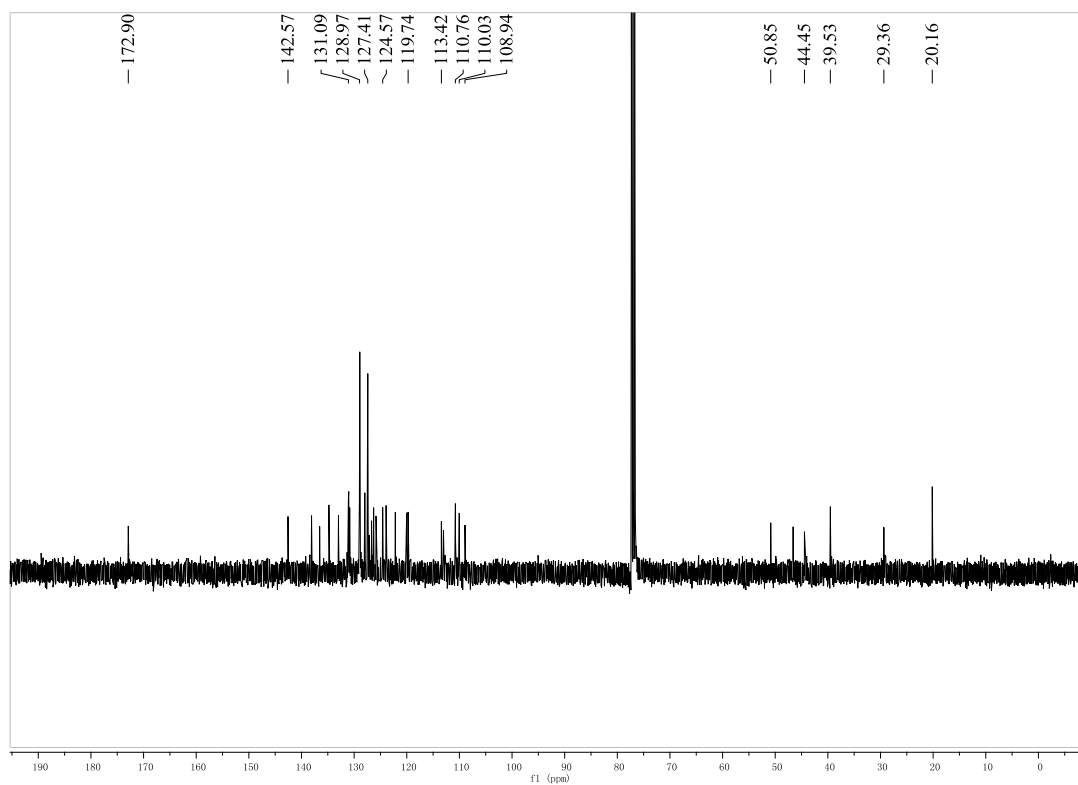




1'-Benzyl-2'-oxo-4-(*o*-tolyl)-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1*H*)-dicarbonitrile (2e')

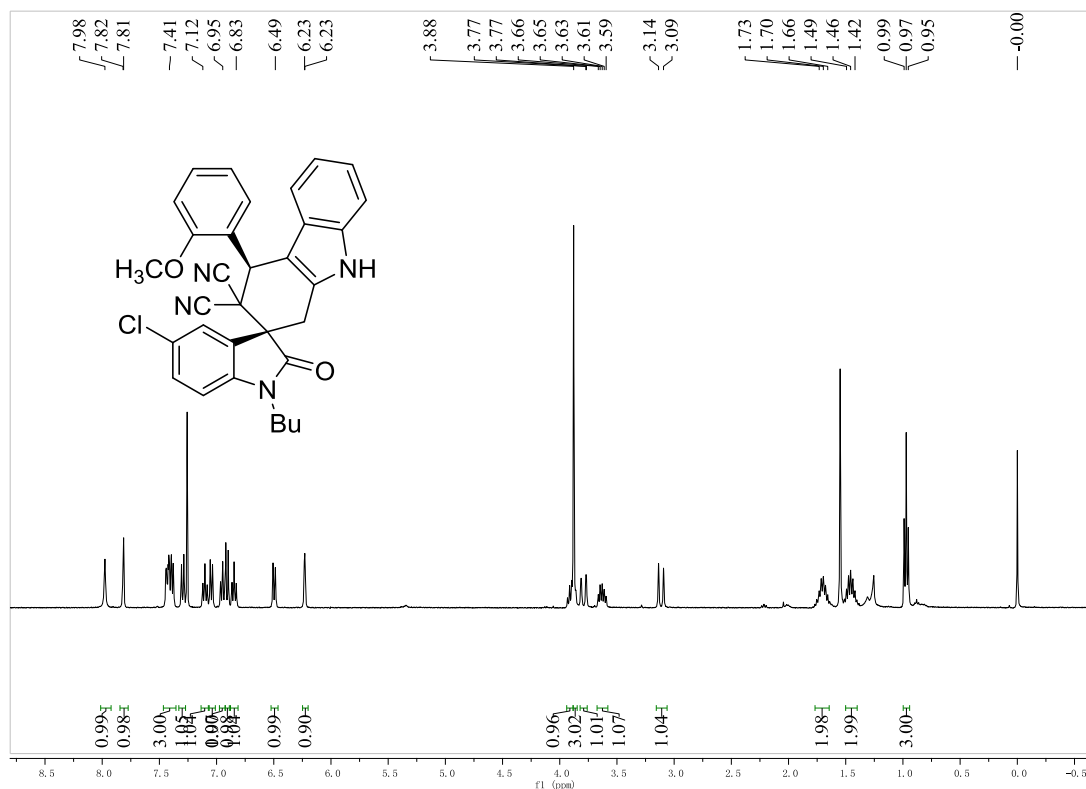
white solid, 7%, m.p. 196-199 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.00 (s, 1H, NH), 7.89 (d, J = 7.6 Hz, 1H, ArH), 7.53 (d, J = 7.6 Hz, 1H, ArH), 7.38-7.33 (m, 6H, ArH), 7.31-7.28 (m, 2H, ArH), 7.20 (t, J = 7.6 Hz, 2H, ArH), 7.09 (t, J = 7.6 Hz, 1H, ArH), 6.82 (t, J = 7.6 Hz, 1H, ArH), 6.28 (d, J = 8.0 Hz, 1H, ArH), 6.10 (s, 1H, CH), 5.07 (d, J = 15.6 Hz, 1H, CH), 4.90 (d, J = 16.0 Hz, 1H, CH), 3.87 (d, J = 16.4 Hz, 1H, CH), 3.18 (d, J = 17.6 Hz, 1H, CH), 2.63 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 172.9, 142.5, 138.0, 136.5, 134.7, 132.9, 131.0, 131.0, 130.8, 129.0, 128.9, 128.9, 128.8, 127.9, 127.4, 126.6, 126.2, 125.8, 124.5, 123.9, 122.1, 120.0, 119.7, 113.4, 110.7, 110.0, 108.9, 50.8, 46.5, 44.4, 39.5, 29.3, 20.1; IR (KBr) ν : 3289, 3231, 3178, 3055, 2948, 2867, 2148, 1855, 1648, 1637, 1546, 1417, 1376, 1236, 1138, 1137, 937, 948, 845, 736 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{35}\text{H}_{16}\text{N}_4\text{O}$ ($[\text{M}+\text{Na}]^+$): 541.1999, found: 541.1990.

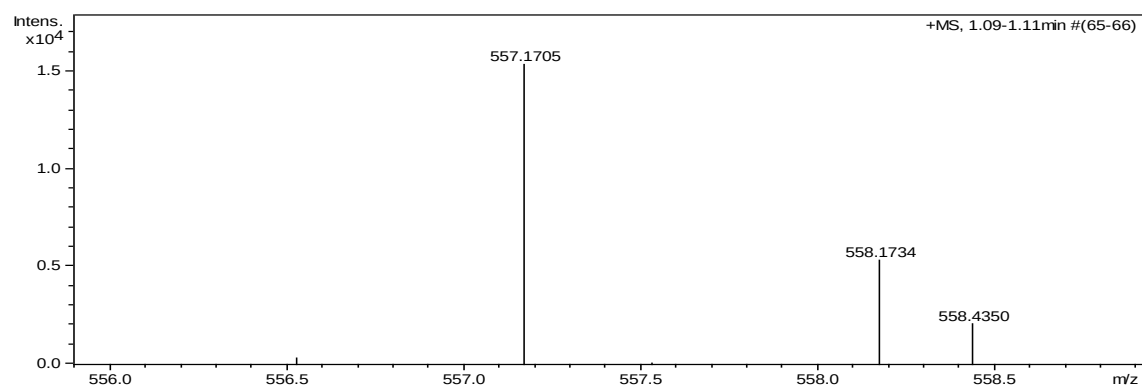
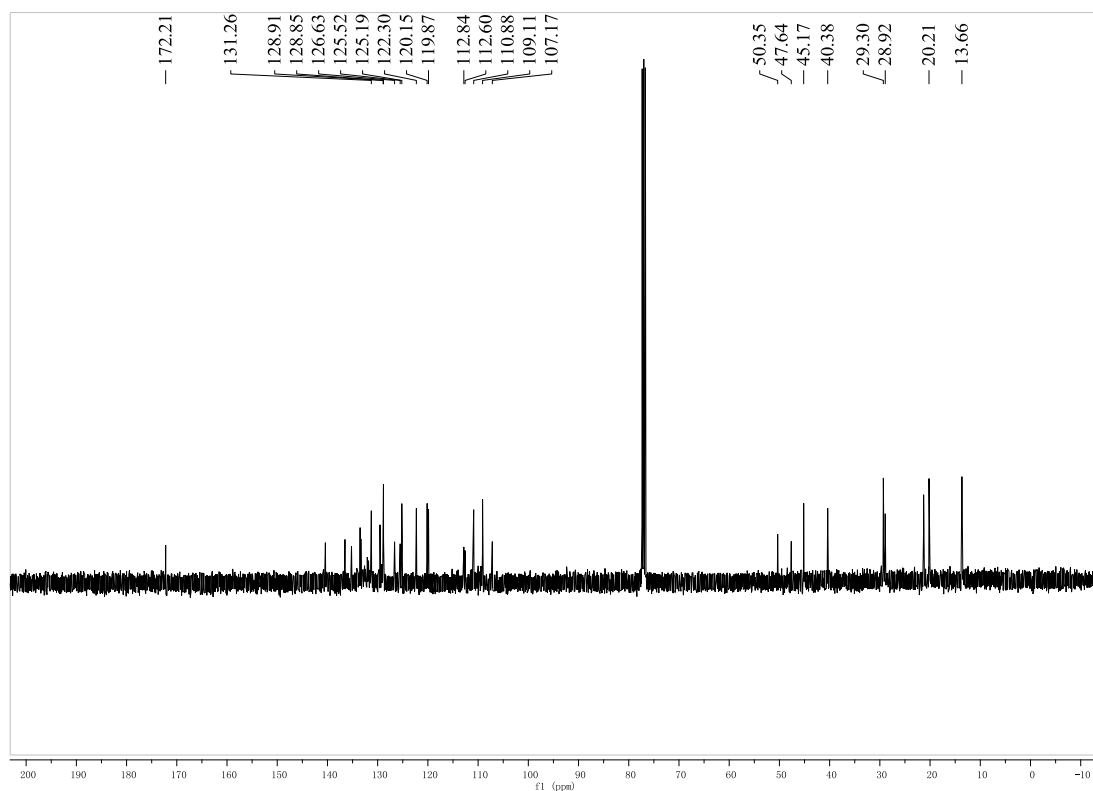




1'-Butyl-5'-chloro-4-(2-methoxyphenyl)-2'-oxo-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1H)-dicarbonitrile (2f):

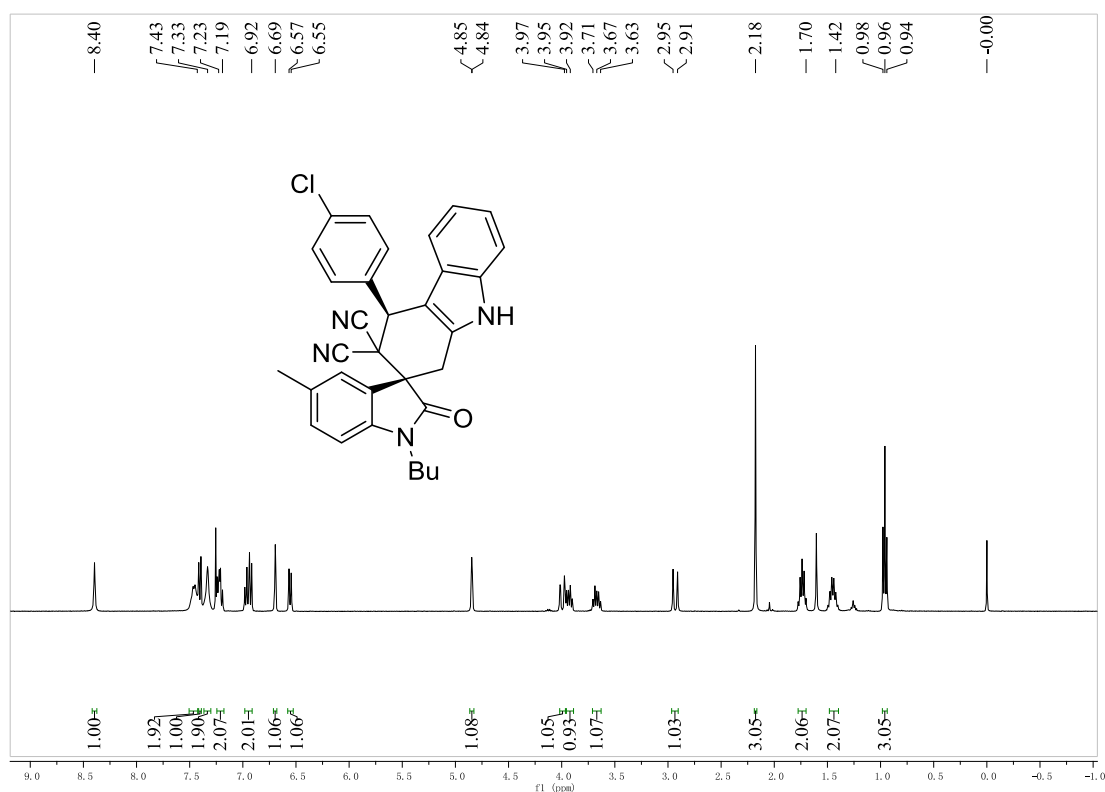
white solid, 58%, m.p. 190-193 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.98 (s, 1H, NH), 7.81 (d, J = 2.0 Hz, 1H, ArH), 7.45-7.38 (m, 3H, ArH), 7.29 (d, J = 8.0 Hz, 1H, ArH), 7.10 (t, J = 8.0 Hz, 1H, ArH), 7.04 (d, J = 8.0 Hz, 1H, ArH), 6.95 (t, J = 7.6 Hz, 1H, ArH), 6.91 (d, J = 8.8 Hz, 1H, ArH), 6.85 (t, J = 7.6 Hz, 1H, ArH), 6.49 (d, J = 8.4 Hz, 1H, ArH), 6.23 (s, 1H, CH), 3.93-3.90 (m, 1H, CH), 3.88 (s, 3H, OCH_3), 3.79 (dd, J_1 = 17.2 Hz, J_2 = 2.0 Hz, 1H, CH), 3.66-3.60 (m, 1H, CH), 3.11 (d, J = 17.6 Hz, 1H, CH), 1.74-1.66 (m, 2H, CH_2), 1.49-1.42 (m, 2H, CH_2), 0.97 (t, J = 7.2 Hz, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 172.2, 140.4, 136.5, 135.2, 133.4, 133.3, 132.0, 131.2, 129.5, 128.9, 128.9, 128.8, 126.6, 125.5, 125.1, 122.2, 120.1, 119.8, 112.8, 112.5, 110.8, 109.1, 107.1, 50.3, 47.6, 45.1, 40.3, 29.3, 28.9, 21.2, 20.2, 13.6; IR (KBr) ν : 3300, 3278, 3158, 3033, 2967, 2856, 2148, 1940, 1654, 1600, 1548, 1417, 1343, 1243, 1155, 1127, 945, 855, 748 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{32}\text{H}_{27}\text{Cl}^{35}\text{N}_4\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 557.1715, found: 557.1705. $\text{C}_{32}\text{H}_{27}\text{Cl}^{37}\text{N}_4\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 558.1748, found: 558.1734.

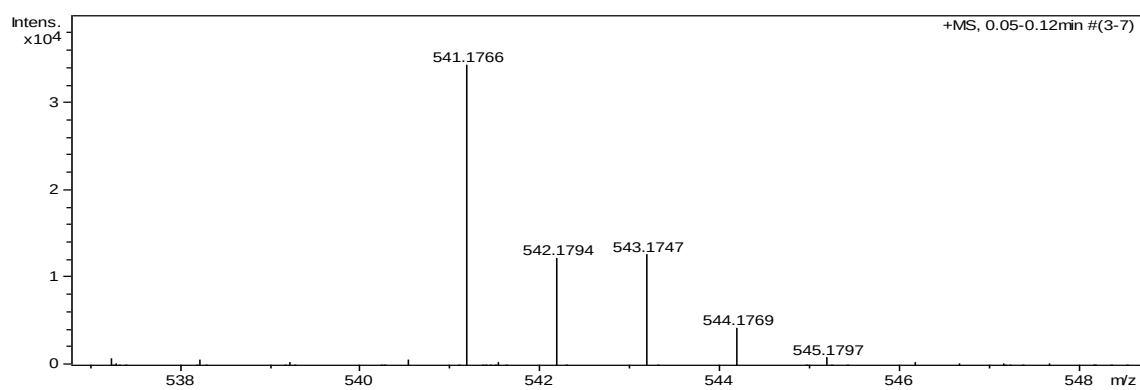
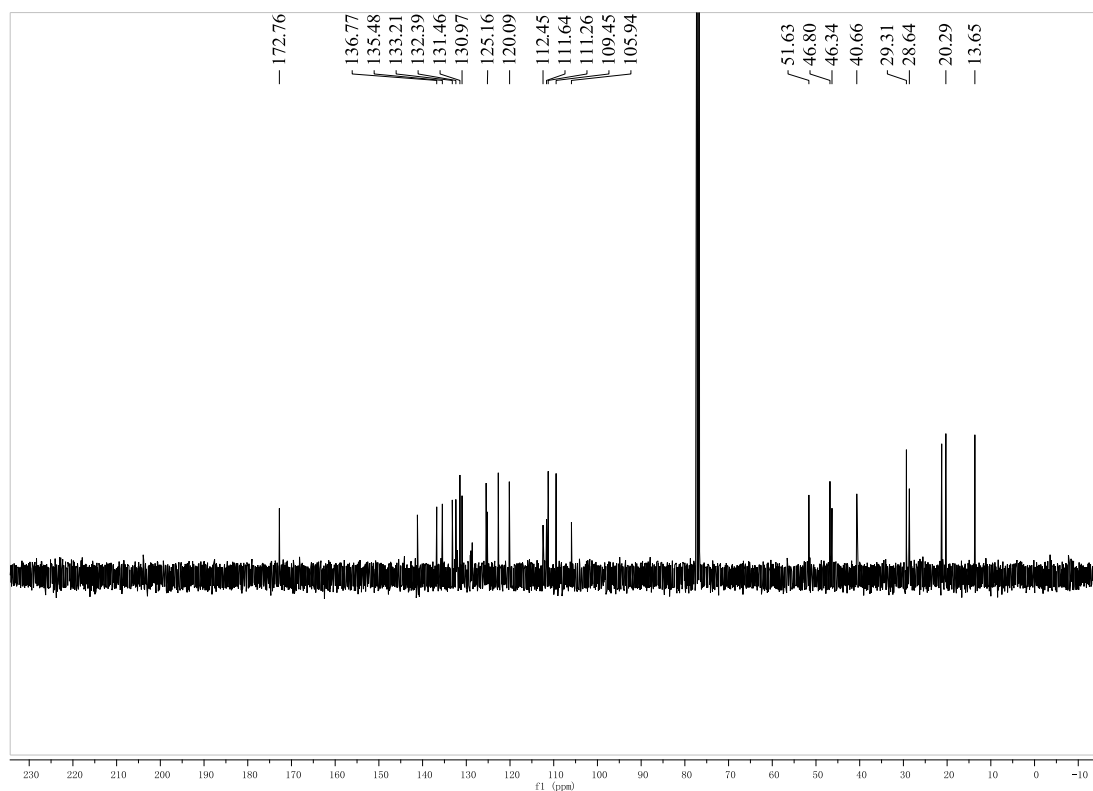




1'-Butyl-4-(4-chlorophenyl)-5'-methyl-2'-oxo-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1H)-dicarbonitrile (2g):

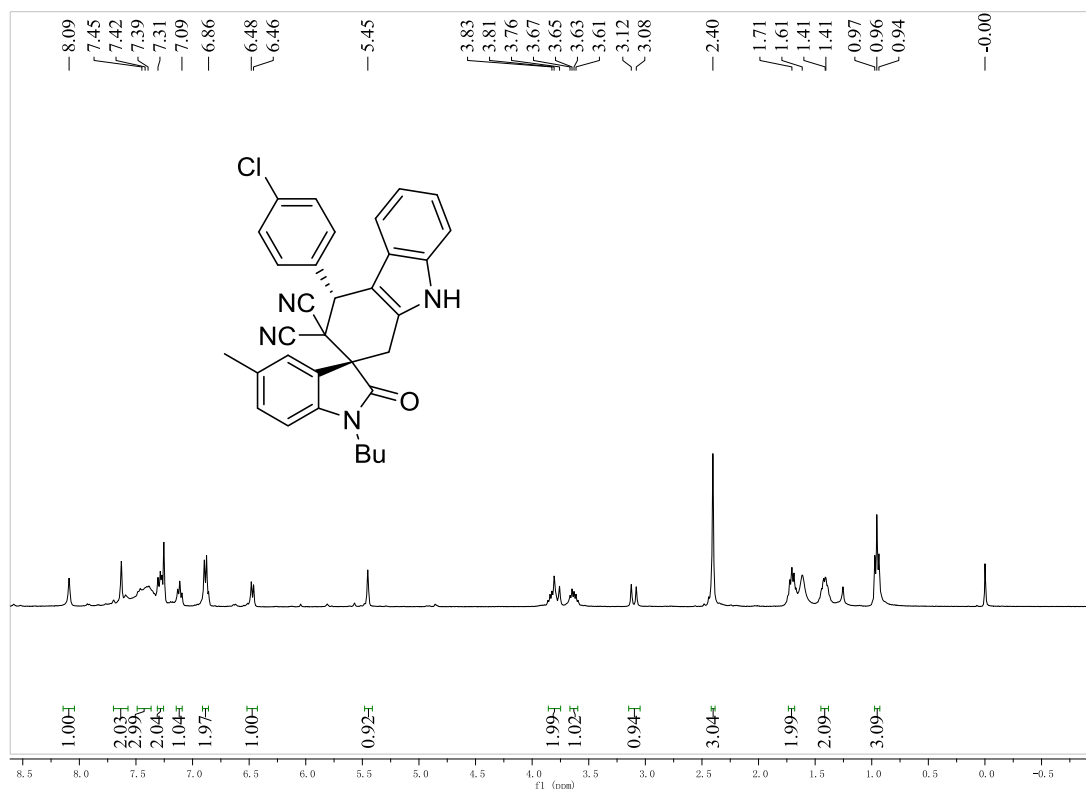
white solid, 54%, m.p. 186-189 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.40 (s, 1H, NH), 7.47-7.43 (m, 2H, ArH), 7.41 (d, $J = 8.4$ Hz, 1H, ArH), 7.34-7.33 (m, 2H, ArH), 7.24-7.19 (m, 2H, ArH), 6.95 (dd, $J_1 = 18.0$ Hz, $J_2 = 8.0$ Hz, 2H, ArH), 6.70-6.69 (m, 1H, ArH), 6.55 (d, $J = 8.0$ Hz, 1H, ArH), 4.84 (d, $J = 1.2$ Hz, 1H, CH), 3.99 (dd, $J_1 = 17.2$ Hz, $J_2 = 2.0$ Hz, 1H, CH), 3.92 (q, $J = 7.2$ Hz, 1H, CH), 3.71-3.63 (m, 1H, CH), 2.93 (d, $J = 17.2$ Hz, 1H, CH), 2.18 (s, 3H, CH_3), 1.78-1.70 (m, 2H, CH_2), 1.48-1.42 (m, 2H, CH_2), 0.96 (t, $J = 7.2$ Hz, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 172.7, 141.1, 136.7, 135.4, 133.2, 132.3, 131.4, 130.9, 125.4, 125.2, 125.1, 122.6, 120.1, 120.0, 112.4, 111.6, 111.2, 109.4, 105.9, 51.6, 46.8, 46.3, 40.6, 29.3, 28.6, 21.2, 20.2, 13.6; IR (KBr) ν : 3300, 3259, 3167, 3017, 2955, 2837, 2148, 1865, 1681, 1631, 1548, 1467, 1355, 1248, 1131, 1101, 947, 902, 846 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{32}\text{H}_{27}\text{Cl}^{35}\text{N}_4\text{O}$ ($[\text{M}+\text{Na}]^+$): 541.1766, found: 541.1766. $\text{C}_{32}\text{H}_{27}\text{Cl}^{37}\text{N}_4\text{O}$ ($[\text{M}+\text{Na}]^+$): 542.1799, found: 542.1794.

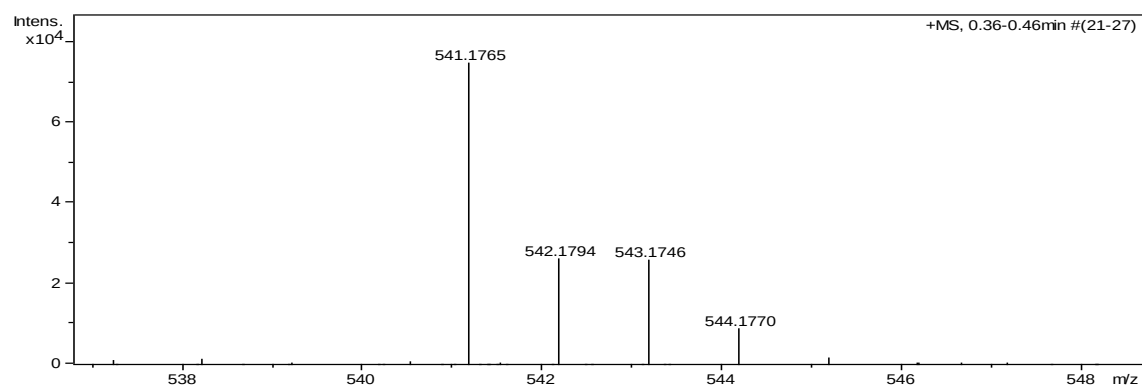
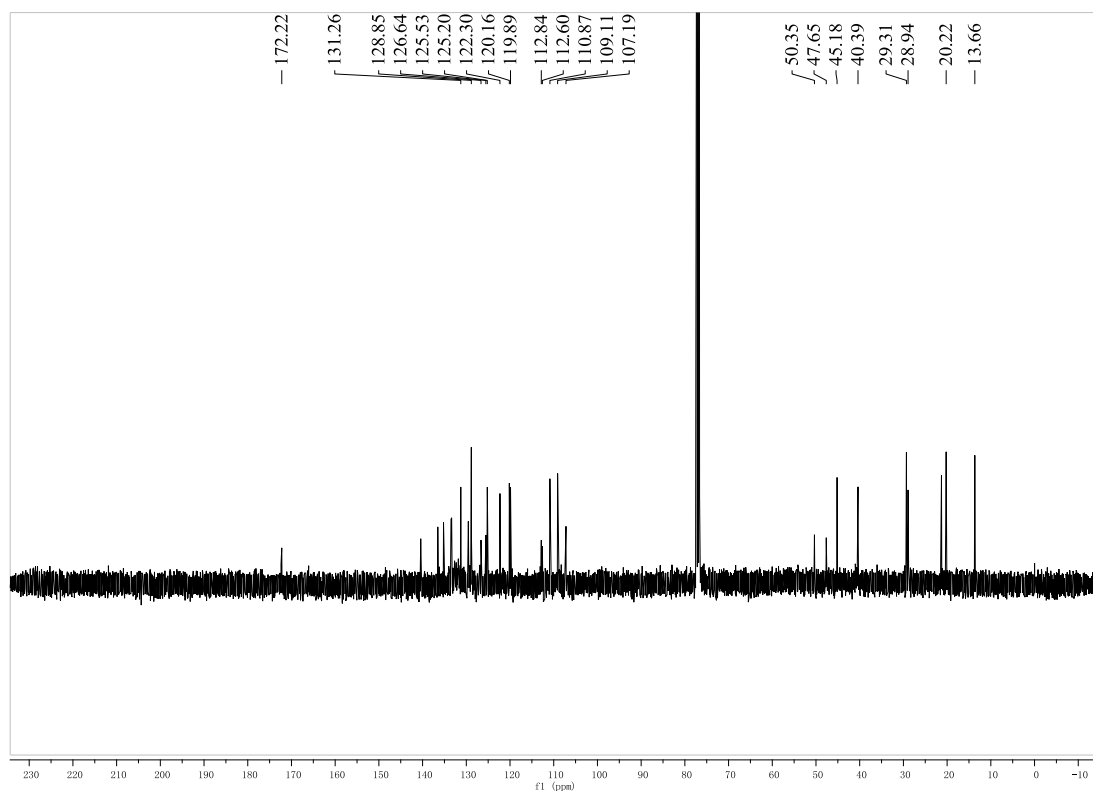




1'-Butyl-4-(4-chlorophenyl)-5'-methyl-2'-oxo-4,9-dihydrospiro[carbazole-2,3'-indoline]-3,3(1H)-dicarbonitrile (2g'):

white solid, 6%, m.p. 193-197 °C; ¹H NMR (400 MHz, CDCl₃) δ: 8.09 (s, 1H, NH), 7.70-7.59 (m, 2H, ArH), 7.48-7.37 (m, 3H, ArH), 7.29 (t, *J* = 8.4 Hz, 2H, ArH), 7.11 (t, *J* = 8.0 Hz, 1H, ArH), 6.88 (t, *J* = 8.0 Hz, 2H, ArH), 6.47 (d, *J* = 8.0 Hz, 1H, ArH), 5.45 (s, 1H, CH), 3.84-3.75 (m, 2H, CH₂), 3.64 (q, *J* = 7.2 Hz, 1H, CH), 3.10 (d, *J* = 17.6 Hz, 1H, CH), 2.40 (s, 3H, CH₃), 1.71 (t, *J* = 7.2 Hz, 2H, CH₂), 1.43-1.41 (m, 2H, CH₂), 0.96 (t, *J* = 7.2 Hz, 3H, CH₃); ¹³C NMR (400 MHz, CDCl₃) δ: 172.2, 140.4, 136.5, 135.2, 133.5, 133.3, 131.2, 129.5, 128.9, 128.8, 126.6, 125.5, 125.1, 122.3, 120.1, 119.8, 112.8, 112.5, 110.8, 109.1, 107.1, 50.3, 47.6, 45.1, 40.3, 29.3, 28.9, 21.2, 20.2, 13.6; IR (KBr) ν: 3300, 3248, 3170, 3049, 2966, 2849, 2170, 1864, 1654, 1600, 1560, 1455, 1348, 1267, 1131, 1100, 970, 917, 855 cm⁻¹; MS (*m/z*): HRMS (ESI) Calcd. for C₃₂H₂₇Cl³⁵N₄O ([M+Na]⁺): 541.1766, found: 541.1765. C₃₂H₂₇Cl³⁷N₄O ([M+Na]⁺): 542.1799, found: 542.1794.

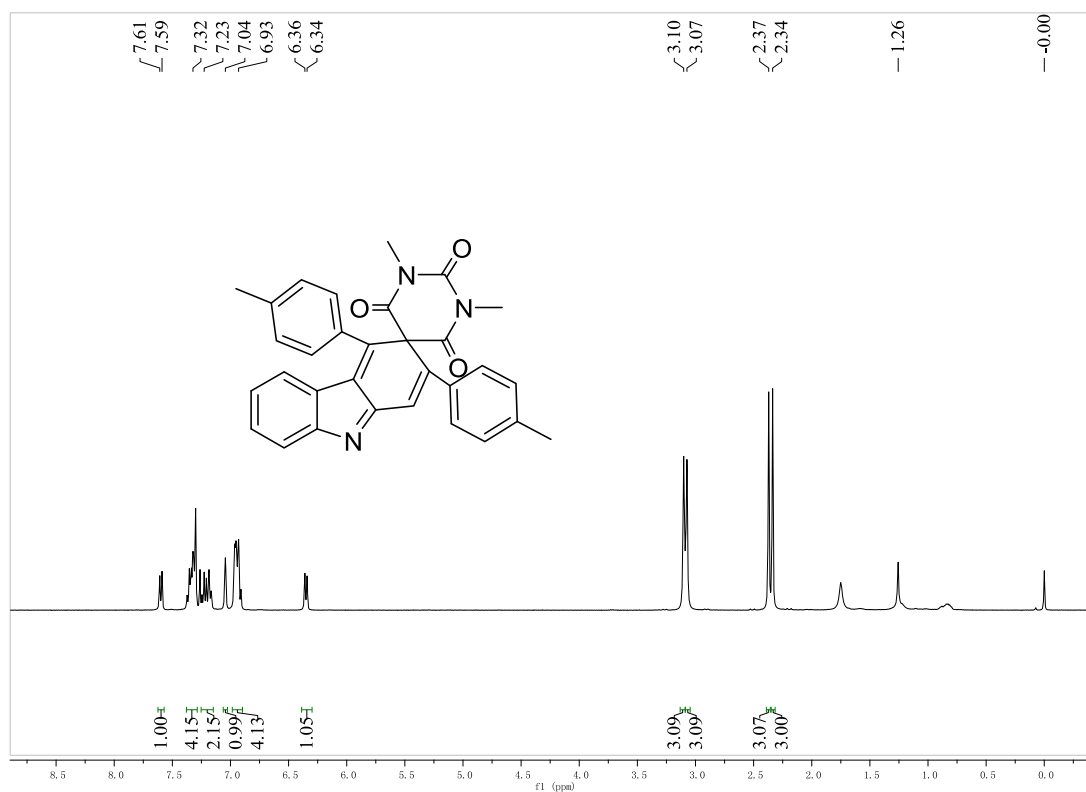


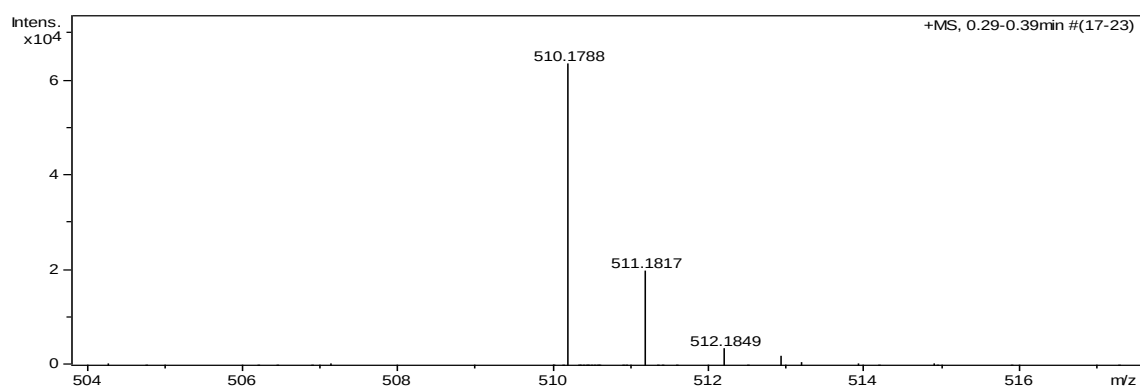
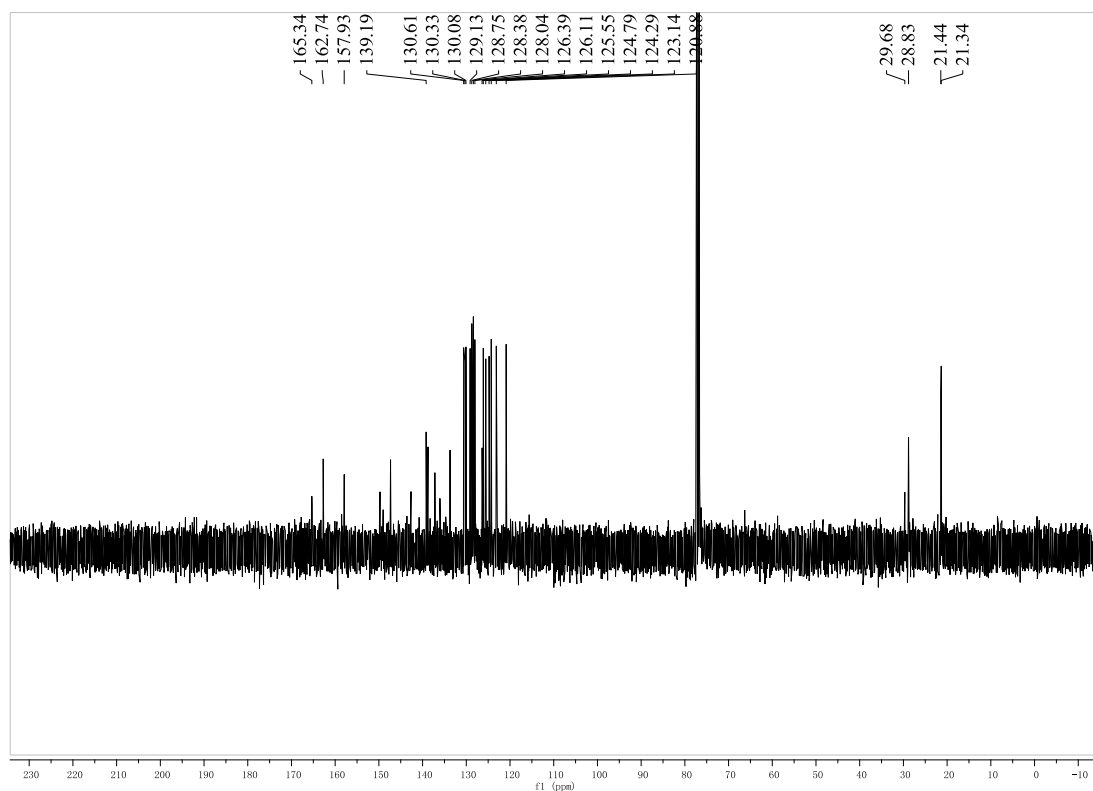


1',3'-Dimethyl-2,4-di-*p*-tolyl-2'*H*-spiro[carbazole-3,5'-pyrimidine]-2',4',6'(1'*H*,3'*H*)-trione

(3a):

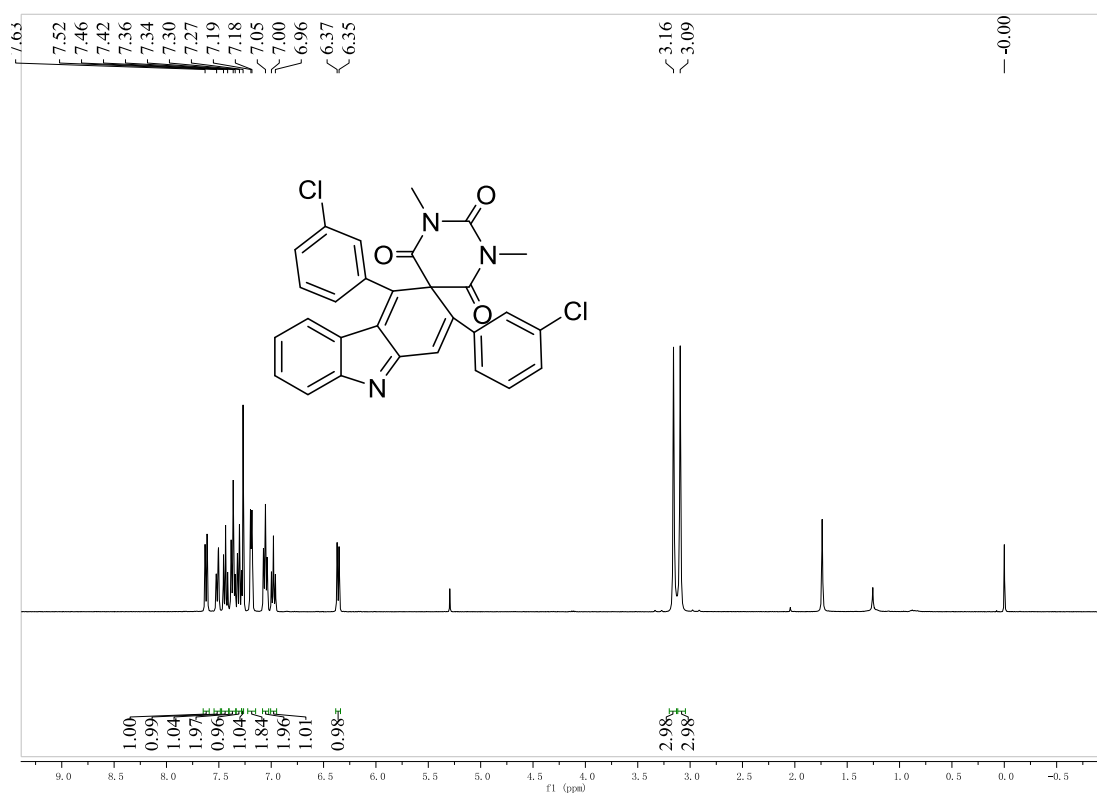
yellow solid, 75%, m.p. 190-192 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.59 (d, $J = 7.6$ Hz, 1H, ArH), 7.35-7.30 (m, 4H, ArH), 7.25-7.17 (m, 2H, ArH), 7.04 (s, 1H, ArH), 6.96-6.91 (m, 4H, ArH), 6.35 (d, $J = 7.6$ Hz, 1H, CH), 3.10 (s, 3H, CH_3), 3.07 (s, 3H, CH_3), 2.37 (s, 3H, CH_3), 2.34 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 165.3, 162.7, 157.9, 149.7, 149.0, 147.3, 142.6, 139.1, 138.7, 137.1, 136.0, 133.7, 130.6, 130.3, 130.0, 129.1, 128.7, 128.3, 128.0, 126.3, 126.1, 125.5, 124.7, 124.2, 123.1, 120.8, 29.6, 28.8, 21.4, 21.3; IR (KBr) ν : 3219, 3158, 3043, 2966, 2900, 1843, 1755, 1648, 1617, 1537, 1466, 1358, 1318, 1266, 1150, 987, 899, 765 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{31}\text{H}_{25}\text{N}_3\text{O}_3$ ($[\text{M}+\text{Na}]^+$): 510.1788, found: 510.1788.

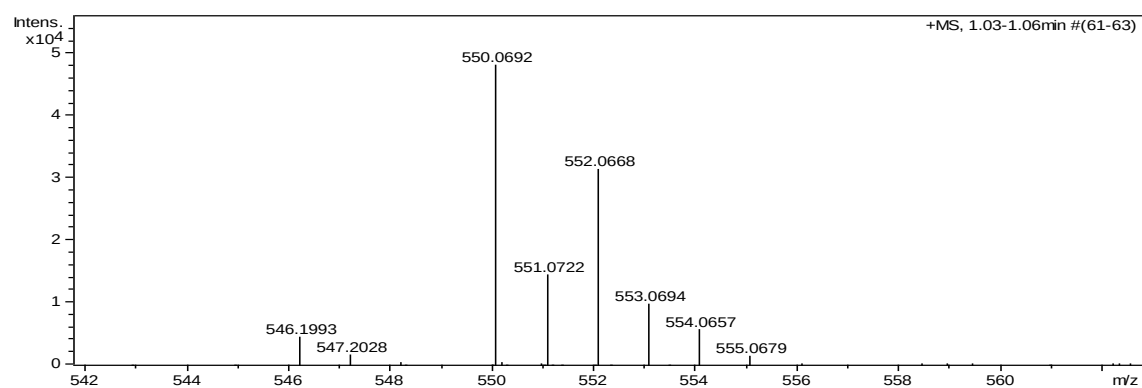
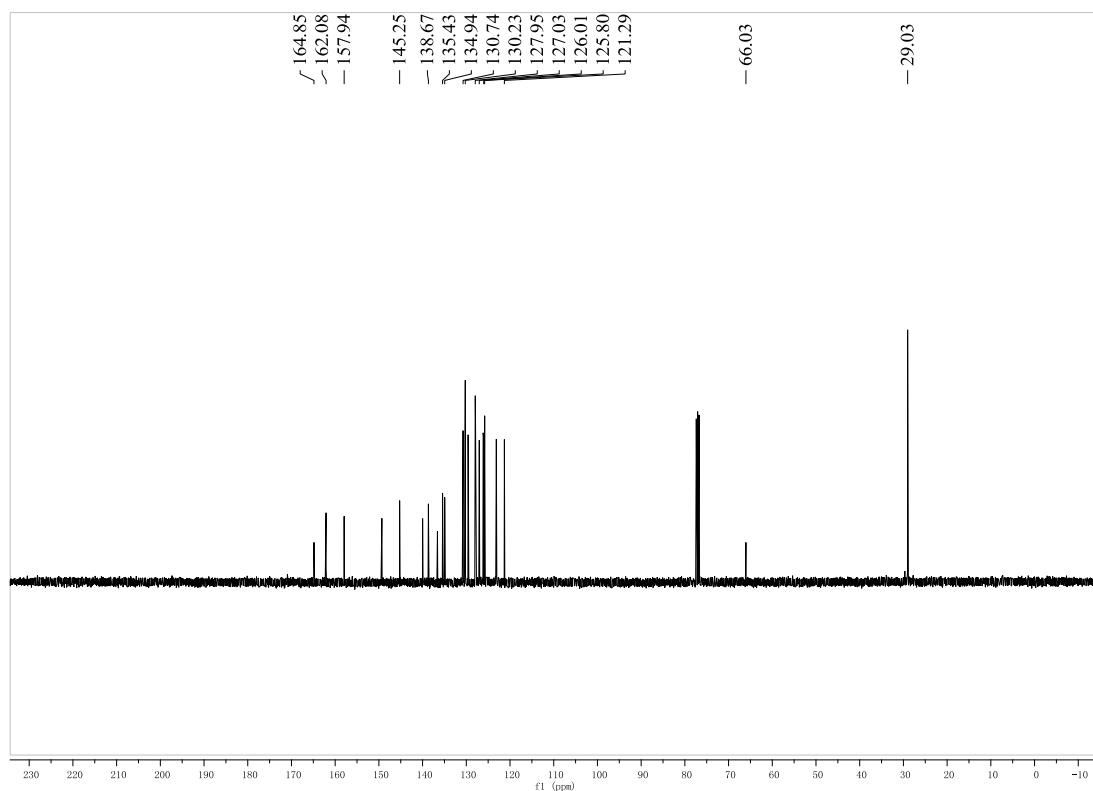




2,4-Bis(3-chlorophenyl)-1',3'-dimethyl-2'H-spiro[carbazole-3,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (3b):

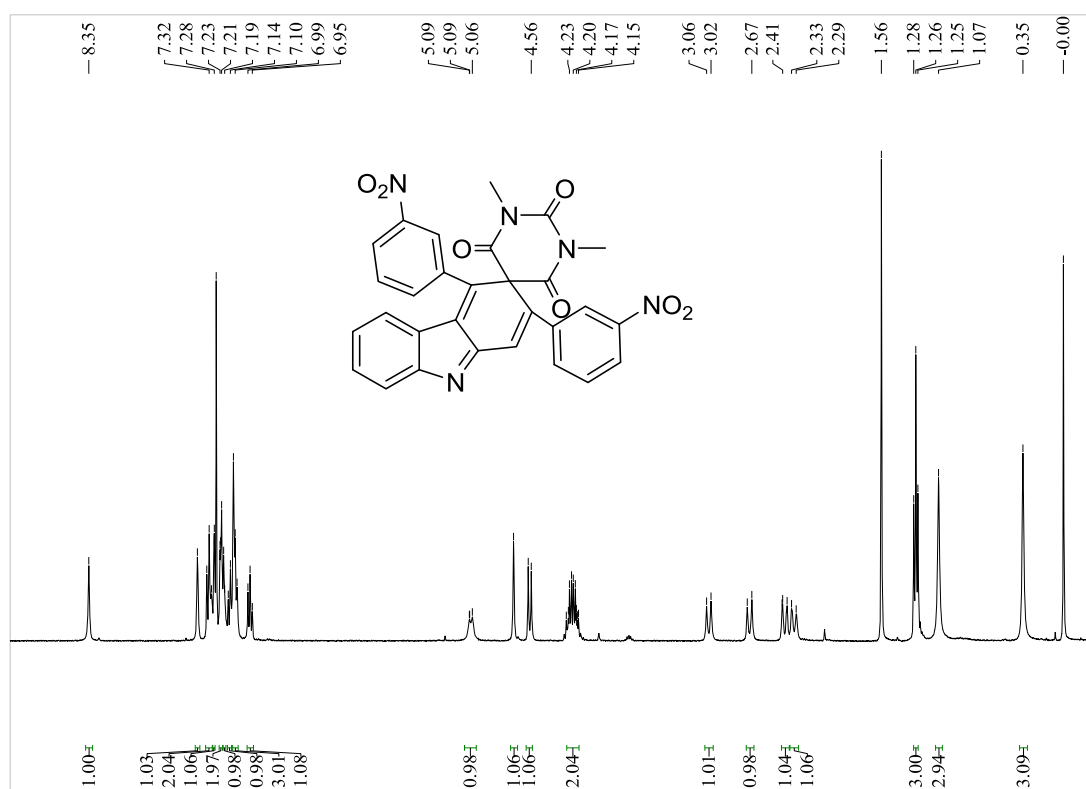
yellow solid, 67%, m.p. 194-197 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.62 (d, $J = 8.0$ Hz, 1H, ArH), 7.53-7.50 (m, 1H, ArH), 7.44 (t, $J = 8.0$ Hz, 1H, ArH), 7.36 (t, $J = 8.0$ Hz, 2H, ArH), 7.30 (t, $J = 8.0$ Hz, 1H, ArH), 7.28-7.27 (m, 1H, ArH), 7.20-7.18 (m, 2H, ArH), 7.06 (t, $J = 7.6$ Hz, 2H, ArH), 6.98 (t, $J = 7.6$ Hz, 1H, ArH), 6.36 (d, $J = 8.0$ Hz, 1H, CH), 3.16 (s, 3H, CH_3), 3.09 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 164.8, 162.0, 157.9, 149.3, 145.2, 139.9, 138.6, 136.6, 135.4, 135.3, 134.9, 130.8, 130.7, 130.2, 130.2, 129.5, 127.9, 127.8, 127.0, 126.1, 126.0, 125.8, 125.7, 123.1, 121.2, 66.0, 29.0; IR (KBr) ν : 3255, 3175, 3048, 2978, 1876, 1756, 1667, 1601, 1556, 1478, 1362, 1258, 1217, 1180, 964, 832, 745 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{29}\text{H}_{19}\text{Cl}_2^{35}\text{N}_3\text{O}_3$ ($[\text{M}+\text{Na}]^+$): 550.0696, found: 550.0692. $\text{C}_{29}\text{H}_{19}\text{Cl}_2^{37}\text{N}_3\text{O}_3$ ($[\text{M}+\text{Na}]^+$): 552.0666, found: 552.0668.

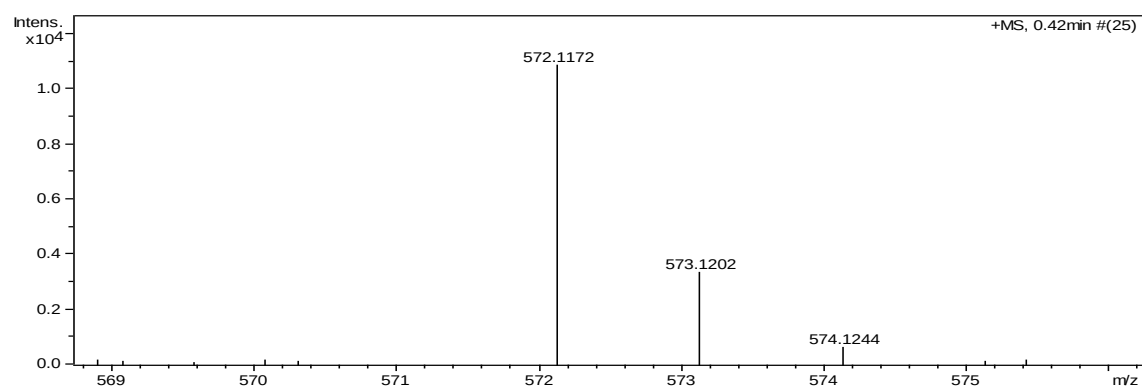
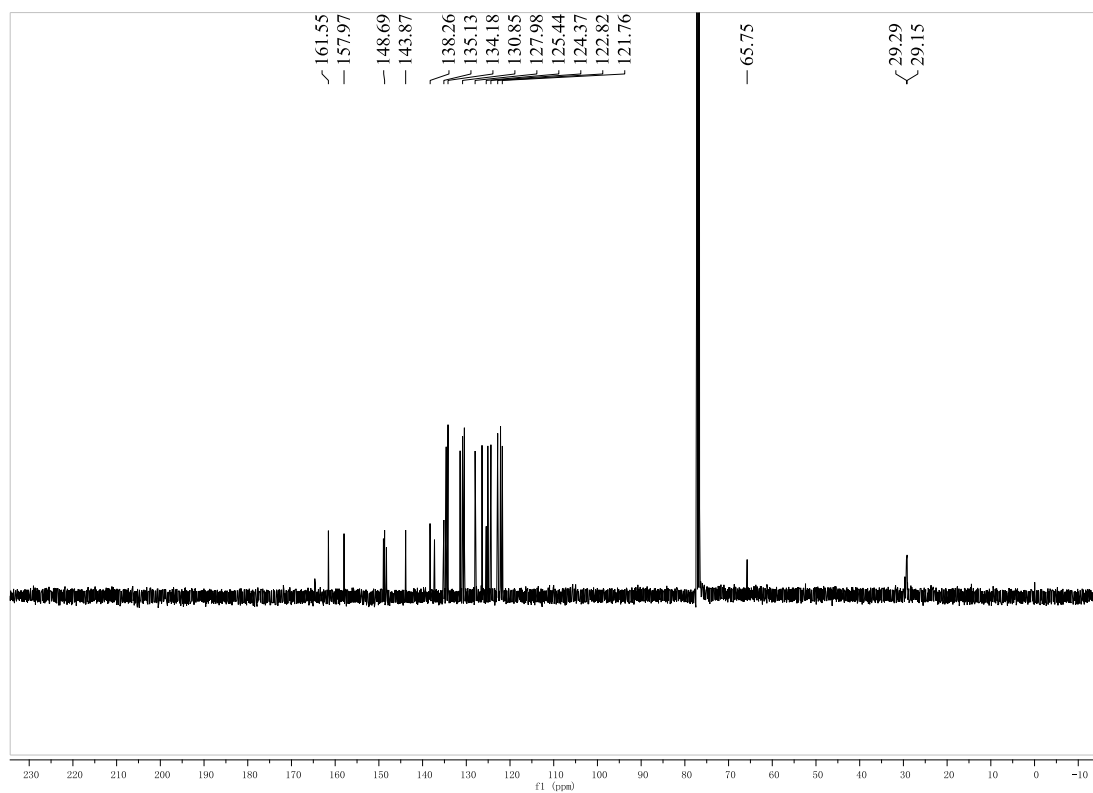




1',3'-Dimethyl-2,4-bis(3-nitrophenyl)-2'H-spiro[carbazole-3,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (3c):

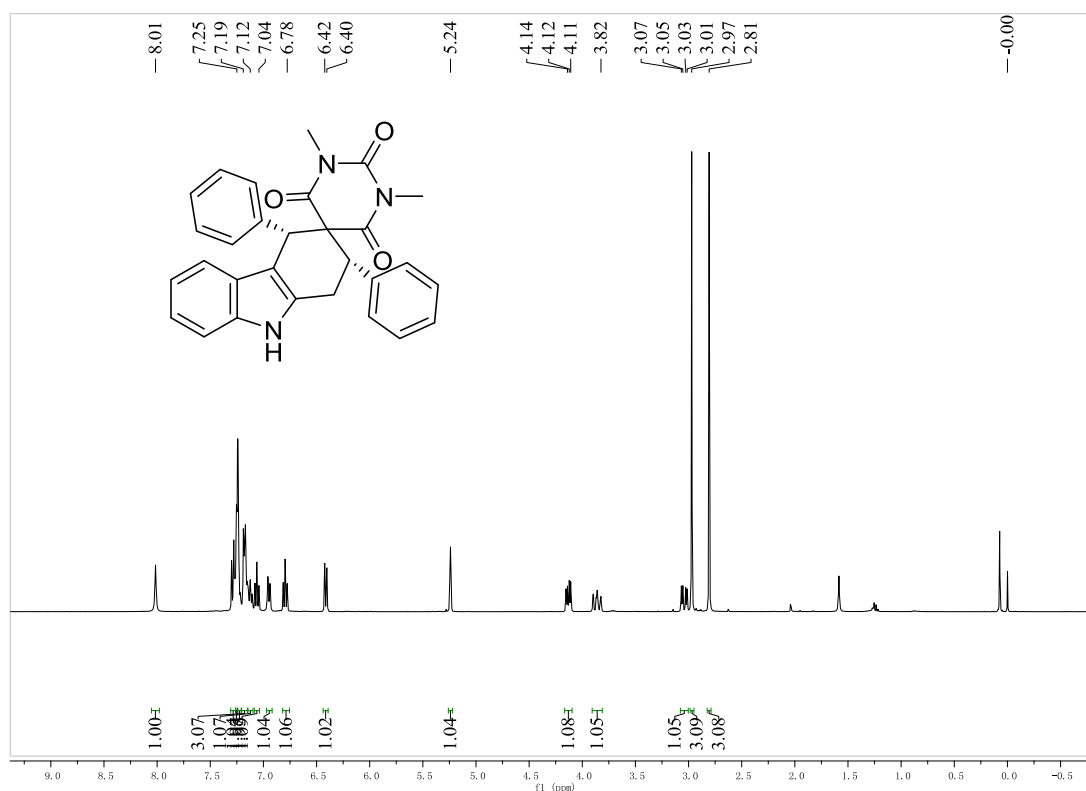
yellow solid, 58%, m.p. 200-203 °C; ¹H NMR (400 MHz, CDCl₃) δ: 8.42 (d, *J* = 8.0 Hz, 1H, ArH), 8.27 (d, *J* = 8.0 Hz, 1H, ArH), 8.07-8.05 (m, 2H, ArH), 7.75 (t, *J* = 8.0 Hz, 1H, ArH), 7.66-7.56 (m, 4H, ArH), 7.42-7.37 (m, 2H, ArH), 6.97 (t, *J* = 8.0 Hz, 1H, ArH), 6.23 (d, *J* = 7.2 Hz, 1H, CH), 3.19 (s, 3H, CH₃), 3.08 (s, 3H, CH₃); ¹³C NMR (400 MHz, CDCl₃) δ: 161.5, 157.9, 148.9, 148.6, 148.2, 143.8, 138.3, 138.2, 137.2, 135.1, 134.6, 134.1, 131.4, 130.8, 130.4, 127.9, 126.4, 125.4, 125.0, 124.3, 122.9, 122.8, 122.1, 121.7, 65.7, 29.2, 29.1; IR (KBr) ν: 3217, 3167, 3005, 2967, 1945, 1832, 1746, 1676, 1523, 1411, 1348, 1234, 1221, 1167, 969, 848, 772 cm⁻¹; MS (*m/z*): HRMS (ESI) Calcd. for C₂₉H₁₉N₅O₇ ([M+Na]⁺): 572.1177, found: 572.1172.

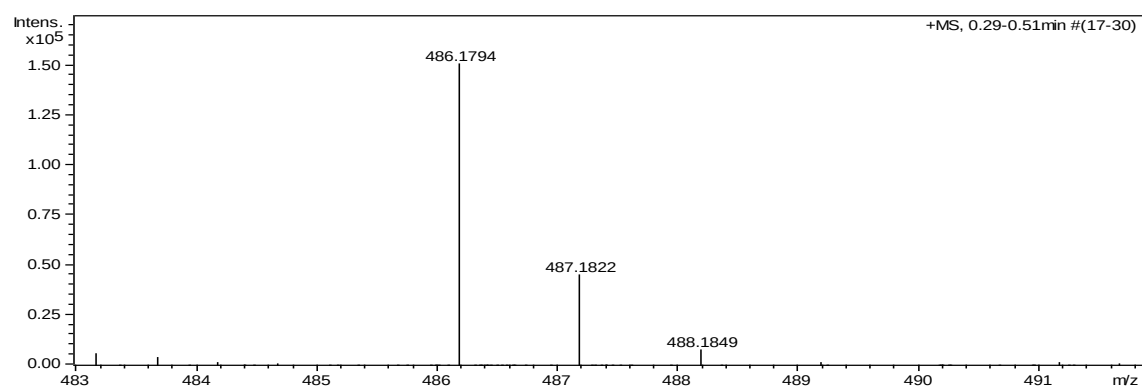
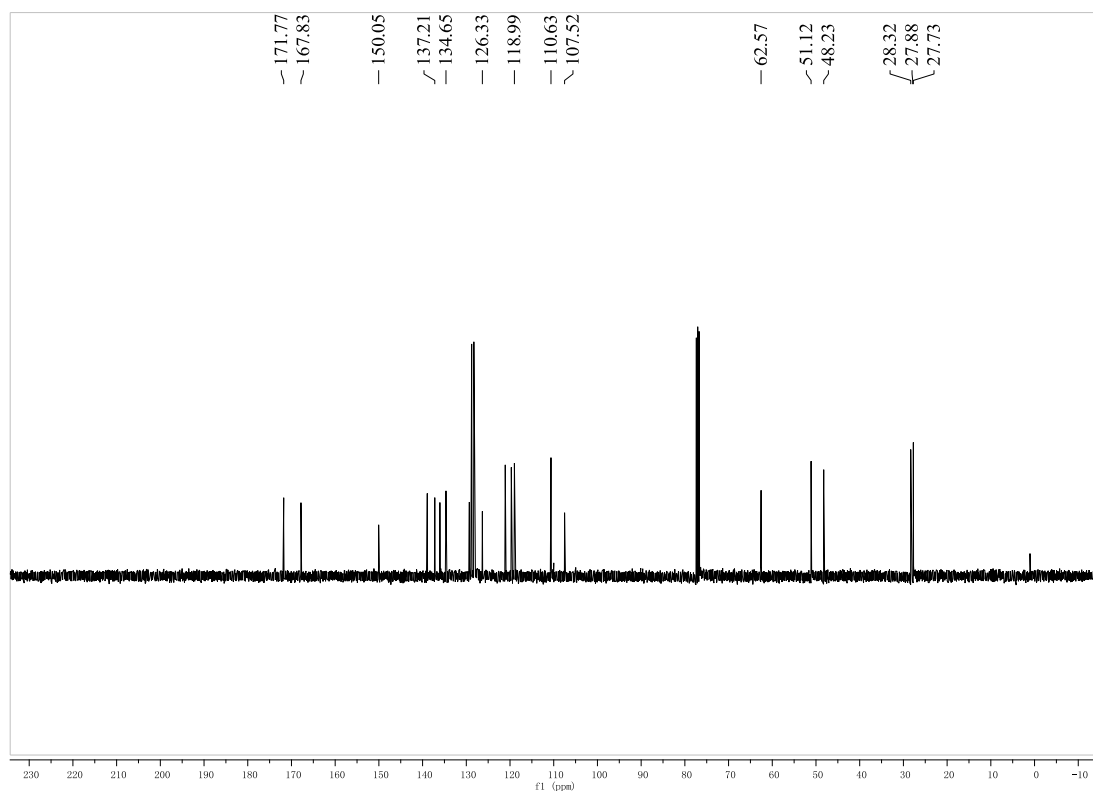




1',3'-Dimethyl-2,4-diphenyl-1,2,4,9-tetrahydro-2'H-spiro[carbazole-3,5'-pyrimidine]-2',4',6'-(1'H,3'H)-trione (4a):

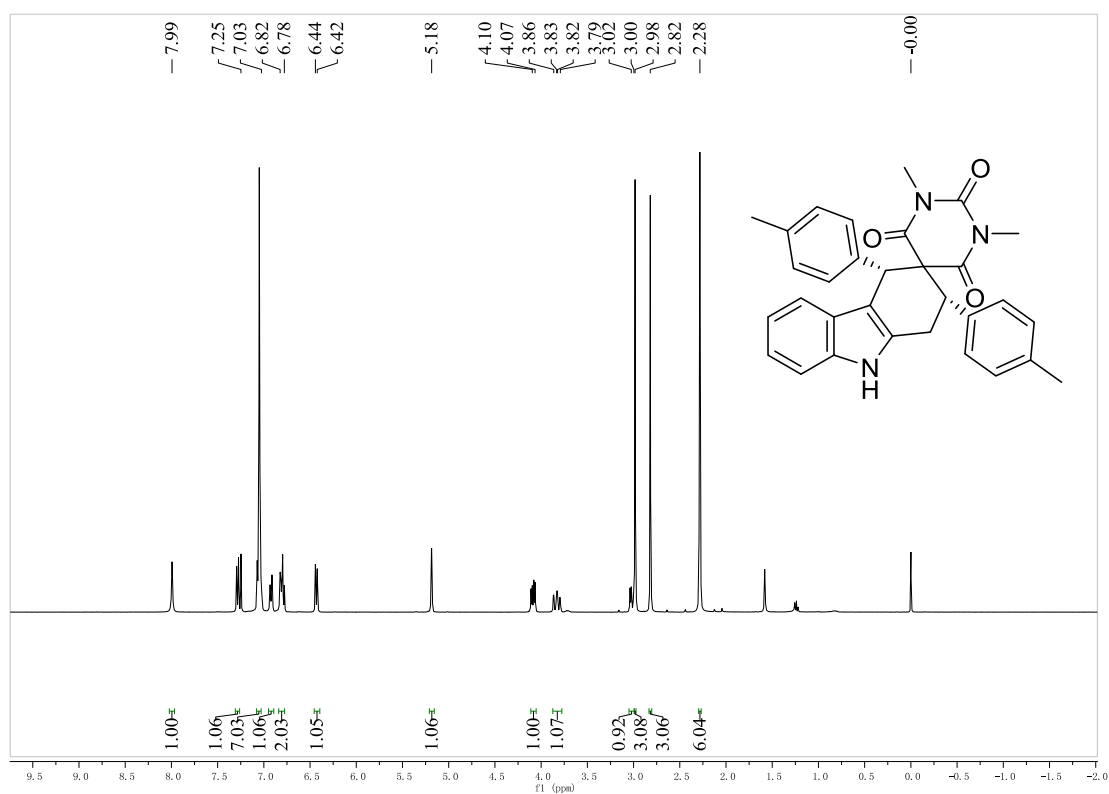
purple solid, 82%, m.p. 205-208 °C; ¹H NMR (400 MHz, CDCl₃) δ: 8.02 (s, 1H, NH), 7.30-7.27 (m, 2H, ArH), 7.25-7.24 (m, 2H, ArH), 7.24-7.22 (m, 2H, ArH), 7.19-7.15 (m, 3H, ArH), 7.15-7.10 (m, 1H, ArH), 7.06 (t, *J* = 7.2 Hz, 1H, ArH), 6.94 (d, *J* = 7.2 Hz, 1H, ArH), 6.80 (t, *J* = 8.0 Hz, 1H, ArH), 6.41 (d, *J* = 8.0 Hz, 1H, ArH), 5.24 (s, 1H, CH), 4.13 (dd, *J*₁ = 12.0 Hz, *J*₂ = 5.6 Hz, 1H, CH), 3.90-3.82 (m, 1H, CH), 3.05 (dd, *J*₁ = 16.4 Hz, *J*₂ = 5.6 Hz, 1H, CH), 2.97 (s, 3H, CH₃), 2.81 (s, 3H, CH₃); ¹³C NMR (400 MHz, CDCl₃) δ: 171.7, 167.8, 150.0, 138.9, 137.2, 136.0, 134.6, 129.3, 128.9, 128.7, 128.2, 128.2, 128.1, 128.1, 128.1, 126.3, 121.0, 119.7, 118.9, 110.6, 107.5, 62.5, 51.1, 48.2, 28.3, 27.8, 27.7; IR (KBr) ν: 3407, 3078, 2981, 1873, 1744, 1658, 1667, 1582, 1466, 1356, 1321, 1221, 1180, 912, 833 cm⁻¹; MS (*m/z*): HRMS (ESI) Calcd. for C₂₉H₂₅N₃O₃ ([M+Na]⁺): 486.1788, found: 486.1794.

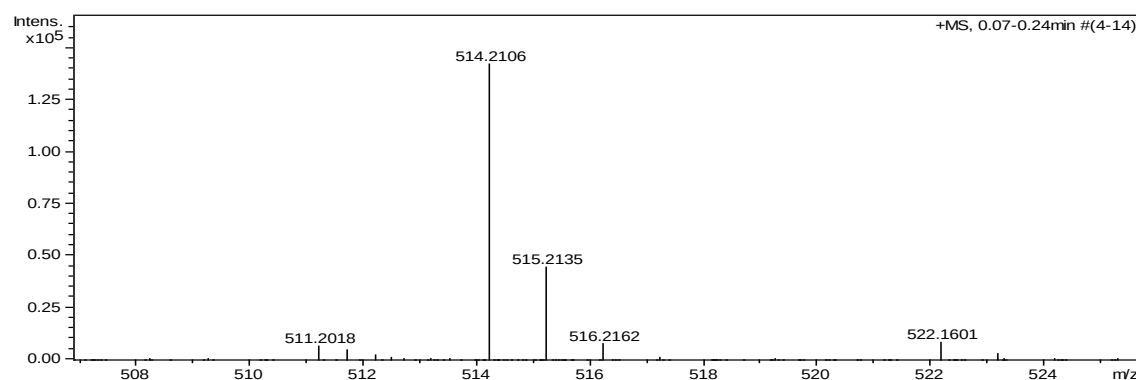
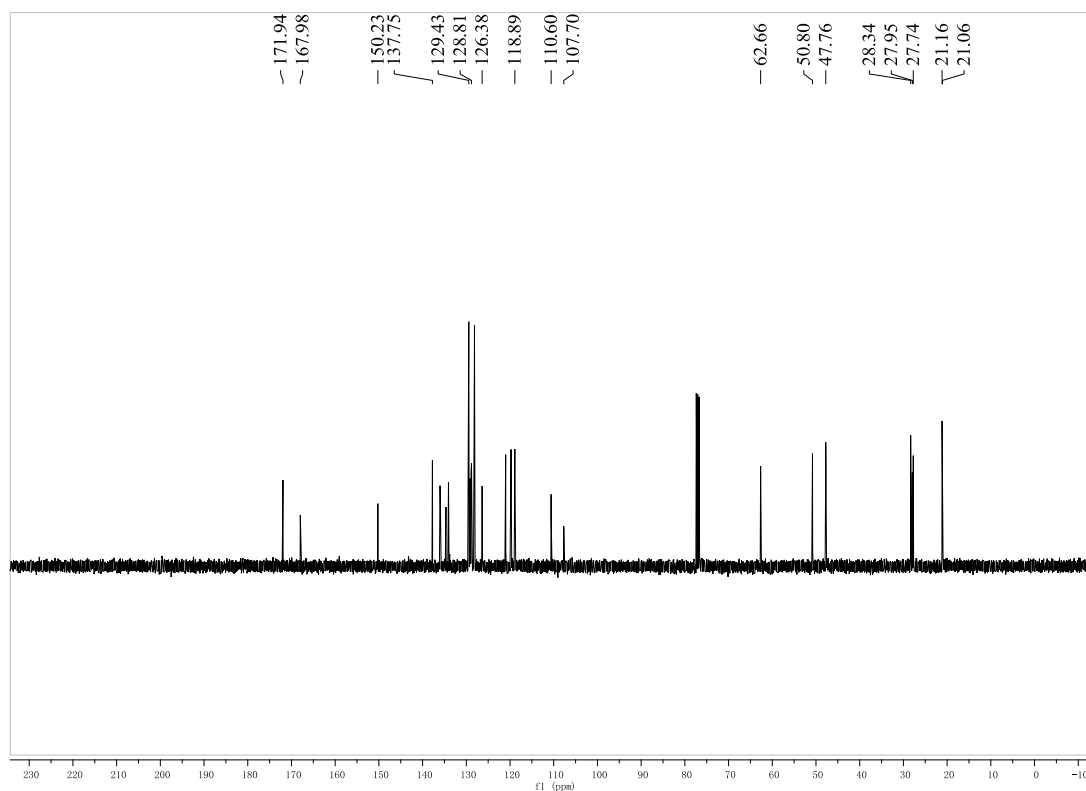




1',3'-Dimethyl-2,4-di-*p*-tolyl-1,2,4,9-tetrahydro-2'*H*-spiro[carbazole-3,5'-pyrimidine]-2',4',6'-(1'*H*,3'*H*)-trione (4b):

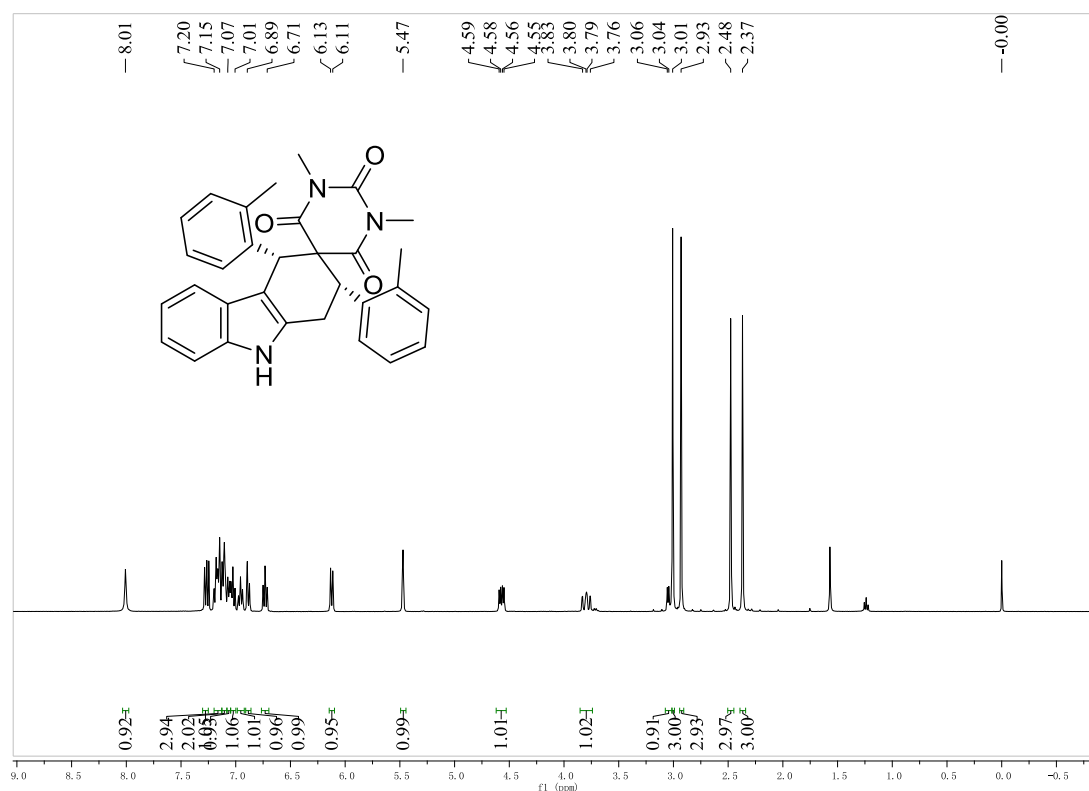
purple solid, 80%, m.p. 206-209 °C; ¹H NMR (400 MHz, CDCl₃) δ: 7.99 (s, 1H, NH), 7.28 (d, *J* = 8.4 Hz, 1H, ArH), 7.07-7.03 (m, 7H, ArH), 6.92 (d, *J* = 8.0 Hz, 1H, ArH), 6.82-6.78 (m, 2H, ArH), 6.43 (d, *J* = 8.0 Hz, 1H, ArH), 5.19 (s, 1H, CH), 4.09 (dd, *J*₁ = 12.0 Hz, *J*₂ = 5.6 Hz, 1H, CH), 3.87-3.79 (m, 1H, CH), 3.00 (dd, *J*₁ = 16.8 Hz, *J*₂ = 5.6 Hz, 1H, CH), 2.98 (s, 3H, CH₃), 2.82 (s, 3H, CH₃), 2.29 (s, 3H, CH₃), 2.28 (s, 3H, CH₃); ¹³C NMR (400 MHz, CDCl₃) δ: 171.9, 167.9, 150.2, 137.7, 137.6, 136.0, 135.9, 134.6, 134.0, 129.4, 129.0, 128.9, 128.8, 128.7, 128.1, 126.3, 120.9, 119.7, 118.8, 110.5, 107.7, 62.6, 50.8, 47.7, 28.3, 27.9, 27.7, 21.1, 21.0; IR (KBr) ν: 3464, 3087, 2967, 1866, 1753, 1662, 1631, 1583, 1462, 1355, 1317, 1280, 1175, 961, 870 cm⁻¹; MS (*m/z*): HRMS (ESI) Calcd. for C₃₁H₂₉N₃O₃ ([M+Na]⁺): 514.2101, found: 514.2106.

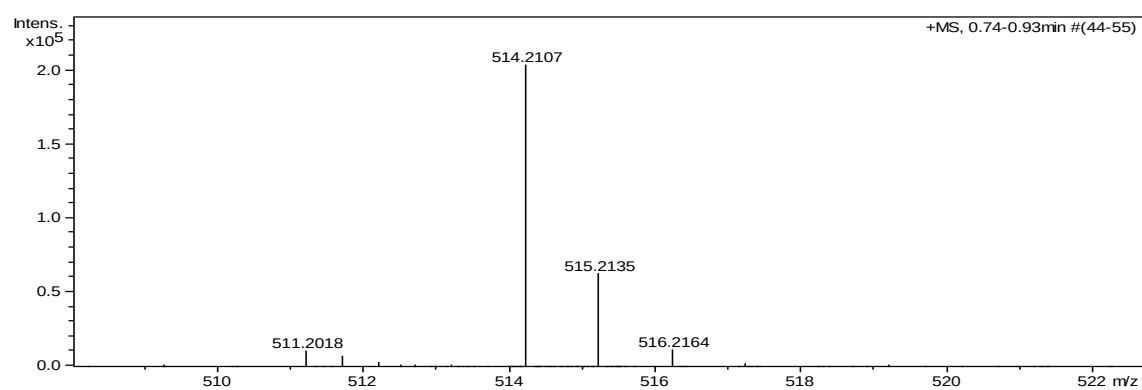
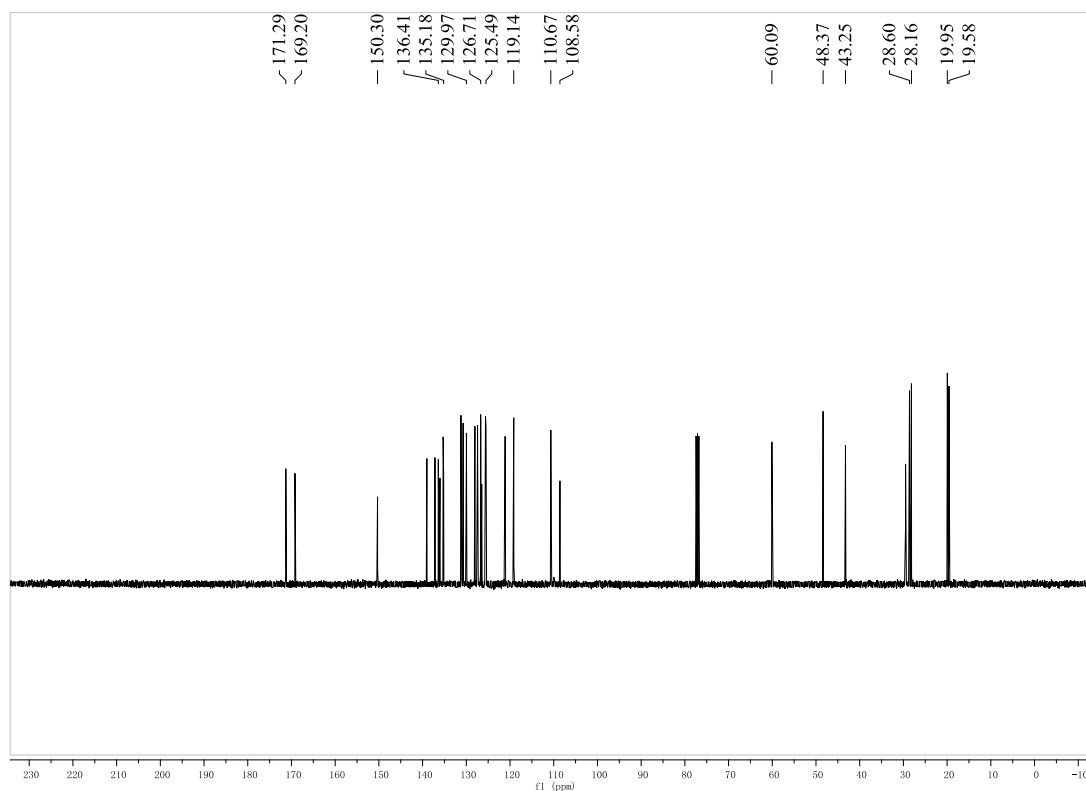




1',3'-Dimethyl-2,4-di-*o*-tolyl-1,2,4,9-tetrahydro-2'*H*-spiro[carbazole-3,5'-pyrimidine]-2',4',6'-(1'*H*,3'*H*)-trione (4c):

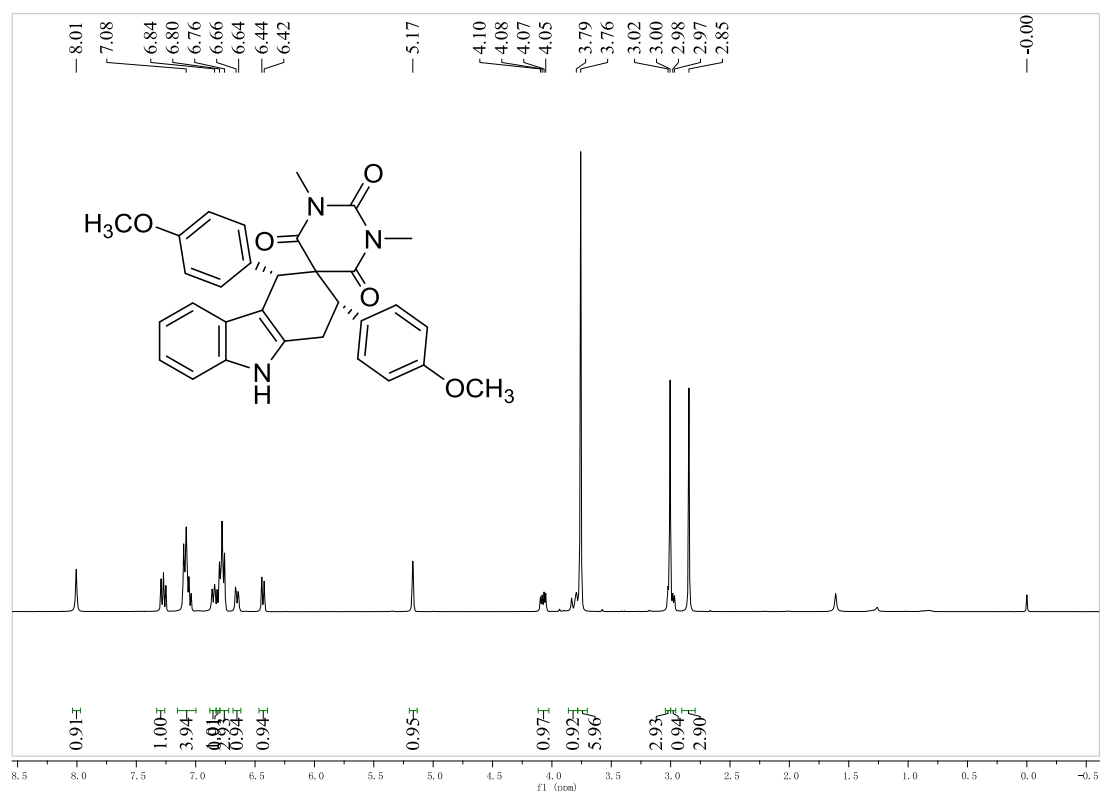
purple solid, 75%, m.p. 212-214 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.01 (s, 1H, NH), 7.27 (d, J = 8.4 Hz, 1H, ArH), 7.20-7.13 (m, 3H, ArH), 7.13-7.09 (m, 2H, ArH), 7.06 (d, J = 7.2 Hz, 1H, ArH), 7.03 (t, J = 7.2 Hz, 1H, ArH), 6.96 (t, J = 7.2 Hz, 1H, ArH), 6.88 (d, J = 7.2 Hz, 1H, ArH), 6.73 (t, J = 7.2 Hz, 1H, ArH), 6.12 (d, J = 8.0 Hz, 1H, ArH), 5.47 (s, 1H, CH), 4.57 (dd, J_1 = 12.0 Hz, J_2 = 5.6 Hz, 1H, CH), 3.84-3.76 (m, 1H, CH), 3.04 (d, J = 5.6 Hz, 1H, CH), 3.01 (s, 3H, CH_3), 2.93 (s, 3H, CH_3), 2.48 (s, 3H, CH_3), 2.37 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 171.2, 169.1, 150.3, 139.0, 137.1, 136.4, 136.0, 135.2, 135.1, 131.2, 130.7, 129.9, 128.0, 127.4, 126.7, 126.4, 125.6, 125.4, 121.1, 119.1, 119.1, 110.6, 108.5, 60.0, 48.3, 43.2, 29.5, 28.6, 28.1, 19.9, 19.5; IR (KBr) ν : 3466, 3010, 2953, 1937, 1841, 1678, 1655, 1572, 1469, 1361, 1310, 1255, 1169, 987, 863 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{31}\text{H}_{29}\text{N}_3\text{O}_3$ ($[\text{M}+\text{Na}]^+$): 514.2101, found: 514.2107.

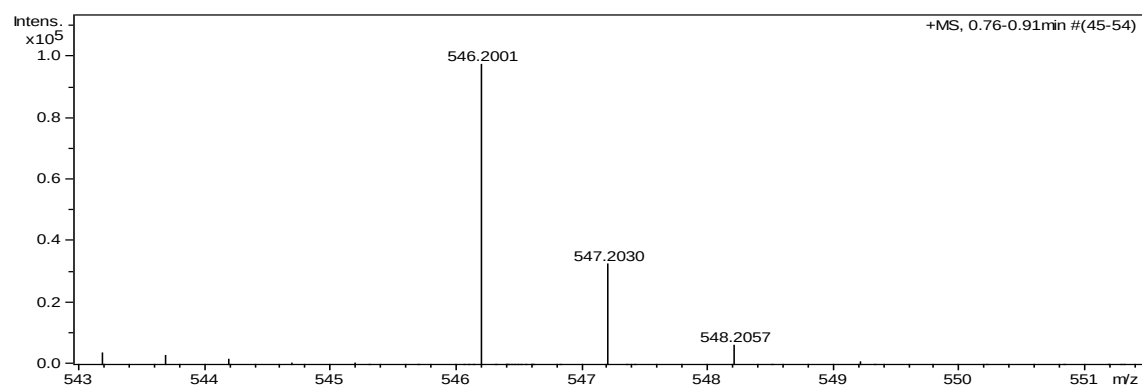
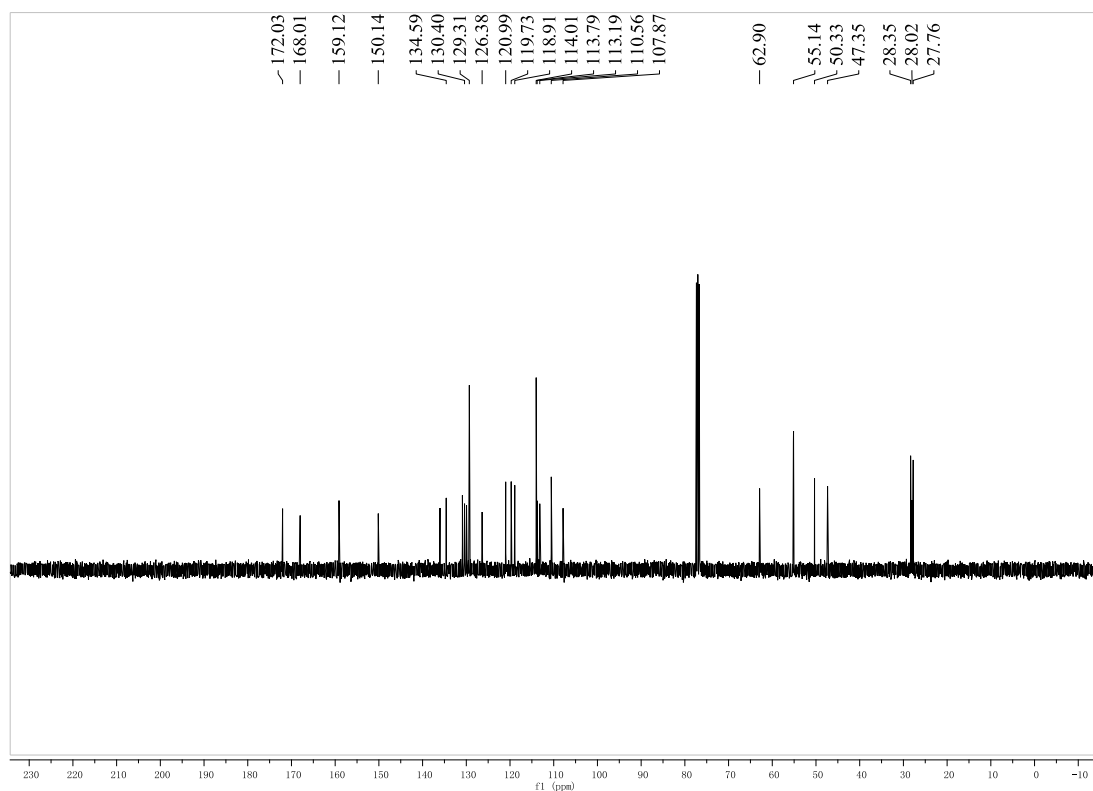




2,4-Bis(4-methoxyphenyl)-1',3'-dimethyl-1,2,4,9-tetrahydro-2'H-spiro[carbazole-3,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (4d):

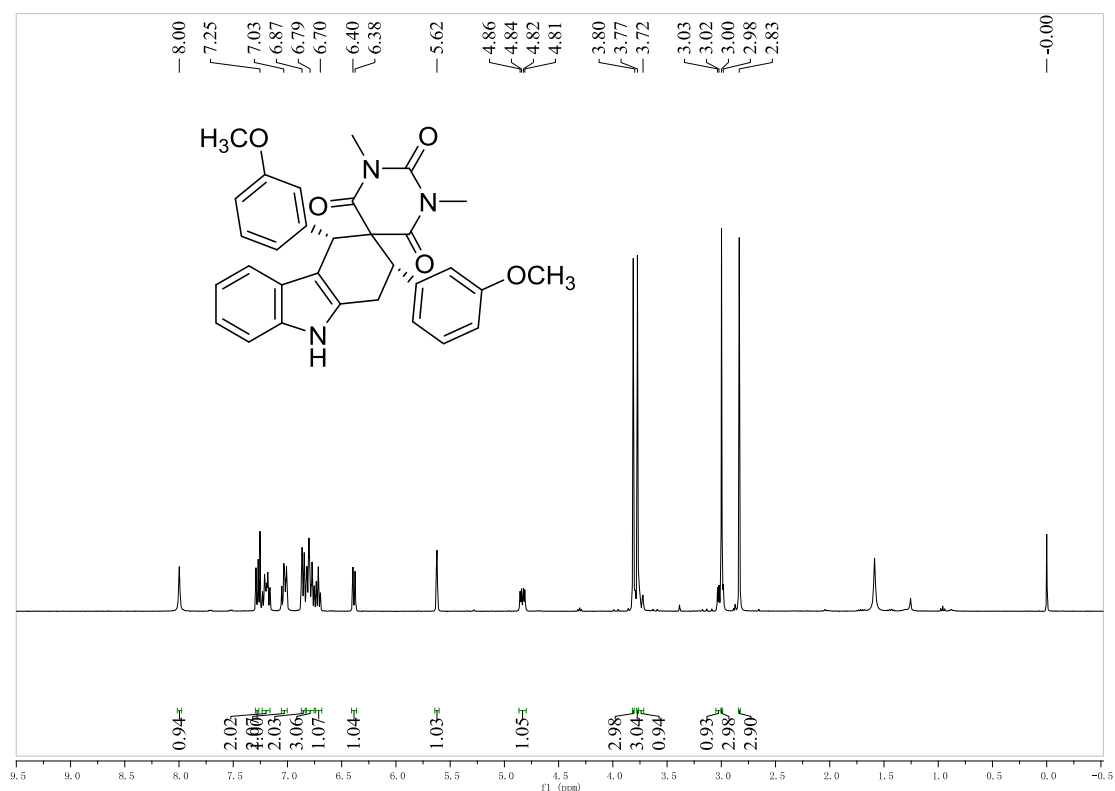
purple solid, 78%, m.p. 213-215 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.01 (s, 1H, NH), 7.28 (d, J = 8.0 Hz, 1H, ArH), 7.10-7.04 (m, 4H, ArH), 6.85 (d, J = 8.8 Hz, 1H, ArH), 6.81 (d, J = 8.0 Hz, 1H, ArH), 6.78-6.76 (m, 3H, ArH), 6.45 (dd, J_1 = 8.8 Hz, J_2 = 2.0 Hz, 1H, ArH), 6.43 (d, J = 8.0 Hz, 1H, ArH), 5.17 (s, 1H, CH), 4.07 (dd, J_1 = 12.0 Hz, J_2 = 5.6 Hz, 1H, CH), 3.81 (d, J = 15.6 Hz, 1H, CH), 3.76 (s, 3H, OCH_3), 3.75 (s, 3H, OCH_3), 3.05 (s, 3H, CH_3), 2.97 (d, J = 5.6 Hz, 1H, CH), 2.85 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 172.0, 168.0, 159.1, 159.1, 150.1, 136.0, 134.5, 130.8, 130.4, 129.9, 129.3, 129.1, 126.3, 120.9, 119.7, 118.9, 114.0, 113.7, 113.1, 110.5, 107.8, 62.9, 55.1, 55.1, 50.3, 47.3, 28.3, 28.0, 27.7; IR (KBr) ν : 3439, 3078, 2988, 1967, 1854, 1676, 1631, 1542, 1453, 1361, 1349, 1255, 1167, 987, 869 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{31}\text{H}_{29}\text{N}_3\text{O}_5$ ($[\text{M}+\text{Na}]^+$): 546.1999, found: 546.2001.

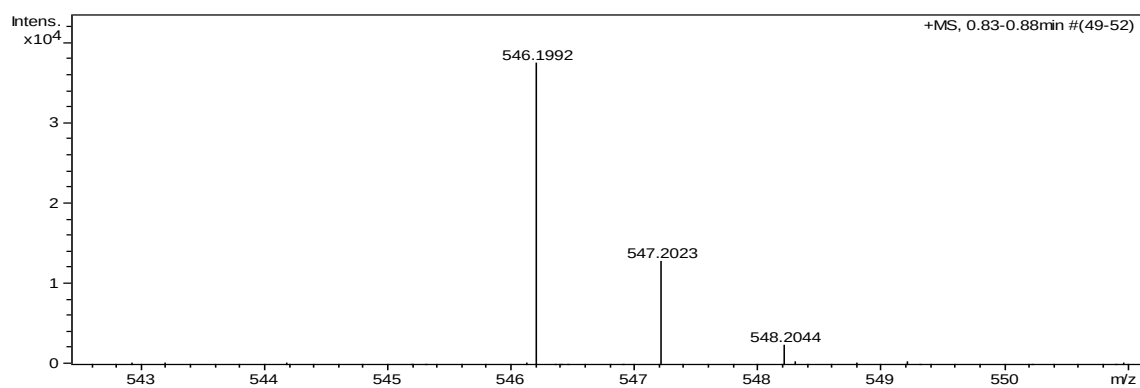
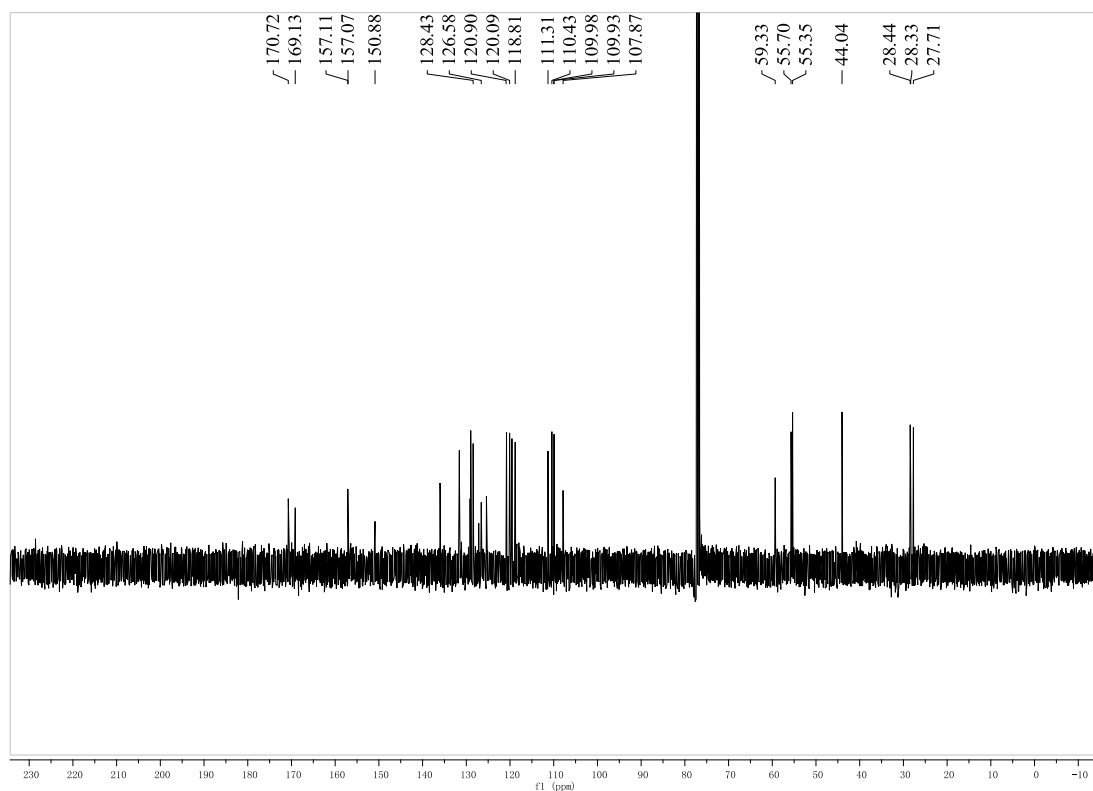




2,4-Bis(3-methoxyphenyl)-1',3'-dimethyl-1,2,4,9-tetrahydro-2'H-spiro[carbazole-3,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (4e):

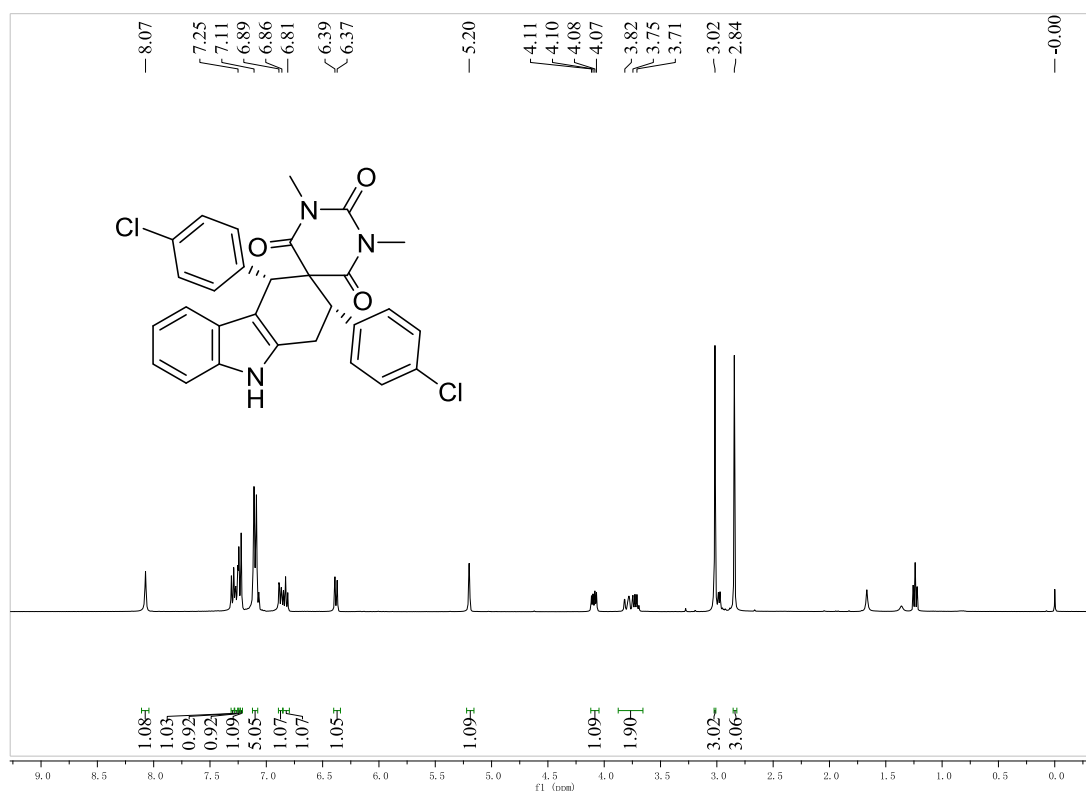
purple solid, 70%, m.p. 215-217 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.00 (s, 1H, NH), 7.28 (d, $J = 8.4$ Hz, 1H, ArH), 7.23-7.16 (m, 2H, ArH), 7.05-7.01 (m, 2H, ArH), 6.85 (d, $J = 8.4$ Hz, 2H, ArH), 6.82-6.75 (m, 3H, ArH), 6.72 (t, $J = 7.6$ Hz, 1H, ArH), 6.38 (d, $J = 8.0$ Hz, 1H, ArH), 5.62 (s, 1H, CH), 4.83 (dd, $J_1 = 12.4$ Hz, $J_2 = 5.6$ Hz, 1H, CH), 3.81 (s, 3H, OCH_3), 3.77 (s, 3H, OCH_3), 3.77-3.72 (m, 1H, CH), 3.04-2.99 (m, 1H, CH), 2.99 (s, 3H, CH_3), 2.84 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 170.7, 169.1, 157.1, 157.0, 150.8, 136.0, 135.9, 131.5, 129.1, 128.9, 128.4, 127.1, 126.5, 125.3, 120.8, 120.8, 120.0, 119.5, 118.8, 111.3, 110.4, 109.9, 109.9, 107.8, 59.3, 55.7, 55.3, 44.0, 28.4, 28.3, 27.7; IR (KBr) ν : 3417, 3083, 2990, 1871, 1743, 1645, 1631, 1581, 1467, 1355, 1371, 1283, 1145, 879, 786 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{31}\text{H}_{29}\text{N}_3\text{O}_5$ ($[\text{M}+\text{Na}]^+$): 546.1999, found: 546.1992.

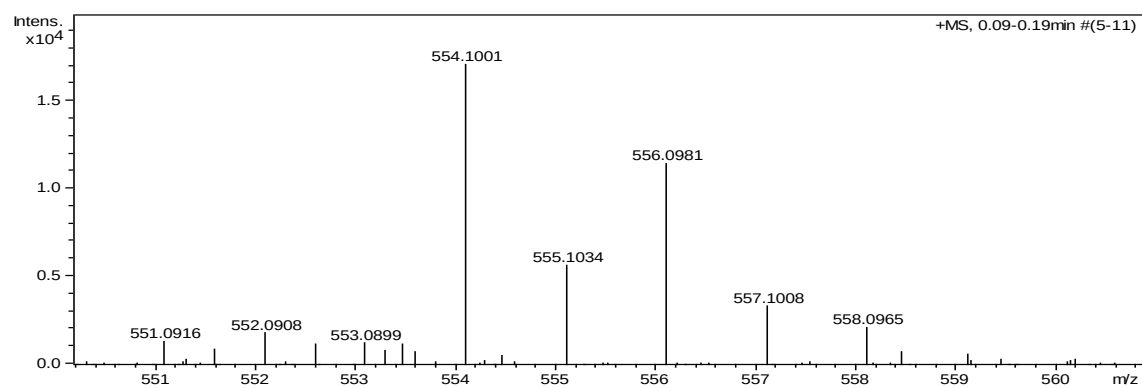
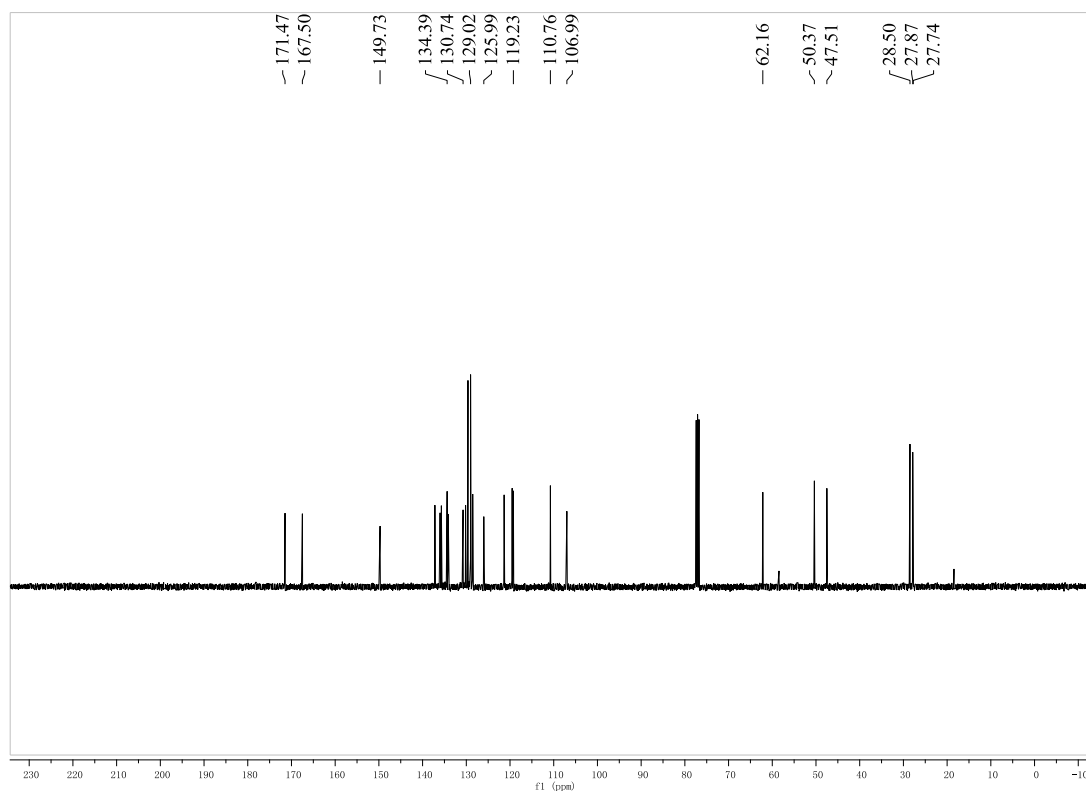




2,4-Bis(4-chlorophenyl)-1',3'-dimethyl-1,2,4,9-tetrahydro-2'H-spiro[carbazole-3,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (4f):

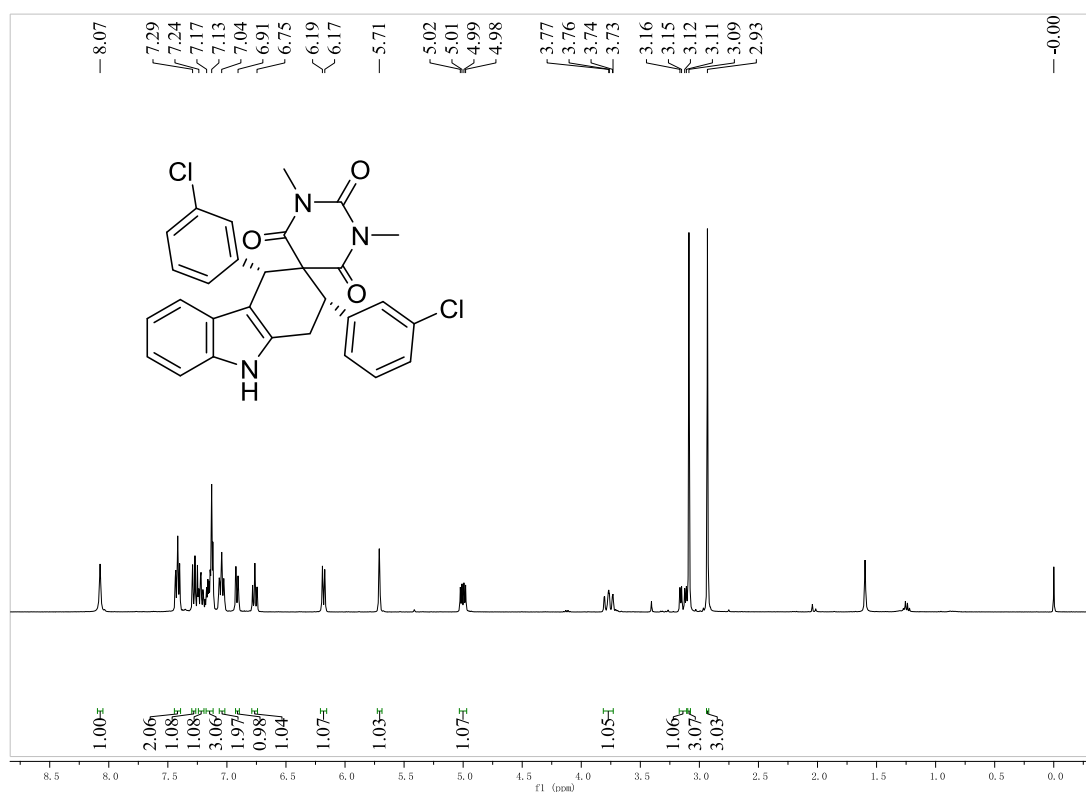
purple solid, 75%, m.p. 203-205 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.07 (s, 1H, NH), 7.30 (d, $J = 8.0$ Hz, 1H, ArH), 7.28-7.25 (m, 1H, ArH), 7.25-7.23 (m, 1H, ArH), 7.23-7.22 (m, 1H, ArH), 7.09 (d, $J = 8.4$ Hz, 5H, ArH), 6.87 (dd, $J_1 = 8.4$ Hz, $J_2 = 2.0$ Hz, 1H, ArH), 6.83 (t, $J = 7.6$ Hz, 1H, ArH), 6.38 (d, $J = 8.0$ Hz, 1H, ArH), 5.20 (s, 1H, CH), 4.09 (dd, $J_1 = 12.0$ Hz, $J_2 = 5.2$ Hz, 1H, CH), 3.82-3.71 (m, 2H, CH_2), 3.02 (s, 3H, CH_3), 2.85 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 171.4, 167.5, 149.7, 137.1, 136.0, 135.7, 134.3, 134.1, 134.0, 130.7, 130.1, 129.6, 129.0, 128.5, 128.4, 125.9, 121.3, 119.5, 119.2, 110.7, 106.9, 62.1, 50.3, 47.5, 28.5, 27.8, 27.7; IR (KBr) ν : 3304, 3113, 3081, 2967, 1746, 1655, 1613, 1569, 1482, 1366, 1312, 1276, 1158, 944, 863 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{29}\text{H}_{23}\text{Cl}_2^{35}\text{N}_3\text{O}_3$ ($[\text{M}+\text{Na}]^+$): 554.1009, found: 554.1001. $\text{C}_{29}\text{H}_{23}\text{Cl}_2^{37}\text{N}_3\text{O}_3$ ($[\text{M}+\text{Na}]^+$): 556.0979, found: 556.0981.

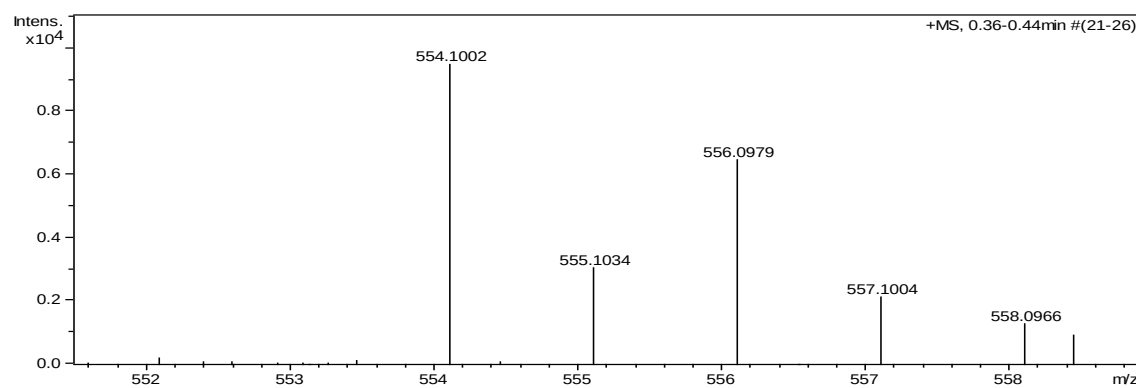
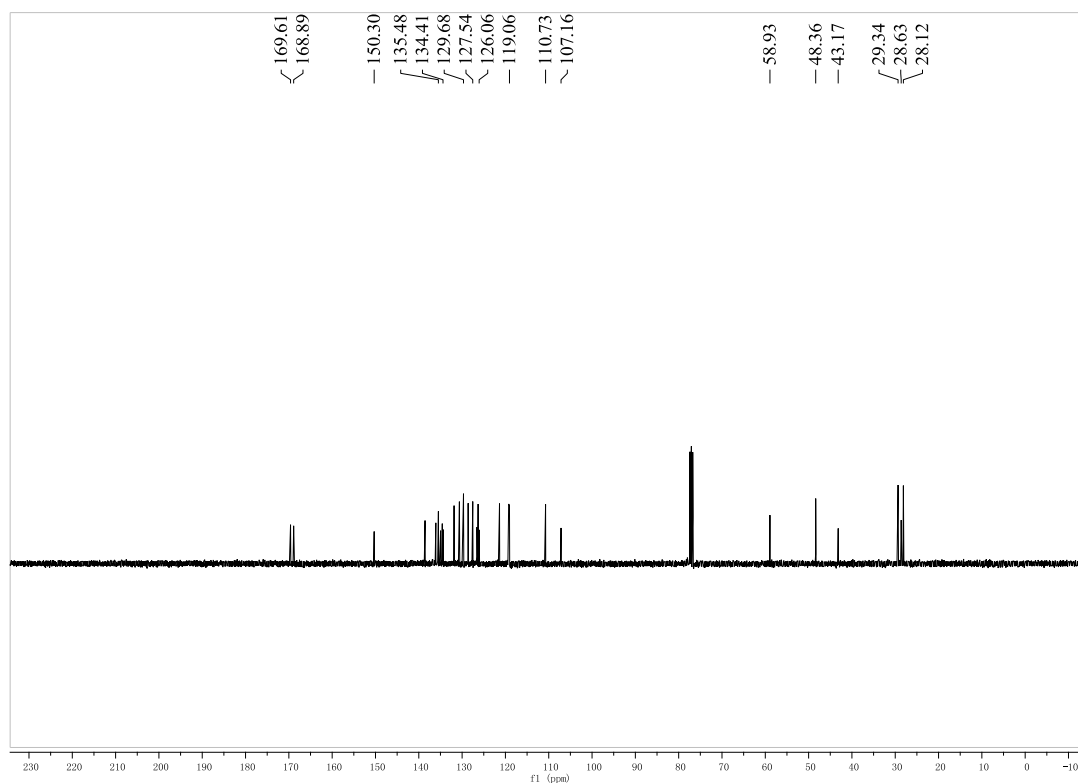




2,4-Bis(3-chlorophenyl)-1',3'-dimethyl-1,2,4,9-tetrahydro-2'*H*-spiro[carbazole-3,5'-pyrimidine]-2',4',6'-(1'*H*,3'*H*)-trione (4g):

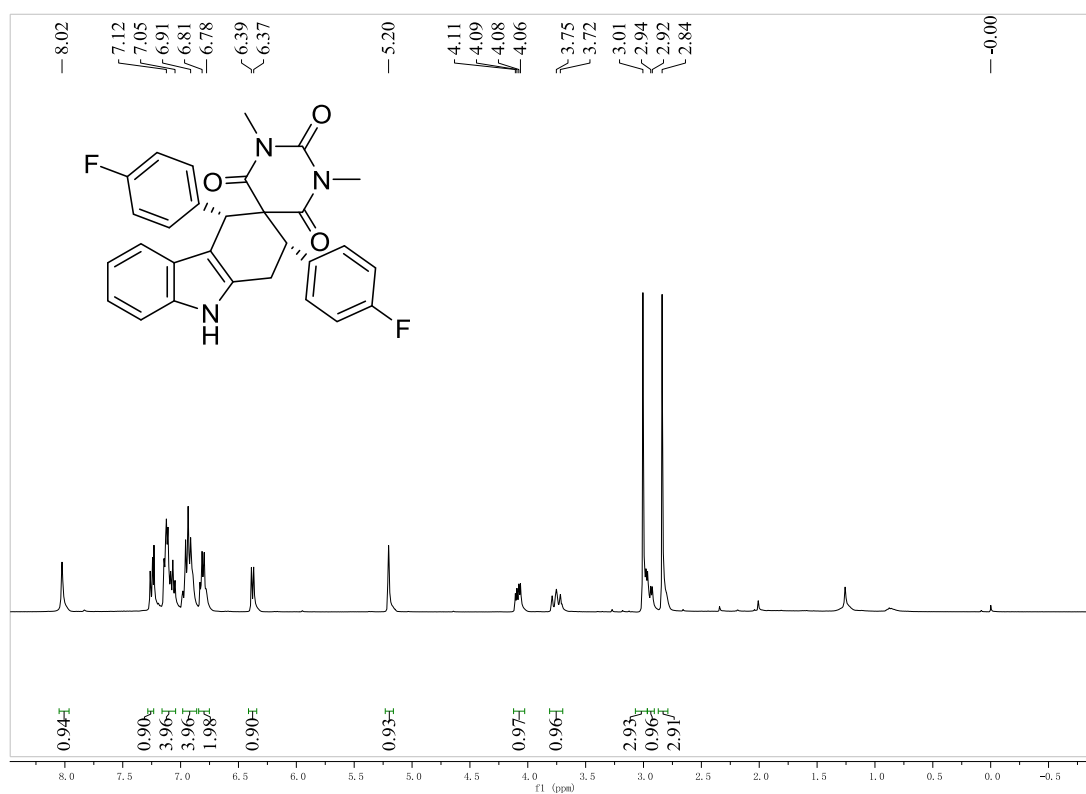
purple solid, 67%, m.p. 211-213 °C; ¹H NMR (400 MHz, CDCl₃) δ: 8.07 (s, 1H, NH), 7.42 (t, *J* = 7.6 Hz, 2H, ArH), 7.28 (d, *J* = 8.4 Hz, 1H, ArH), 7.24-7.20 (m, 1H, ArH), 7.17-7.12 (m, 3H, ArH), 7.04 (t, *J* = 8.4 Hz, 2H, ArH), 6.91 (dd, *J*₁ = 8.0 Hz, *J*₂ = 1.6 Hz, 1H, ArH), 6.76 (t, *J* = 7.6 Hz, 1H, ArH), 6.18 (d, *J* = 8.0 Hz, 1H, ArH), 5.71 (s, 1H, CH), 5.00 (dd, *J*₁ = 12.0 Hz, *J*₂ = 5.6 Hz, 1H, CH), 3.77 (t, *J* = 16.8 Hz, 1H, CH), 3.13 (dd, *J*₁ = 16.0 Hz, *J*₂ = 5.6 Hz, 1H, CH), 3.09 (s, 3H, CH₃), 2.93 (s, 3H, CH₃); ¹³C NMR (400 MHz, CDCl₃) δ: 169.6, 168.8, 150.3, 138.5, 136.0, 135.4, 134.9, 134.5, 134.4, 131.8, 130.6, 129.6, 129.6, 128.6, 127.5, 126.5, 126.3, 126.0, 121.3, 119.2, 119.0, 110.7, 107.1, 58.9, 48.3, 43.1, 29.3, 28.6, 28.1; IR (KBr) ν: 3356, 3168, 3044, 2973, 1767, 1654, 1638, 1566, 1452, 1374, 1331, 1250, 1147, 967, 839 cm⁻¹; MS (*m/z*): HRMS (ESI) Calcd. for C₂₉H₂₃Cl₂³⁵N₃O₃ ([M+Na]⁺): 554.1009, found: 554.1002. C₂₉H₂₃Cl₂³⁷N₃O₃ ([M+Na]⁺): 556.0979, found: 556.0979.

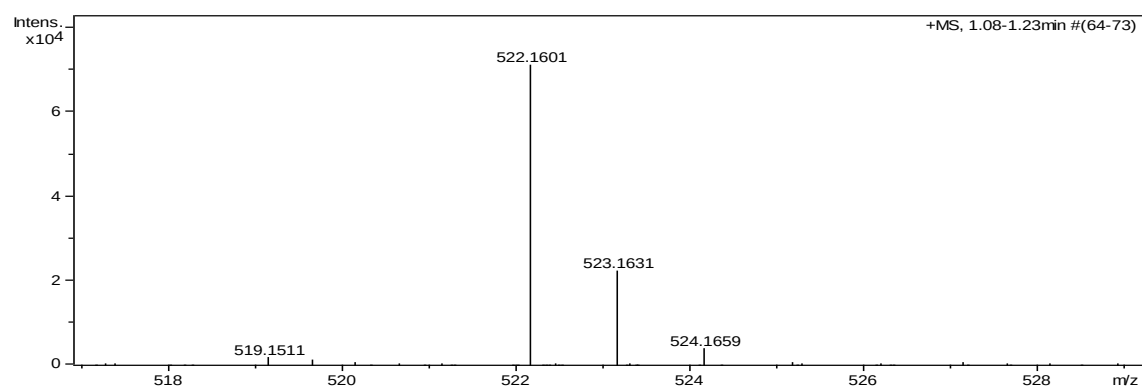
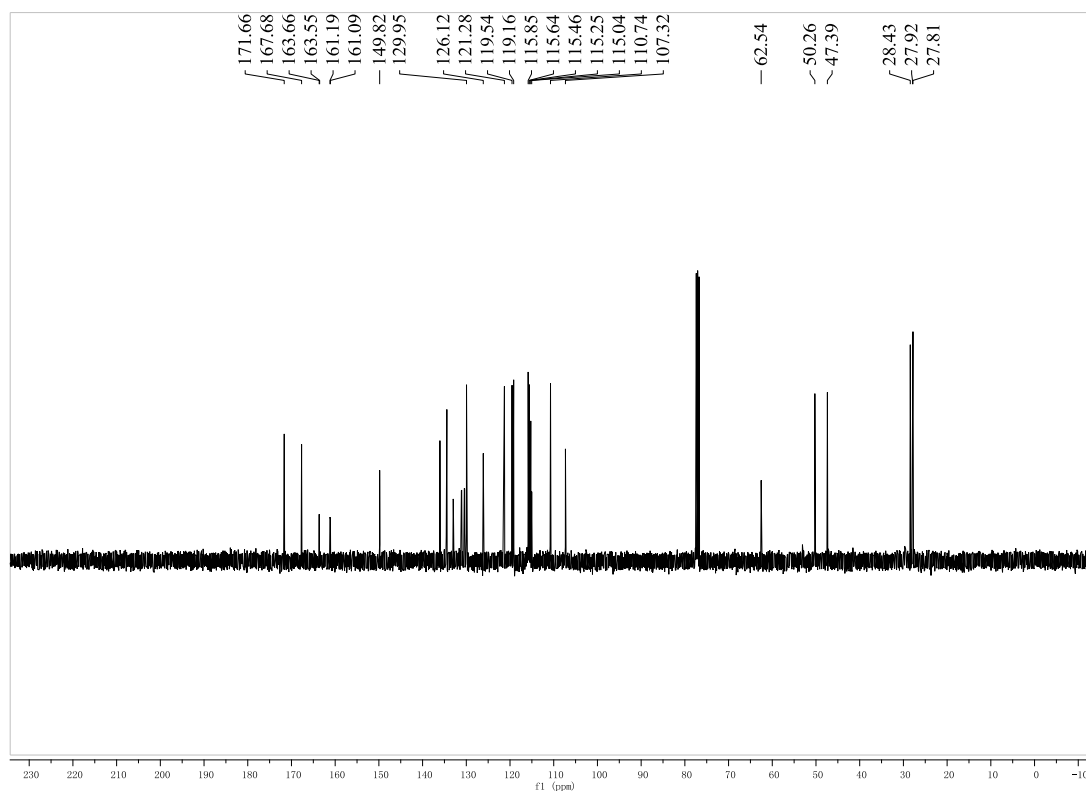




**2,4-Bis(4-fluorophenyl)-1',3'-dimethyl-1,2,4,9-tetrahydro-2'H-spiro[carbazole-3,5'-pyrimidin
e]-2',4',6'(1'H,3'H)-trione (4h):**

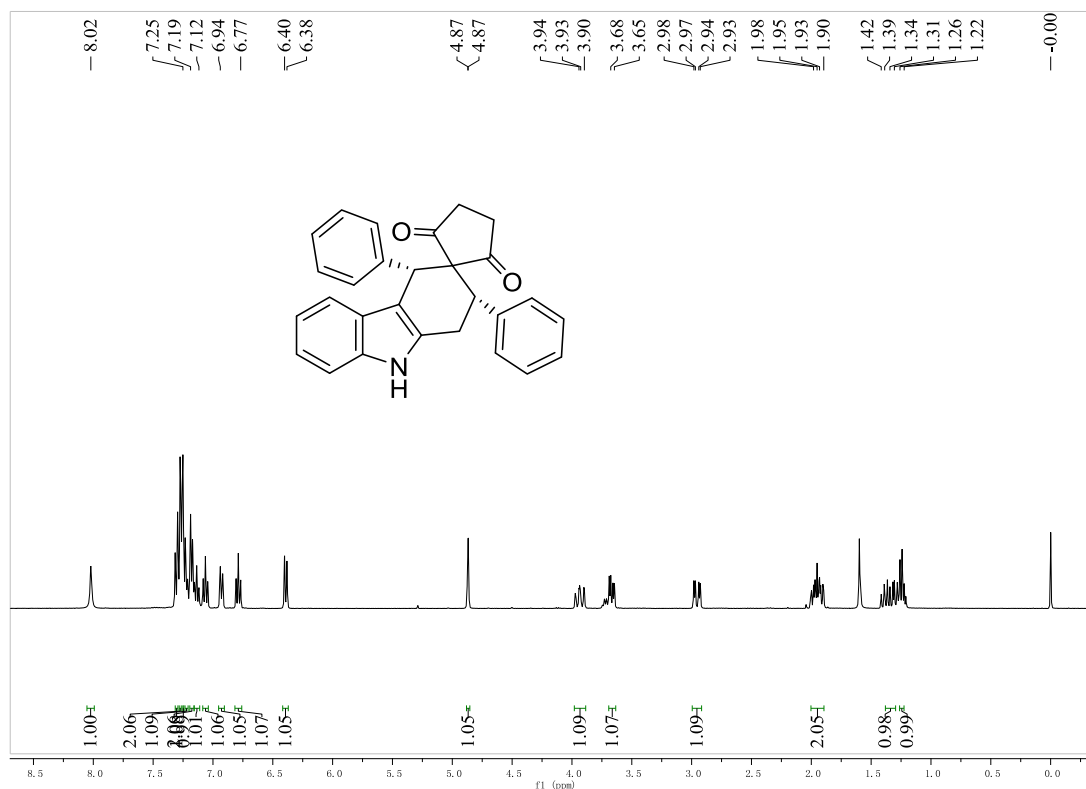
purple solid, 62%, m.p. 214-216 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.03 (s, 1H, NH), 7.25 (d, J = 8.0 Hz, 1H, ArH), 7.14-7.05 (m, 4H, ArH), 6.98-6.90 (m, 4H, ArH), 6.83-6.78 (m, 2H, ArH), 6.37 (d, J = 8.0 Hz, 1H, ArH), 5.20 (s, 1H, CH), 4.08 (dd, J_1 = 12.0 Hz, J_2 = 5.6 Hz, 1H, CH), 3.79-3.72 (m, 1H, CH), 3.01 (s, 3H, CH_3), 2.93 (d, J = 5.6 Hz, 1H, CH), 2.84 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 171.6, 167.6, 163.6, 163.5, 161.1, 161.0, 149.8, 136.0, 134.5, 134.5, 134.4, 133.0, 132.9, 131.1, 131.0, 130.4, 130.3, 129.9, 129.8, 126.1, 121.2, 119.5, 119.1, 115.8, 115.6, 115.4, 115.2, 115.0, 110.7, 107.3, 62.5, 50.2, 47.3, 28.4, 27.9, 27.8; IR (KBr) ν : 3403, 3162, 3079, 2977, 1781, 1663, 1640, 1531, 1442, 1338, 1308, 1271, 1165, 978, 834 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{29}\text{H}_{23}\text{F}_2\text{N}_3\text{O}_3$ ($[\text{M}+\text{Na}]^+$): 522.1600, found: 522.1601.

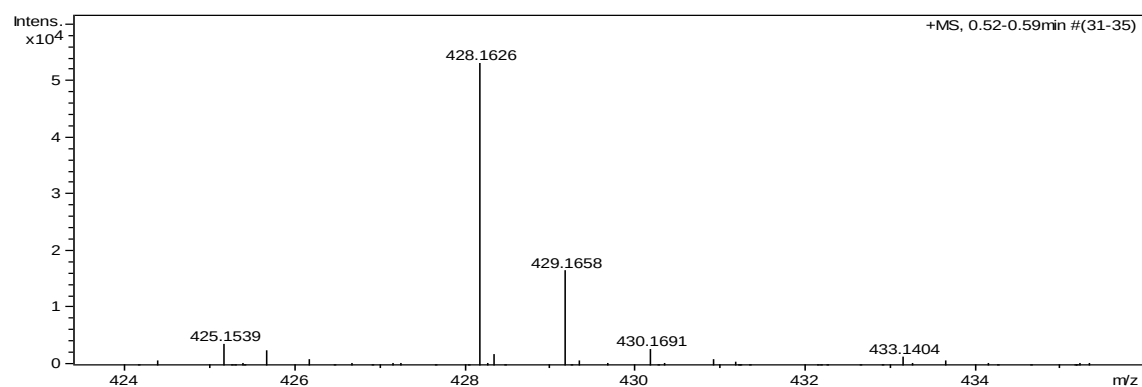
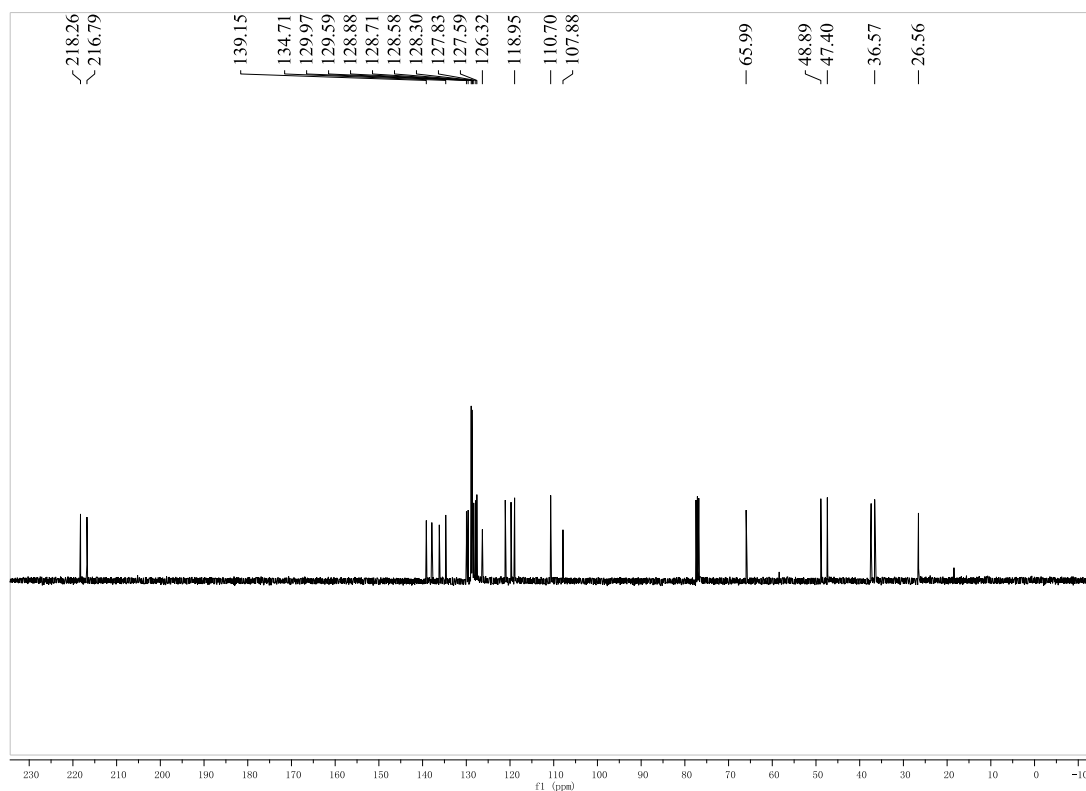




2,4-Diphenyl-1,2,4,9-tetrahydrospiro[carbazole-3,1'-cyclopentane]-2',5'-dione (5a):

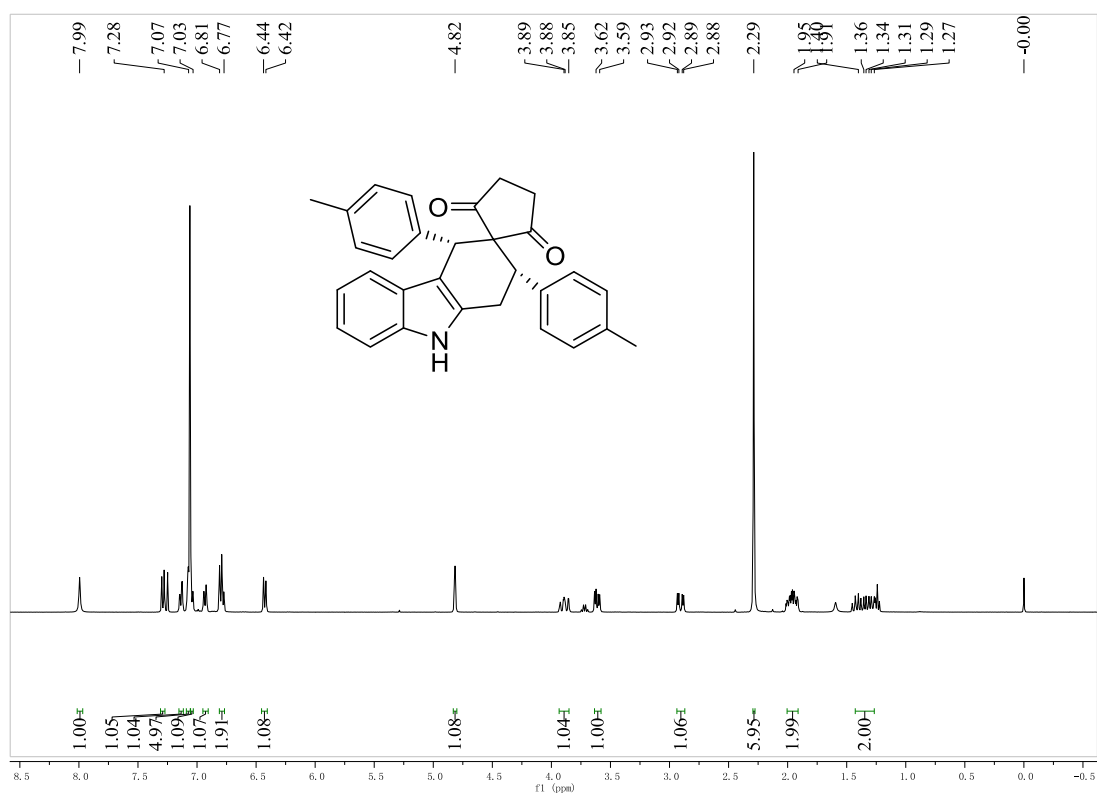
purple solid, 74%, m.p. 178-180 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.02 (s, 1H, NH), 7.30 (d, $J = 8.4$ Hz, 1H, ArH), 7.28-7.27 (m, 2H, ArH), 7.26-7.25 (m, 1H, ArH), 7.22 (d, $J = 8.0$ Hz, 1H, ArH), 7.19-7.17 (m, 2H, ArH), 7.14 (t, $J = 7.6$ Hz, 1H, ArH), 7.06 (t, $J = 7.2$ Hz, 1H, ArH), 6.92 (d, $J = 8.0$ Hz, 1H, ArH), 6.79 (t, $J = 7.2$ Hz, 1H, ArH), 6.39 (d, $J = 8.0$ Hz, 1H, ArH), 4.87 (s, 1H, CH), 3.97-3.90 (m, 1H, CH), 3.66 (dd, $J_1 = 12.4$ Hz, $J_2 = 4.8$ Hz, 1H, CH), 2.95 (dd, $J_1 = 16.0$ Hz, $J_2 = 4.8$ Hz, 1H, CH), 2.01-1.90 (m, 2H, CH_2), 1.39-1.31 (m, 1H, CH), 1.26-1.22 (m, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 218.2, 216.7, 139.1, 137.8, 136.1, 134.7, 129.9, 129.5, 128.8, 128.7, 128.5, 128.2, 127.8, 127.5, 126.3, 121.0, 119.7, 118.9, 110.7, 107.8, 65.9, 48.8, 47.4, 37.3, 36.5, 26.5; IR (KBr) ν : 3200, 3173, 3064, 2978, 1861, 1745, 1677, 1631, 1567, 1467, 1382, 1311, 1254, 1132, 960, 841, 781 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{28}\text{H}_{23}\text{NO}_2$ ($[\text{M}+\text{Na}]^+$): 428.1621, found: 428.1626.

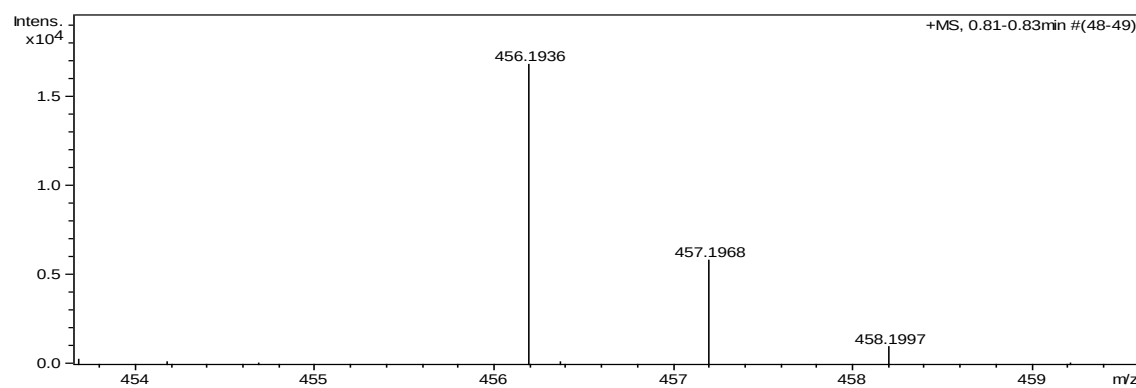
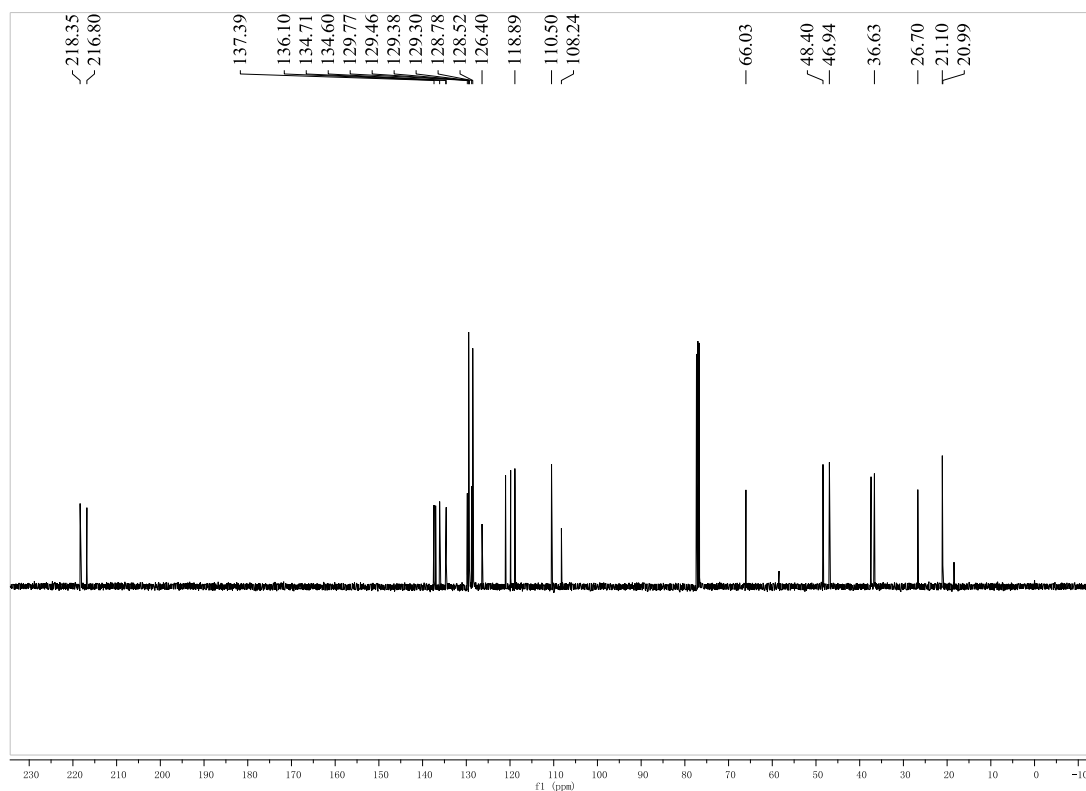




2,4-Di-p-tolyl-1,2,4,9-tetrahydrospiro[carbazole-3,1'-cyclopentane]-2',5'-dione (5b):

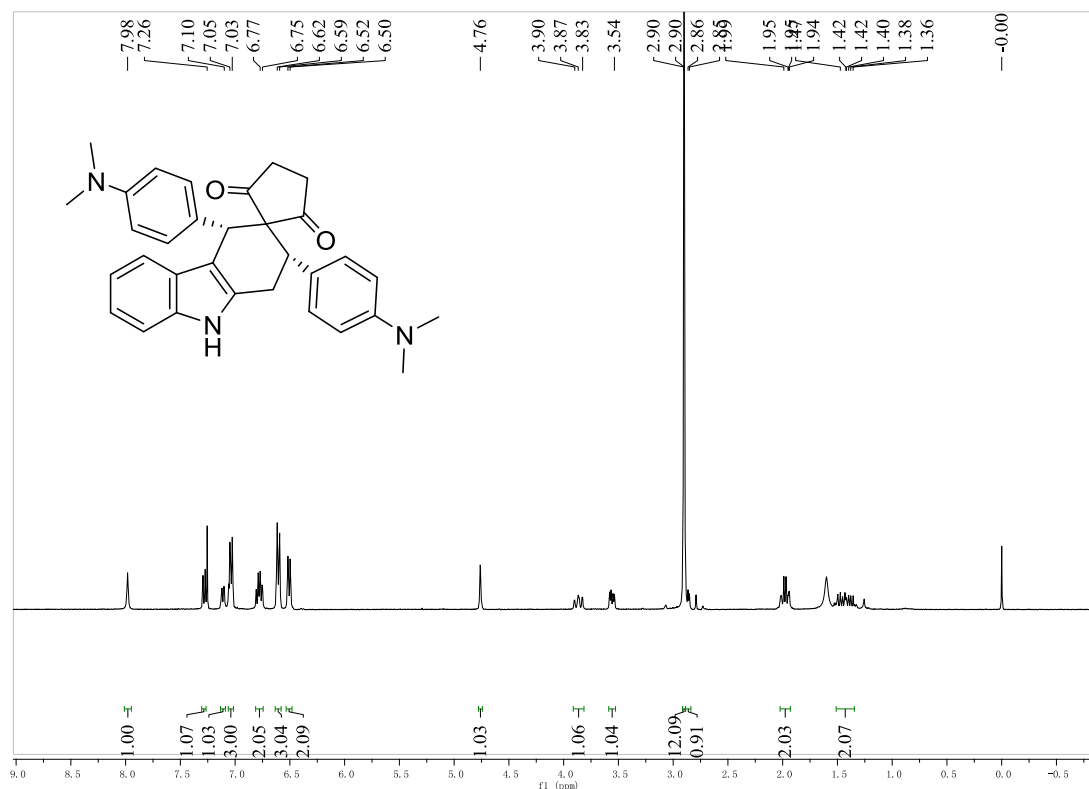
purple solid, 78%, m.p. 176-178 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.99 (s, 1H, NH), 7.28 (d, J = 8.4 Hz, 1H, ArH), 7.13 (dd, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H, ArH), 7.08-7.06 (m, 5H, ArH), 7.05-7.03 (m, 1H, ArH), 6.93 (d, J = 8.0 Hz, 1H, ArH), 6.79 (t, J = 7.2 Hz, 2H, ArH), 6.42 (d, J = 8.0 Hz, 1H, ArH), 4.82 (s, 1H, CH), 3.93-3.85 (m, 1H, CH), 3.61 (dd, J_1 = 12.4 Hz, J_2 = 5.2 Hz, 1H, CH), 2.91 (dd, J_1 = 16.4 Hz, J_2 = 5.2 Hz, 1H, CH), 2.28 (s, 3H, CH_3), 2.27 (s, 3H, CH_3), 2.01-1.91 (m, 2H, CH_2), 1.43-1.27 (m, 2H, CH_2); ^{13}C NMR (400 MHz, CDCl_3) δ : 218.3, 216.8, 137.3, 137.0, 136.1, 134.7, 134.6, 129.7, 129.4, 129.3, 129.3, 128.7, 128.5, 126.4, 121.0, 119.8, 118.8, 110.5, 108.2, 66.0, 48.3, 46.9, 37.4, 36.6, 26.7, 21.1, 20.9; IR (KBr) ν : 3213, 3172, 3063, 2956, 1876, 1732, 1654, 1632, 1578, 1456, 1367, 1257, 1237, 1169, 899, 763 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{30}\text{H}_{27}\text{NO}_2$ ($[\text{M}+\text{Na}]^+$): 456.1934, found: 456.1936.

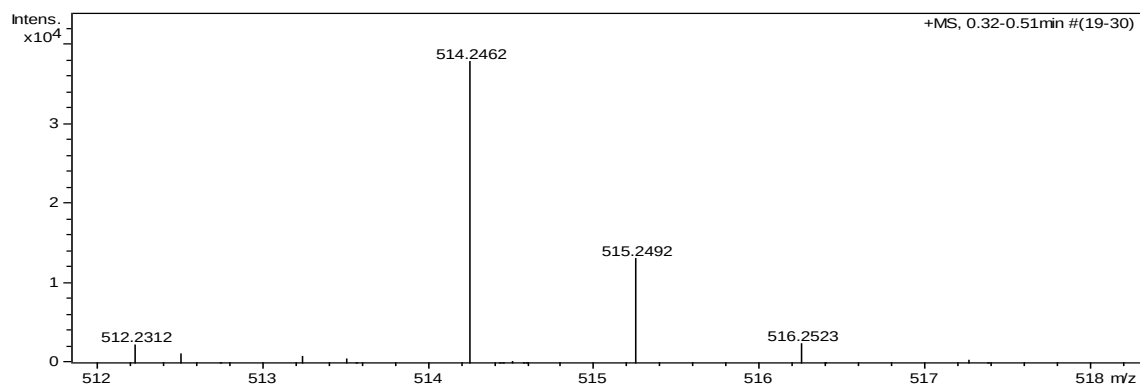
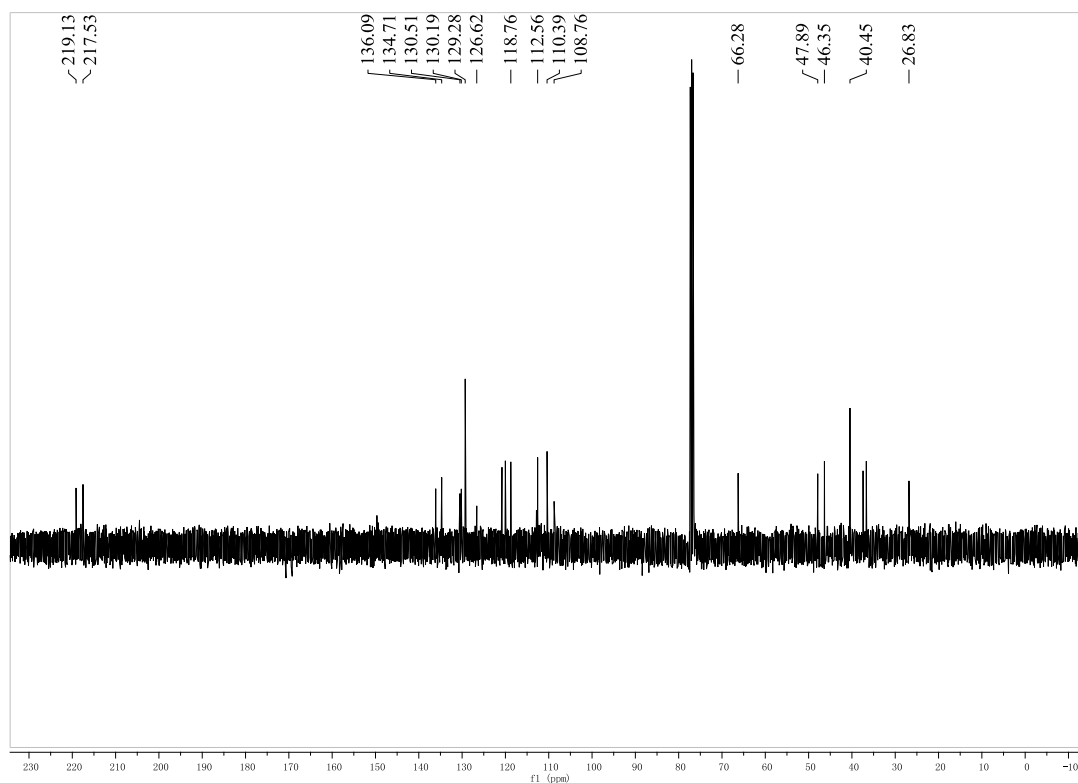




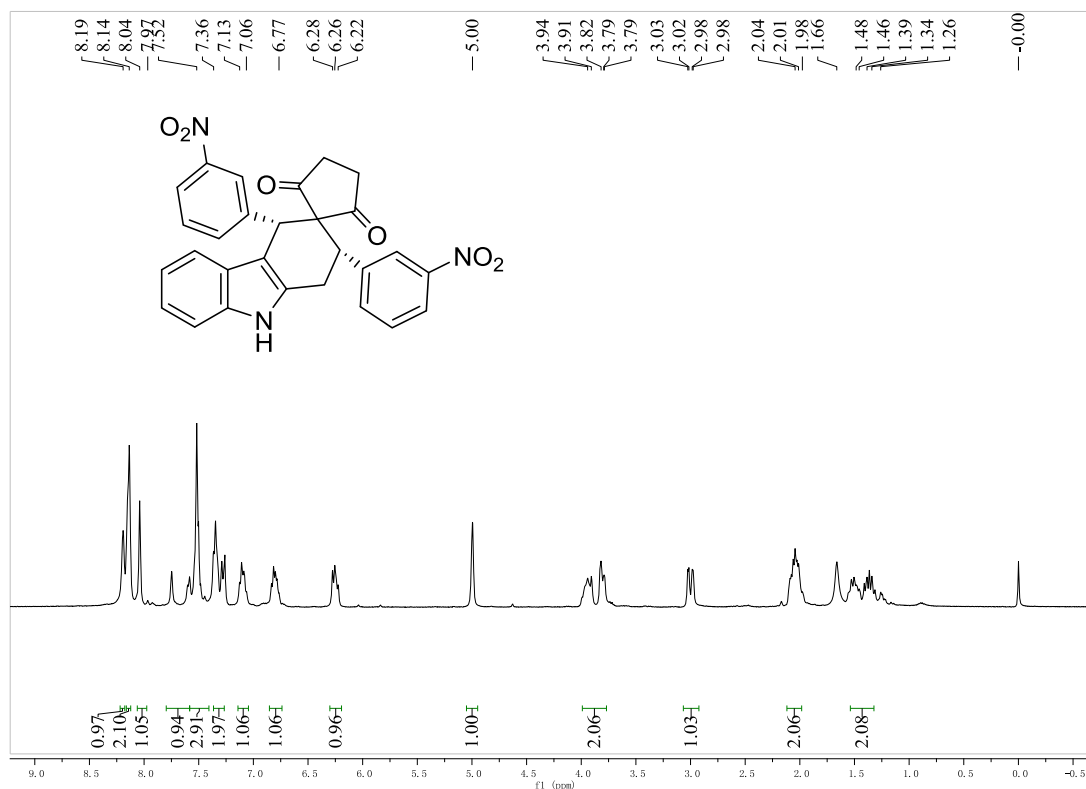
2,4-Bis(4-(dimethylamino)phenyl)-1,2,4,9-tetrahydrospiro[carbazole-3,1'-cyclopentane]-2',5'-dione (5c):

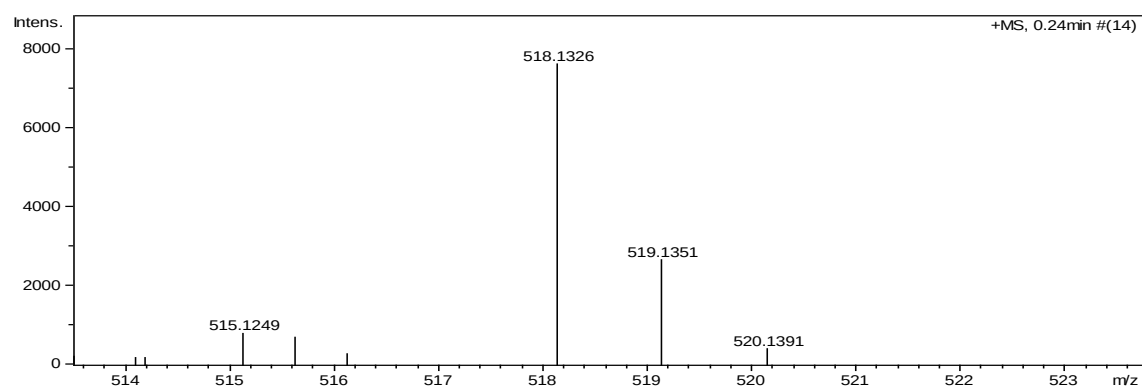
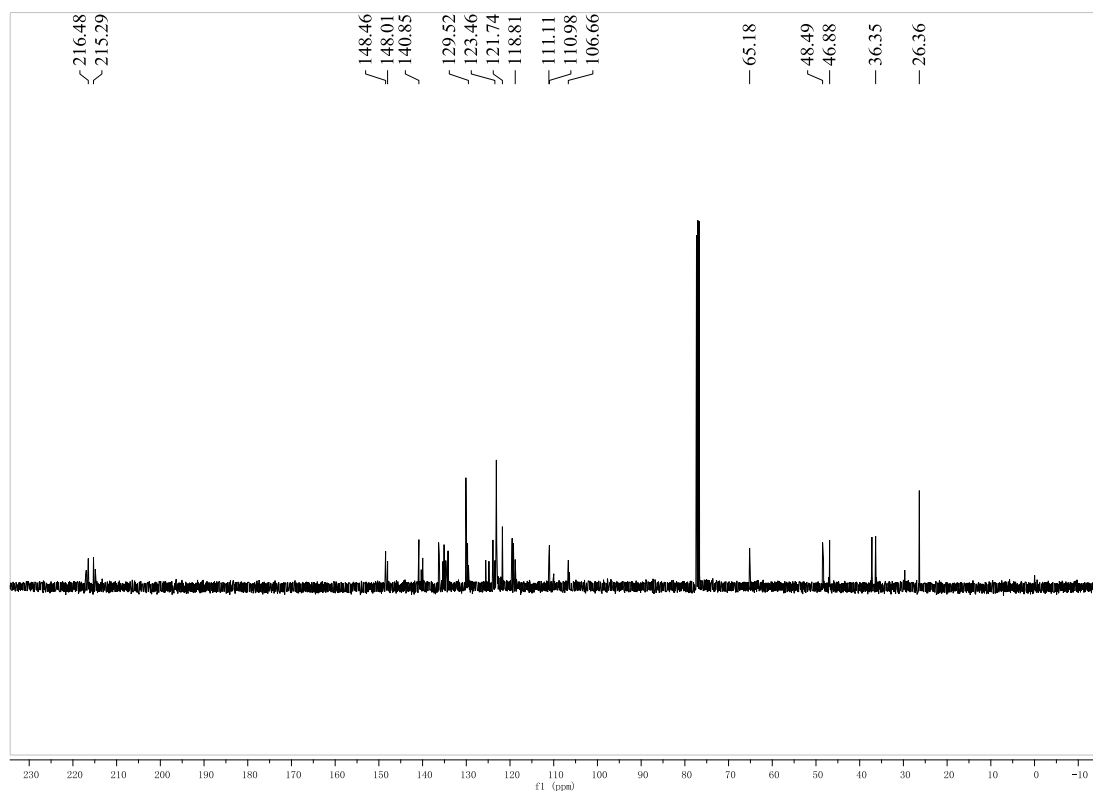
purple solid, 73%, m.p. 180-182 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.98 (s, 1H, NH), 7.28 (d, J = 8.0 Hz, 1H, ArH), 7.11 (dd, J_1 = 8.4 Hz, J_2 = 2.0 Hz, 1H, ArH), 7.06-7.03 (m, 3H, ArH), 6.78 (q, J = 7.6 Hz, 2H, ArH), 6.60 (d, J = 8.4 Hz, 3H, ArH), 6.50 (d, J = 8.0 Hz, 2H, ArH), 4.76 (s, 1H, CH), 3.90-3.83 (m, 1H, CH), 3.56 (dd, J_1 = 12.0 Hz, J_2 = 5.2 Hz, 1H, CH), 2.90 (s, 3H, CH_3), 2.89 (s, 3H, CH_3), 2.88 (s, 3H, CH_3), 2.87 (s, 3H, CH_3), 2.86 (d, J = 5.2 Hz, 1H, CH), 2.02-1.94 (m, 2H, CH_2), 1.50-1.36 (m, 2H, CH_2); ^{13}C NMR (400 MHz, CDCl_3) δ : 219.1, 217.5, 136.0, 134.7, 130.5, 130.1, 129.2, 126.6, 120.8, 120.0, 118.7, 112.5, 110.3, 108.7, 66.2, 47.8, 46.3, 40.5, 40.4, 37.4, 36.6, 26.8; IR (KBr) ν : 3217, 3189, 3046, 2983, 2817, 2156, 1833, 1678, 1611, 1534, 1429, 1351, 1269, 1138, 1131, 962, 888, 817, 764 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{32}\text{H}_{33}\text{N}_3\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 514.2465, found: 514.2462.





2,4-Bis(3-nitrophenyl)-1,2,4,9-tetrahydrospiro[carbazole-3,1'-cyclopentane]-2',5'-dione (5d): purple solid, 59%, m.p. 190-192 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.19 (s, 1H, NH), 8.15-8.13 (m, 2H, ArH), 8.04-7.97 (m, 1H, ArH), 7.75-7.58 (m, 1H, ArH), 7.58-7.42 (m, 3H, ArH), 7.36-7.26 (m, 2H, ArH), 7.13-7.06 (m, 1H, ArH), 6.83-6.77 (m, 1H, ArH), 6.26 (t, $J = 8.4$ Hz, 1H, ArH), 5.00 (s, 1H, CH), 3.97-3.79 (m, 2H, CH_2), 3.30 (dd, $J_1 = 16.4$ Hz, $J_2 = 3.6$ Hz, 1H, CH), 2.09-1.98 (m, 2H, CH_2), 1.55-1.34 (m, 2H, CH_2); ^{13}C NMR (400 MHz, CDCl_3) δ : 216.4, 215.2, 148.4, 148.0, 140.8, 139.9, 136.3, 136.2, 136.1, 135.3, 135.0, 134.6, 134.1, 130.1, 129.7, 129.5, 125.5, 124.8, 123.9, 123.4, 123.1, 123.0, 122.9, 121.7, 119.6, 119.5, 119.2, 118.8, 111.1, 110.9, 106.6, 65.1, 48.4, 46.8, 37.2, 36.3, 26.3; IR (KBr) ν : 3218, 3178, 3049, 2967, 2867, 2133, 1871, 1649, 1600, 1548, 1467, 1358, 1261, 1180, 1135, 997, 941, 878, 749 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{28}\text{H}_{21}\text{N}_3\text{O}_6$ ($[\text{M}+\text{Na}]^+$): 518.1323, found: 518.1326.

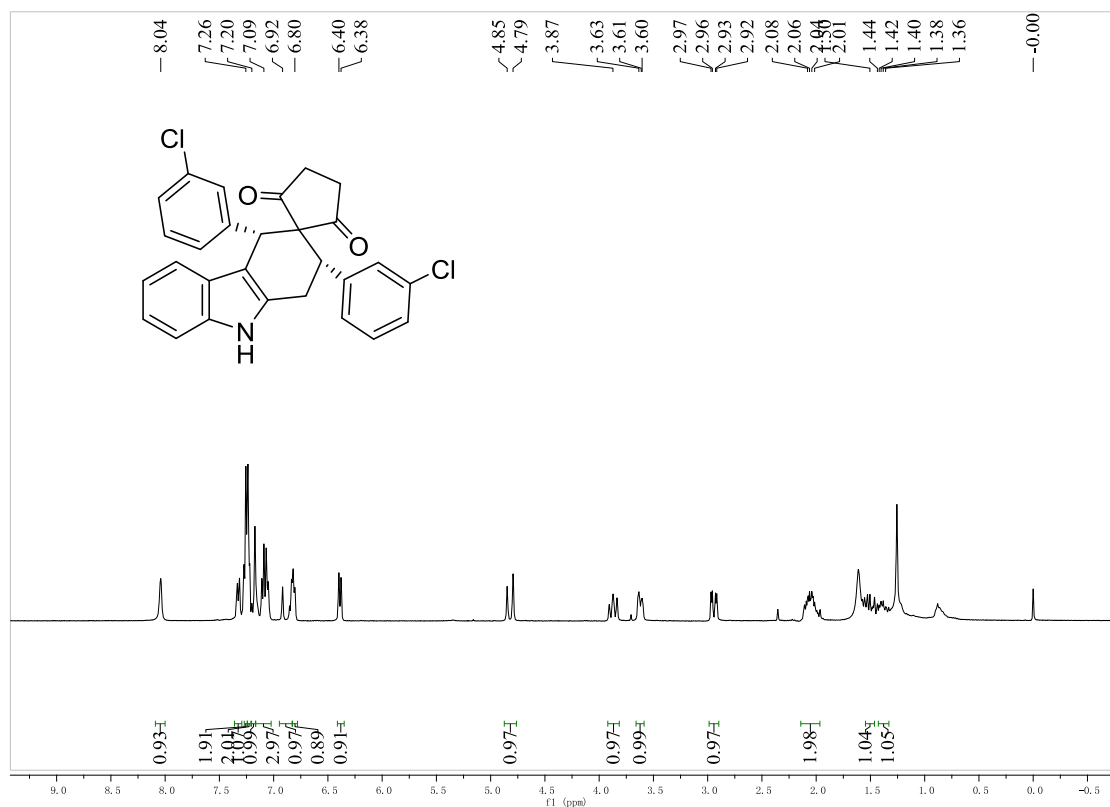


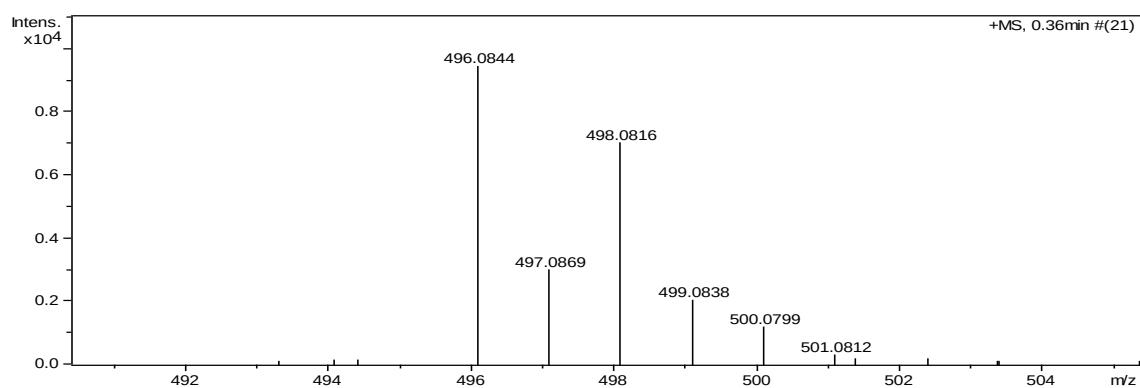
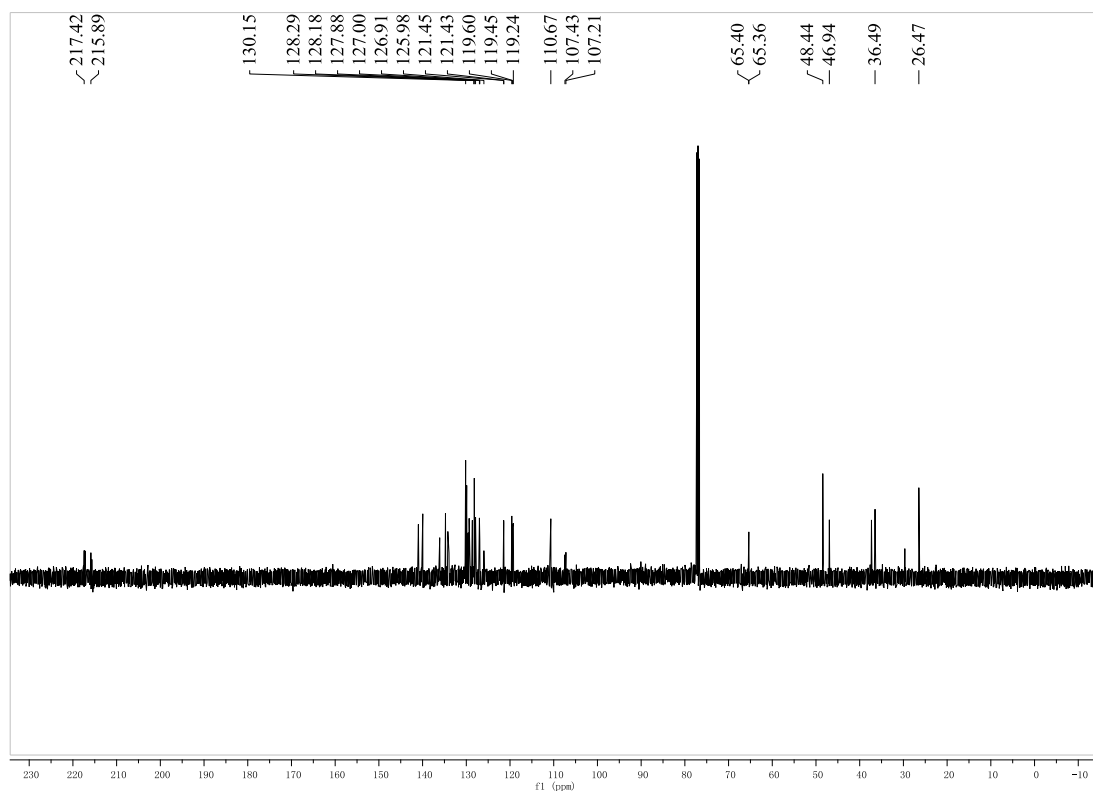


2,4-Bis(3-chlorophenyl)-1,2,4,9-tetrahydrospiro[carbazole-3,1'-cyclopentane]-2',5'-dione

(5e):

purple solid, 64%, m.p. 183-186 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.04 (s, 1H, NH), 7.52 (d, J = 8.0 Hz, 1H, ArH), 7.27-7.26 (m, 2H, ArH), 7.24-7.22 (m, 2H, ArH), 7.18 (d, J = 7.2 Hz, 1H, ArH), 7.17-7.05 (m, 3H, ArH), 6.92-6.84 (m, 1H, ArH), 6.82-6.81 (m, 1H, ArH), 6.38 (d, J = 8.0 Hz, 1H, ArH), 4.82 (d, J = 21.6 Hz, 1H, CH), 3.87 (t, J = 14.8 Hz, 1H, CH), 3.63-3.61 (m, 1H, CH), 2.93 (dd, J_1 = 16.4 Hz, J_2 = 4.4 Hz, 1H, CH), 2.11-2.02 (m, 2H, CH_2), 1.56-1.46 (m, 1H, CH), 1.44-1.36 (m, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 217.4, 215.8, 140.9, 139.9, 136.0, 134.7, 134.2, 133.9, 130.1, 129.9, 129.5, 129.3, 128.6, 128.2, 128.1, 127.8, 127.0, 126.9, 125.9, 121.4, 121.4, 119.5, 119.4, 119.2, 110.6, 107.4, 107.2, 65.3, 48.4, 46.9, 37.3, 36.4, 26.4; IR (KBr) ν : 3211, 3167, 3049, 2971, 2855, 2149, 1854, 1633, 1617, 1559, 1473, 1351, 1249, 1139, 982, 966, 873, 755 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{28}\text{H}_{21}\text{Cl}_2^{35}\text{NO}_2$ ($[\text{M}+\text{Na}]^+$): 496.0842, found: 496.0844. $\text{C}_{28}\text{H}_{21}\text{Cl}_2^{37}\text{NO}_2$ ($[\text{M}+\text{Na}]^+$): 498.0812, found: 498.0816.

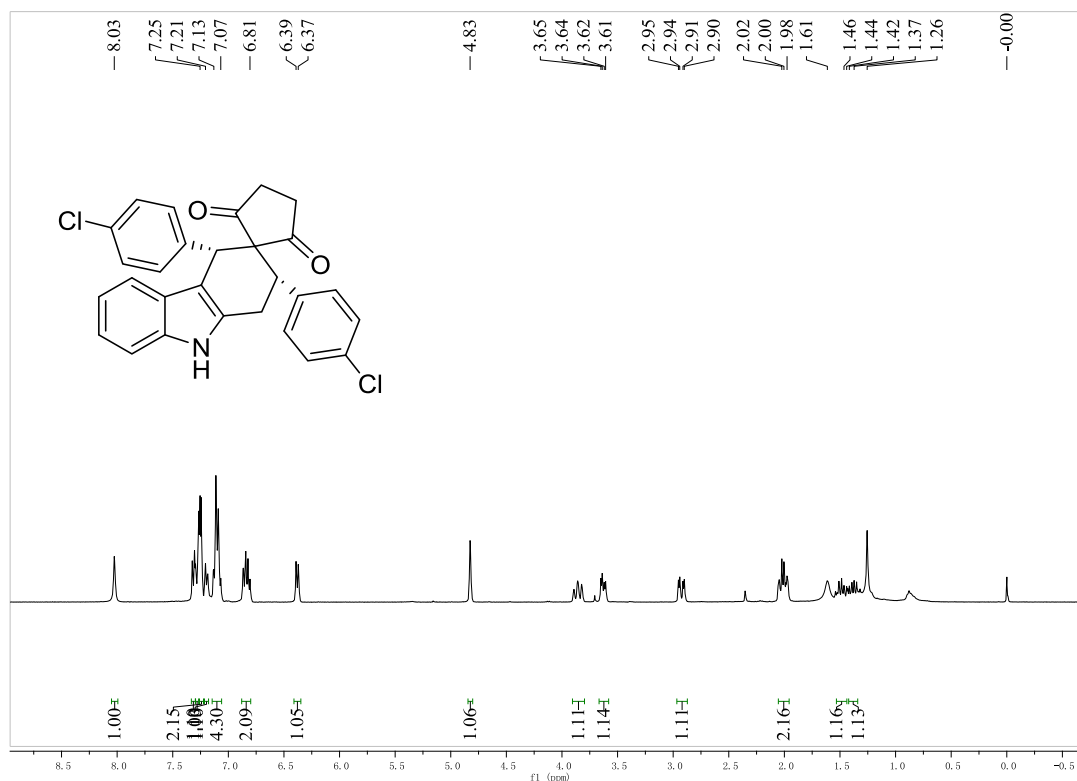


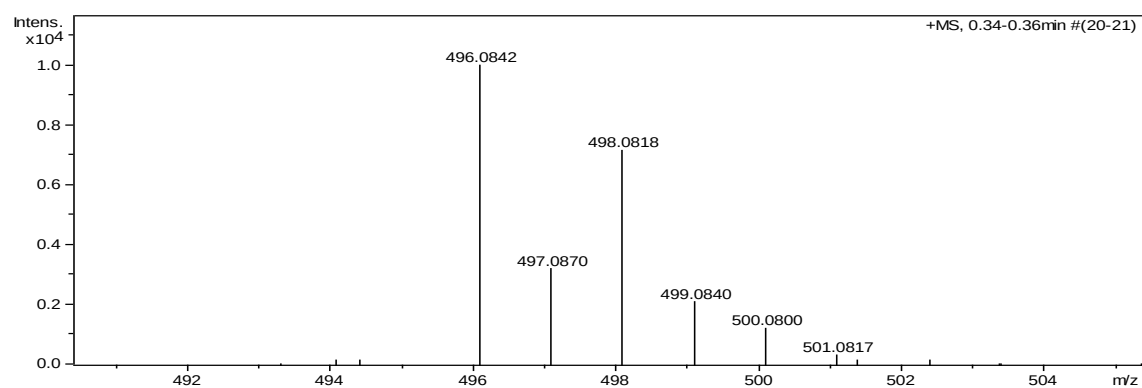
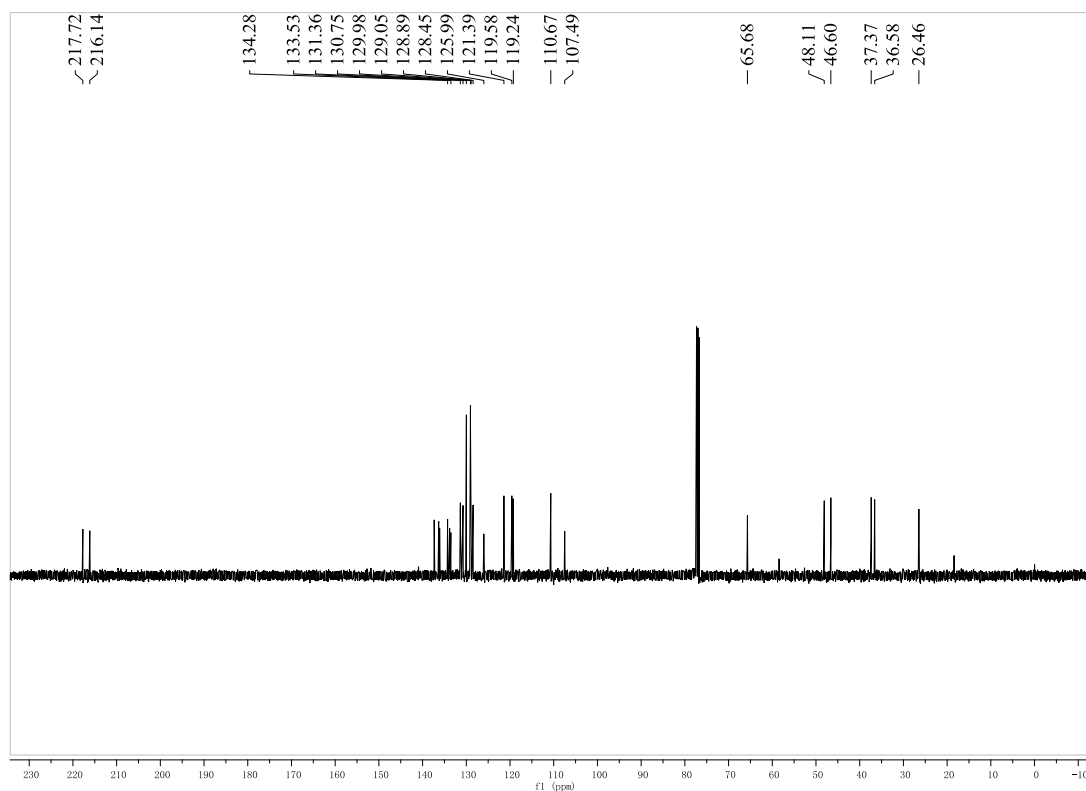


2,4-Bis(4-chlorophenyl)-1,2,4,9-tetrahydrospiro[carbazole-3,1'-cyclopentane]-2',5'-dione

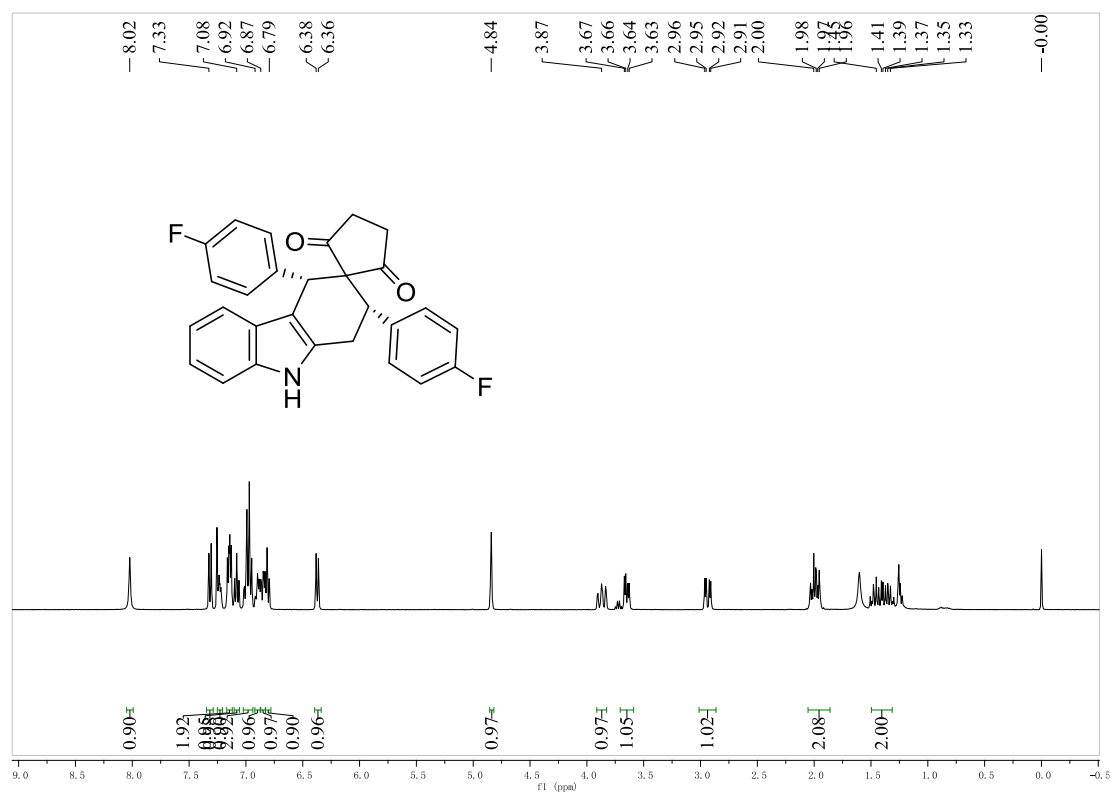
(5f):

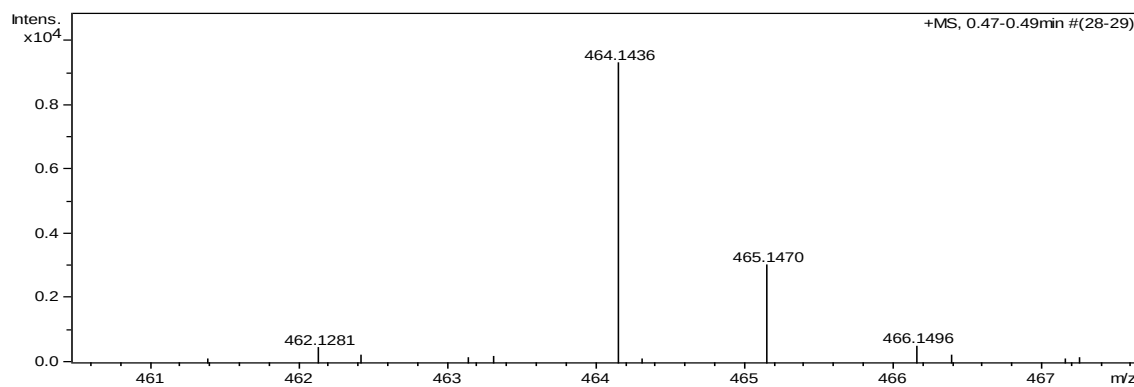
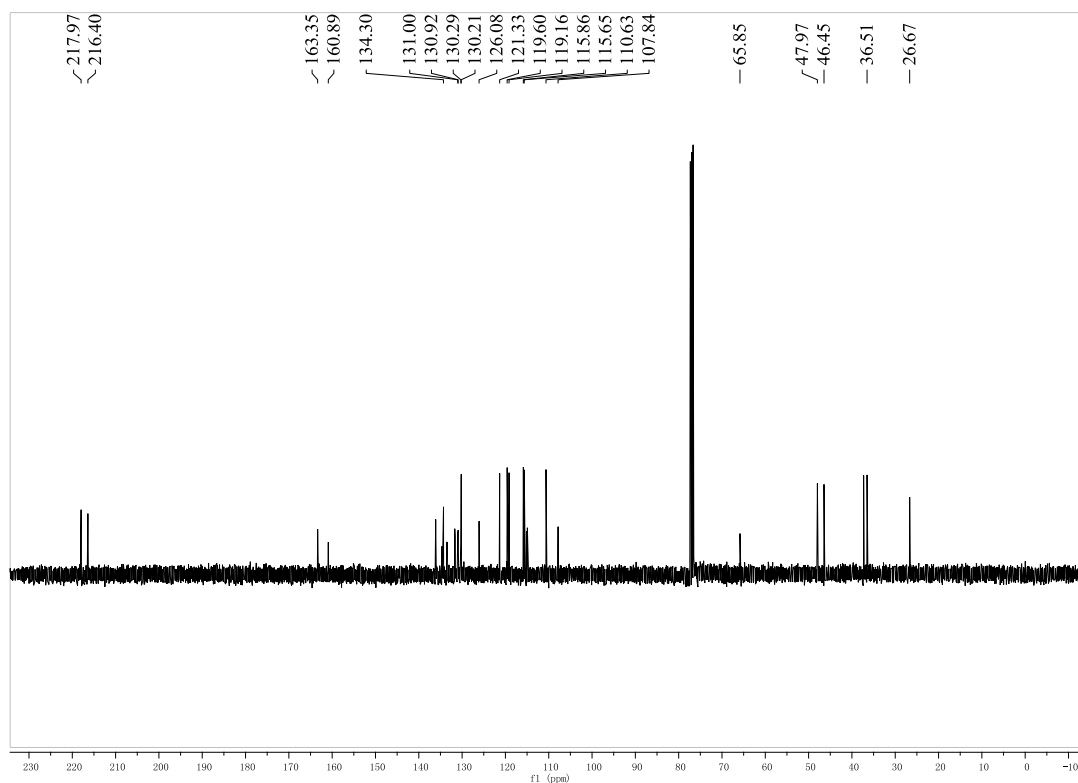
purple solid, 73%, m.p. 180-183 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.03 (s, 1H, NH), 7.31 (d, $J = 8.4$ Hz, 1H, ArH), 7.30-7.27 (m, 1H, ArH), 7.25-7.24 (m, 2H, ArH), 7.19 (d, $J = 8.4$ Hz, 1H, ArH), 7.14-7.07 (m, 4H, ArH), 6.83 (q, $J = 7.6$ Hz, 2H, ArH), 6.38 (d, $J = 7.6$ Hz, 1H, ArH), 4.83 (s, 1H, CH), 3.86 (t, $J = 13.2$ Hz, 1H, ArH), 3.63 (dd, $J_1 = 12.0$ Hz, $J_2 = 4.8$ Hz, 1H, CH), 2.92 (dd, $J_1 = 16.4$ Hz, $J_2 = 4.8$ Hz, 1H, CH), 2.06-1.98 (m, 2H, CH_2), 1.55-1.44 (m, 1H, CH), 1.42-1.35 (m, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 217.7, 216.1, 137.3, 136.3, 136.0, 134.2, 133.8, 133.5, 131.3, 130.7, 129.9, 129.0, 128.8, 128.4, 125.9, 121.3, 119.5, 119.2, 110.6, 107.4, 65.6, 48.1, 46.6, 37.3, 36.5, 26.4; IR (KBr) ν : 3217, 3167, 3034, 2980, 2848, 2167, 1850, 1632, 1600, 1535, 1470, 1362, 1278, 1143, 1101, 972, 900, 824, 768 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{28}\text{H}_{21}\text{Cl}_2^{35}\text{NO}_2$ ($[\text{M}+\text{Na}]^+$): 496.0842, found: 496.0842. $\text{C}_{28}\text{H}_{21}\text{Cl}_2^{37}\text{NO}_2$ ($[\text{M}+\text{Na}]^+$): 498.0812, found: 498.0818.





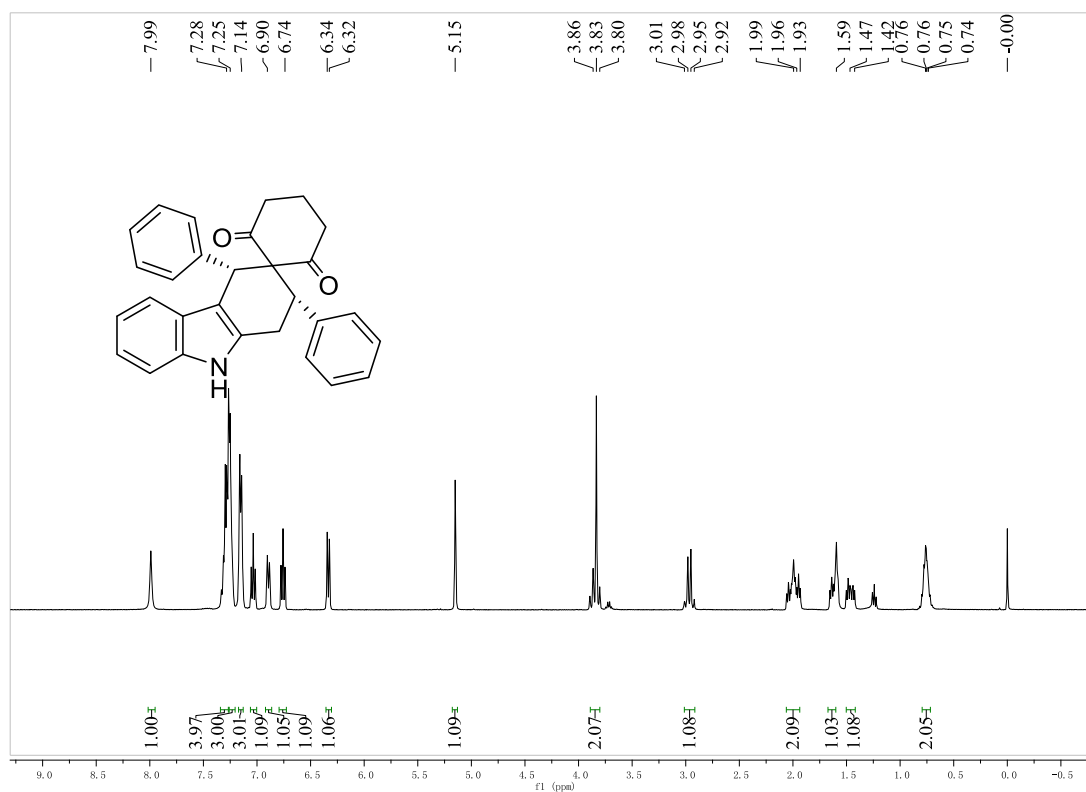
purple solid, 65%, m.p. 179-182 °C; ¹H NMR (400 MHz, CDCl₃) δ: 8.02 (s, 1H, NH), 7.31 (d, *J* = 8.0 Hz, 1H, ArH), 7.24-7.22 (m, 1H, ArH), 7.16-7.13 (m, 2H, ArH), 7.08 (t, *J* = 7.6 Hz, 1H, ArH), 7.01-6.95 (m, 3H, ArH), 6.92-6.88 (m, 1H, ArH), 6.87-6.83 (m, 1H, ArH), 6.80 (d, *J* = 8.0 Hz, 1H, ArH), 6.37 (d, *J* = 8.0 Hz, 1H, ArH), 4.84 (s, 1H, CH), 3.87-3.83 (m, 1H, CH), 3.65 (dd, *J*₁ = 12.0 Hz, *J*₂ = 4.8 Hz, 1H, CH), 2.93 (dd, *J*₁ = 16.4 Hz, *J*₂ = 4.8 Hz, 1H, CH), 2.03-1.96 (m, 2H, CH₂), 1.48-1.33 (m, 2H, CH₂); ¹³C NMR (400 MHz, CDCl₃) δ: 217.9, 216.3, 163.3, 160.8, 136.1, 134.2, 131.6, 131.6, 130.9, 130.9, 130.2, 130.2, 126.0, 121.3, 119.6, 119.1, 115.8, 115.6, 110.6, 107.8, 65.8, 47.9, 46.4, 37.3, 36.5, 26.6; IR (KBr) ν: 3217, 3168, 3049, 2973, 2850, 2146, 1853, 1617, 1600, 1554, 1423, 1331, 1245, 1178, 1116, 973, 888, 807, 759 cm⁻¹; MS (*m/z*): HRMS (ESI) Calcd. for C₂₈H₂₁F₂NO₂ ([M+Na]⁺): 464.1433, found: 464.1436.

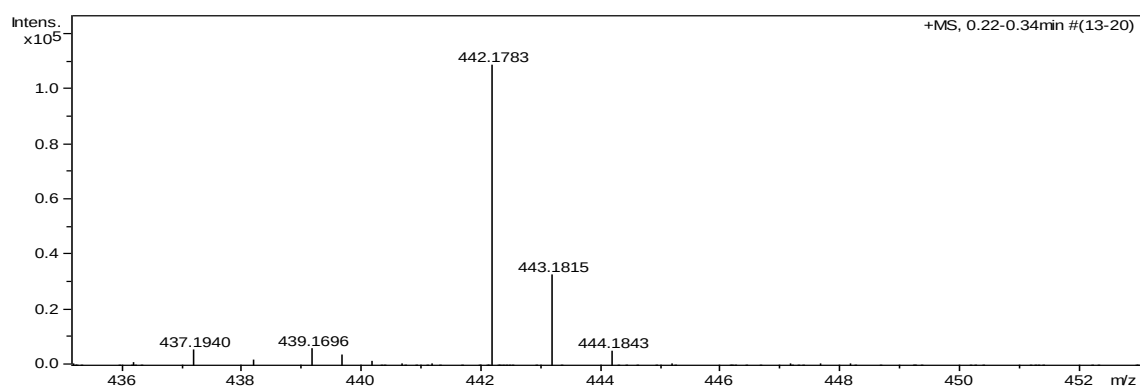
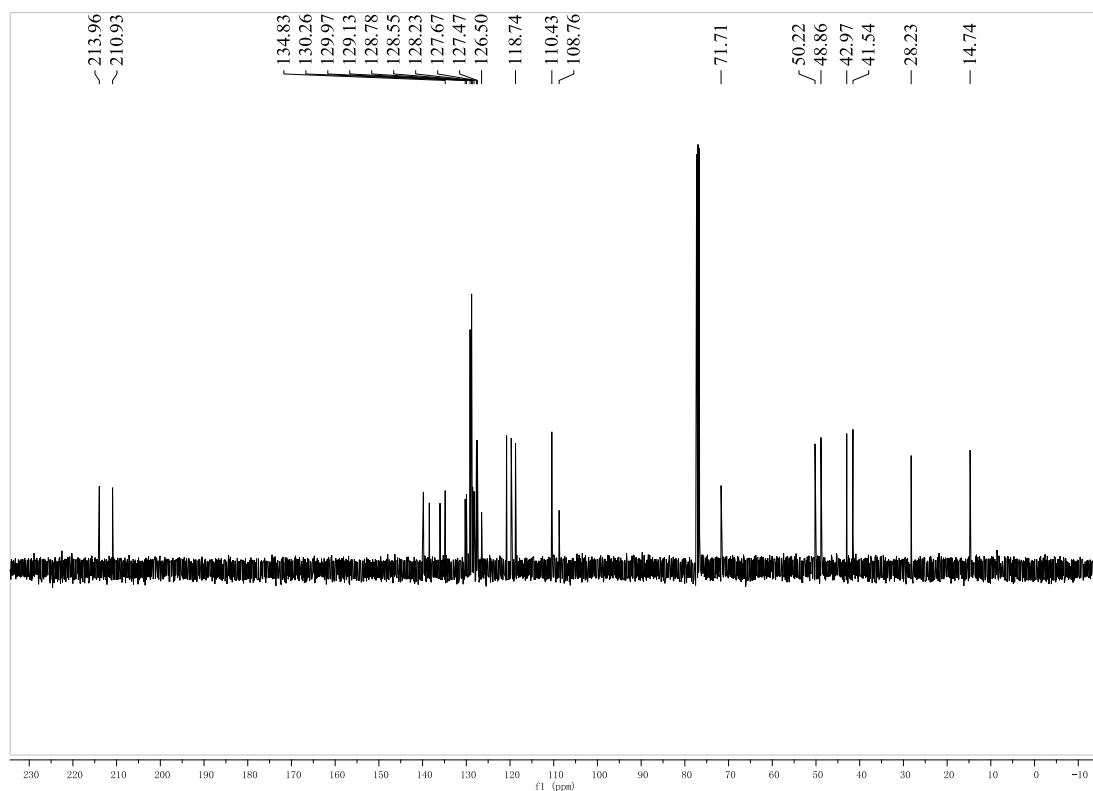




2,4-Diphenyl-1,2,4,9-tetrahydrospiro[carbazole-3,1'-cyclohexane]-2',6'-dione (5h):

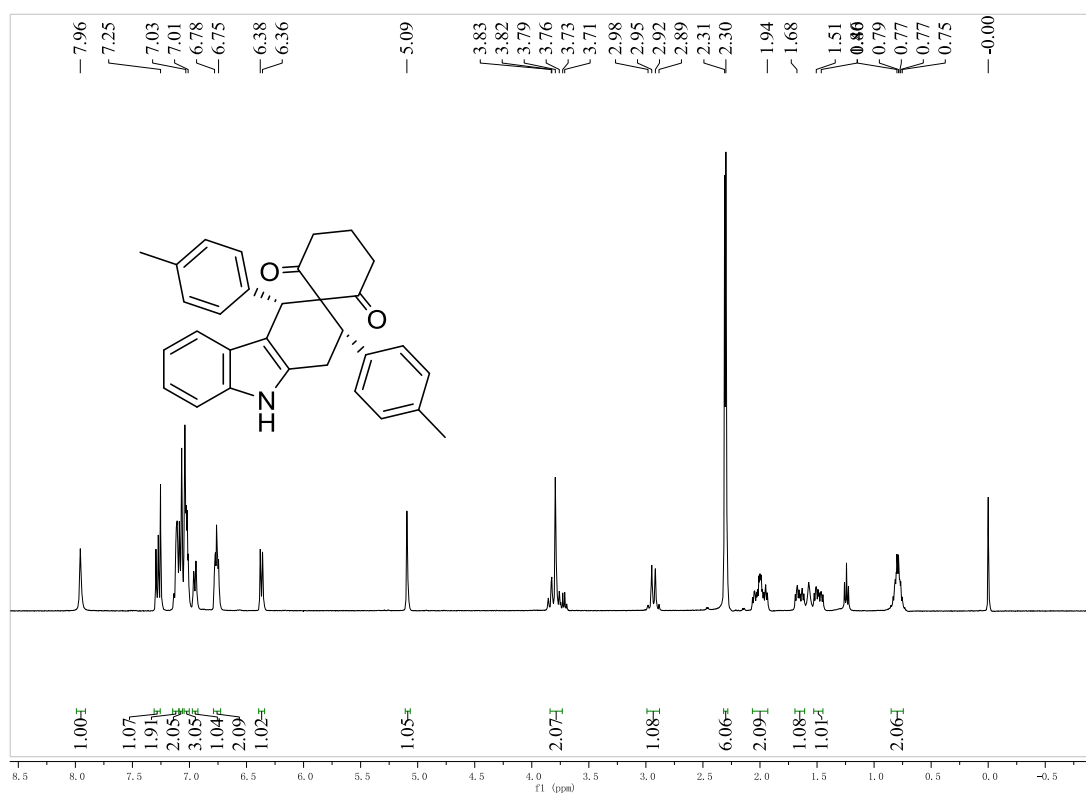
purple solid, 76%, m.p. 182-185 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.99 (s, 1H, NH), 7.33-7.28 (m, 4H, ArH), 7.26-7.23 (m, 3H, ArH), 7.16-7.14 (m, 3H, ArH), 7.04 (t, $J = 7.6$ Hz, 1H, ArH), 6.89 (d, $J = 7.6$ Hz, 1H, ArH), 6.33 (d, $J = 8.0$ Hz, 1H, ArH), 5.15 (s, 1H, CH), 3.83 (t, $J = 12.0$ Hz, 2H, CH_2), 2.97 (t, $J = 12.4$ Hz, 1H, CH), 2.04-1.93 (m, 2H, CH_2), 1.66-1.59 (m, 1H, CH), 1.50-1.43 (m, 1H, CH), 0.80-0.74 (m, 2H, CH_2); ^{13}C NMR (400 MHz, CDCl_3) δ : 213.9, 210.9, 139.8, 138.4, 136.0, 134.8, 130.2, 129.9, 129.1, 128.7, 128.5, 128.2, 127.6, 127.4, 126.5, 120.7, 119.7, 118.7, 110.4, 108.7, 71.7, 50.2, 48.8, 42.9, 41.5, 28.2, 14.7; IR (KBr) ν : 3407, 3271, 3063, 2962, 1871, 1763, 1645, 1600, 1538, 1462, 1345, 1321, 1266, 1148, 961, 844 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{29}\text{H}_{25}\text{NO}_2$ ($[\text{M}+\text{Na}]^+$): 442.1778, found: 442.1783.

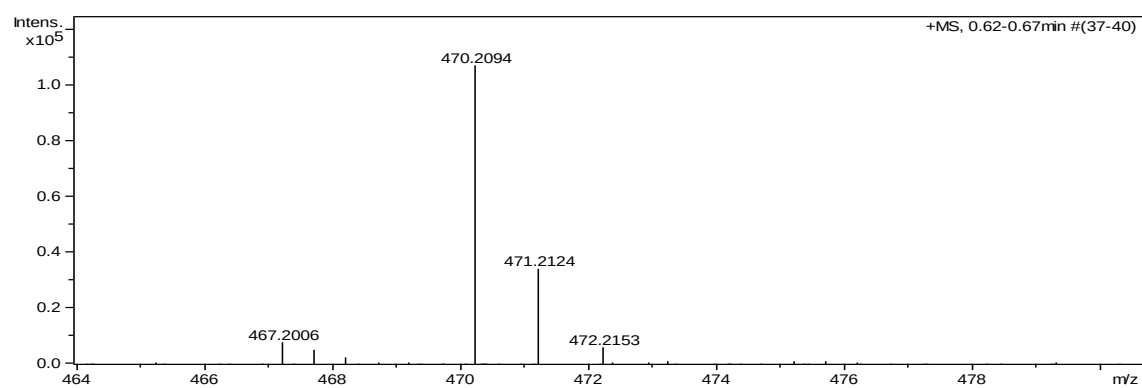
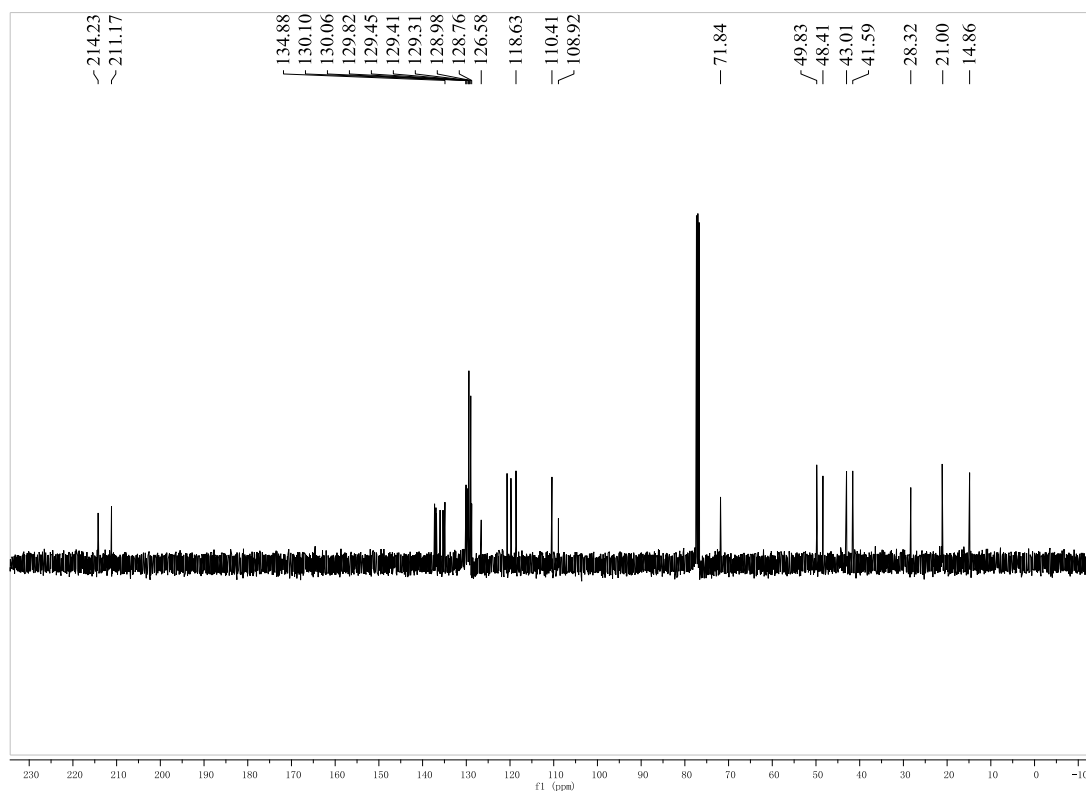




2,4-Di-p-tolyl-1,2,4,9-tetrahydrospiro[carbazole-3,1'-cyclohexane]-2',6'-dione (5i):

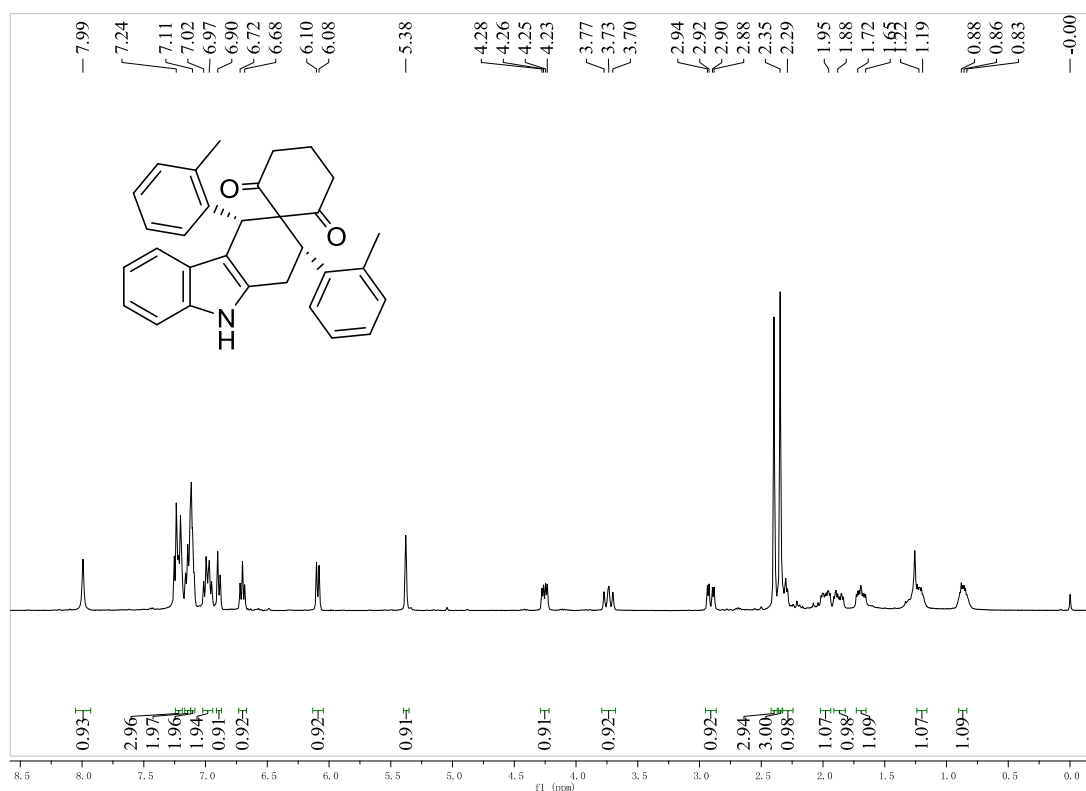
purple solid, 75%, m.p. 182-185 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.96 (s, 1H, NH), 7.28 (d, J = 8.0 Hz, 1H, ArH), 7.14-7.11 (m, 2H, ArH), 7.09-7.07 (m, 2H, ArH), 7.04-7.01 (m, 3H, ArH), 6.95 (d, J = 8.0 Hz, 1H, ArH), 6.78-6.75 (m, 2H, ArH), 6.37 (d, J = 8.0 Hz, 1H, ArH), 5.10 (s, 1H, CH), 3.83-3.73 (m, 2H, CH_2), 2.92 (q, J = 12.8 Hz, 1H, CH), 2.31 (s, 3H, CH_3), 2.30 (s, 3H, CH_3), 2.07-1.94 (m, 2H, CH_2), 1.69-1.62 (m, 1H, CH), 1.53-1.46 (m, 1H, CH), 0.83-0.75 (m, 2H, CH_2); ^{13}C NMR (400 MHz, CDCl_3) δ : 214.2, 211.1, 137.2, 136.9, 136.8, 135.9, 135.3, 134.8, 130.1, 130.0, 129.8, 129.4, 129.4, 129.3, 128.9, 128.7, 126.5, 120.6, 119.7, 118.6, 110.4, 108.9, 71.8, 49.8, 48.4, 43.0, 41.5, 28.3, 21.1, 21.0, 14.8; IR (KBr) ν : 3441, 3245, 3001, 2946, 1867, 1750, 1638, 1617, 1549, 1461, 1355, 1321, 1249, 1157, 969, 864, 771 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{31}\text{H}_{29}\text{NO}_2$ ($[\text{M}+\text{Na}]^+$): 470.2091, found: 470.2094.

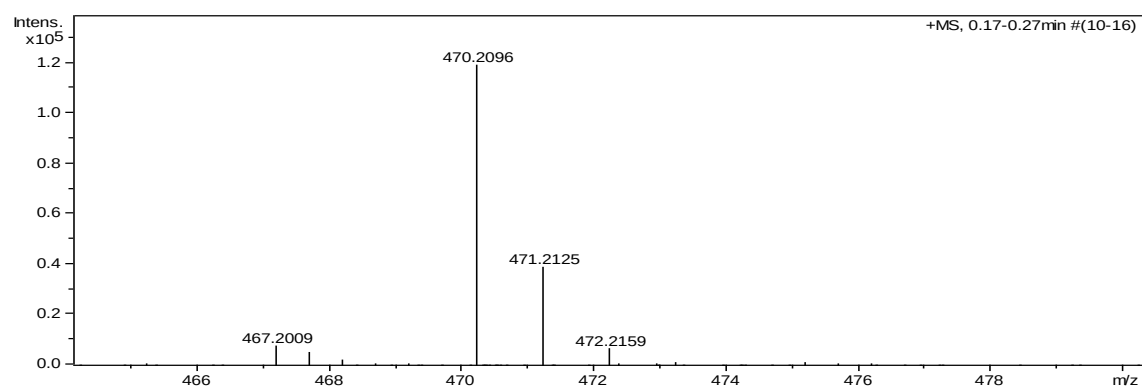
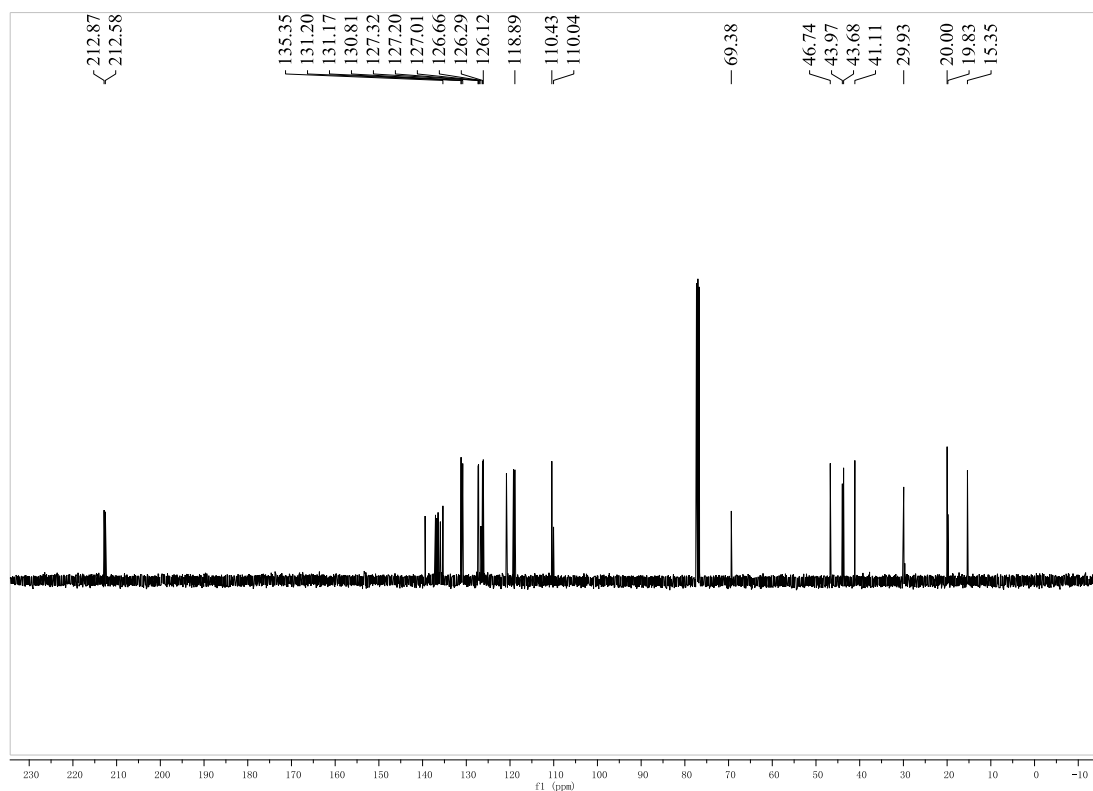




2,4-Di-*o*-tolyl-1,2,4,9-tetrahydrospiro[carbazole-3,1'-cyclohexane]-2',6'-dione (5j):

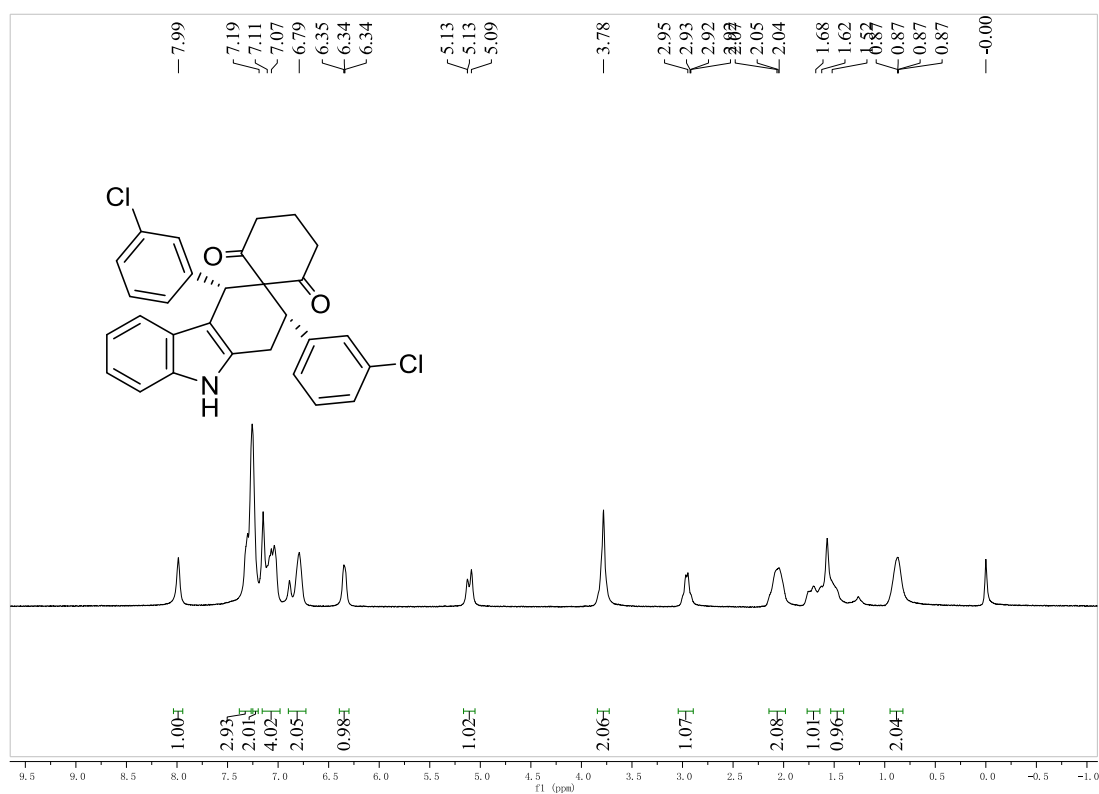
purple solid, 78%, m.p. 188-191 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.99 (s, 1H, NH), 7.24-7.20 (m, 3H, ArH), 7.16-7.12 (m, 2H, ArH), 7.12-7.09 (m, 2H, ArH), 6.98 (dd, $J_1 = 18.4$ Hz, $J_2 = 8.0$ Hz, 2H, ArH), 6.89 (d, $J = 8.0$ Hz, 1H, ArH), 6.70 (t, $J = 8.0$ Hz, 1H, ArH), 6.09 (d, $J = 7.6$ Hz, 1H, ArH), 5.38 (s, 1H, CH), 4.25 (q, $J = 5.6$ Hz, 1H, CH), 3.74 (t, $J = 15.2$ Hz, 1H, CH), 2.90 (dd, $J_1 = 16.0$ Hz, $J_2 = 5.2$ Hz, 1H, CH), 2.40 (s, 3H, CH_3), 2.35 (s, 3H, CH_3), 2.30-2.29 (m, 1H, CH), 2.02-1.94 (m, 1H, CH), 1.91-1.84 (m, 1H, CH), 1.73-1.66 (m, 1H, CH), 1.24-1.19 (m, 1H, CH), 0.89-0.84 (m, 1H, CH); ^{13}C NMR (400 MHz, CDCl_3) δ : 212.8, 212.5, 139.4, 137.0, 136.7, 136.4, 135.9, 135.3, 131.2, 131.1, 130.8, 127.3, 127.1, 127.0, 126.6, 126.2, 126.1, 120.8, 119.2, 118.8, 110.4, 110.0, 69.3, 46.7, 43.9, 43.6, 41.1, 29.9, 20.0, 19.8, 15.3; IR (KBr) ν : 3403, 3169, 3064, 2971, 1846, 1732, 1654, 1645, 1581, 1472, 1331, 1300, 1280, 1157, 969, 867, 782 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{31}\text{H}_{29}\text{NO}_2$ ($[\text{M}+\text{Na}]^+$): 470.2091, found: 470.2096.

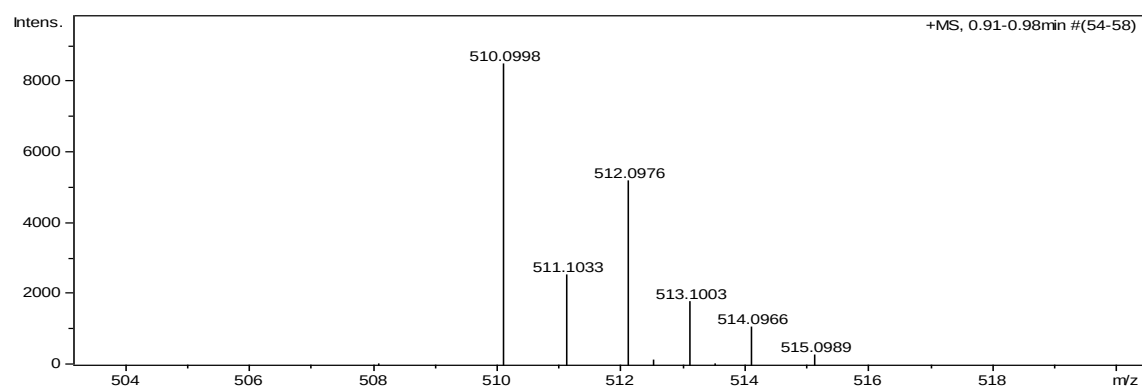
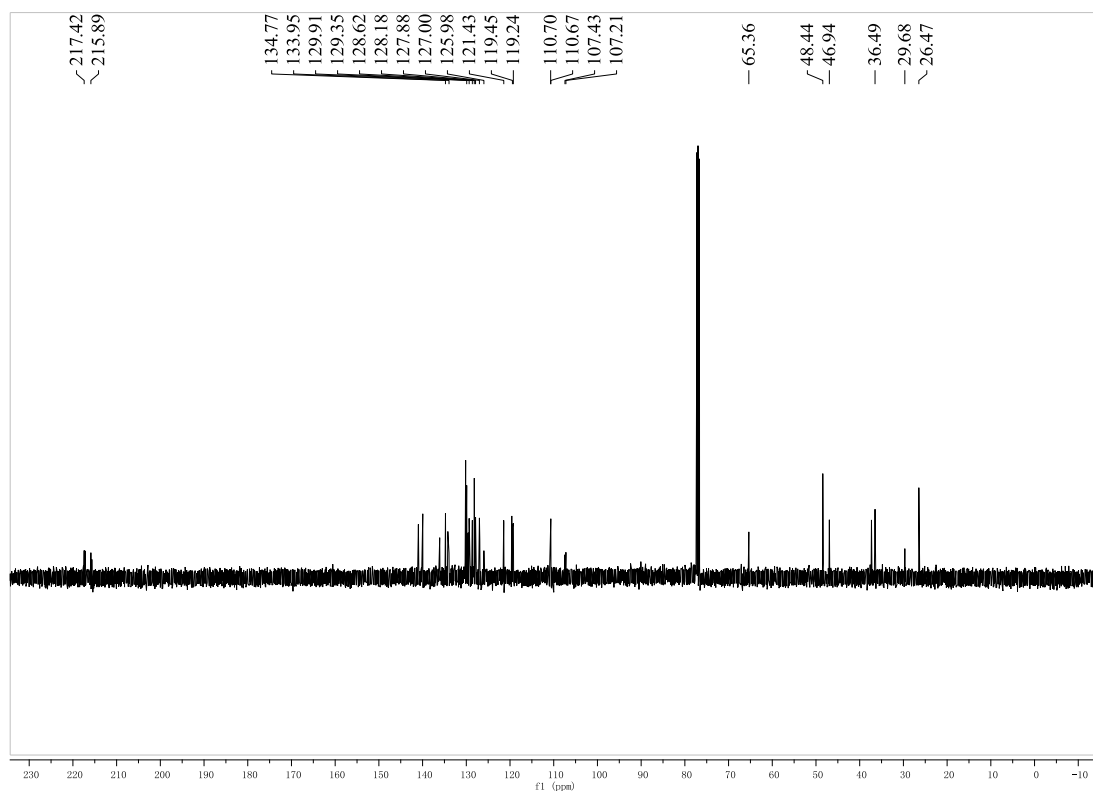




2,4-Bis(3-chlorophenyl)-1,2,4,9-tetrahydrospiro[carbazole-3,1'-cyclohexane]-2',6'-dione (5k):

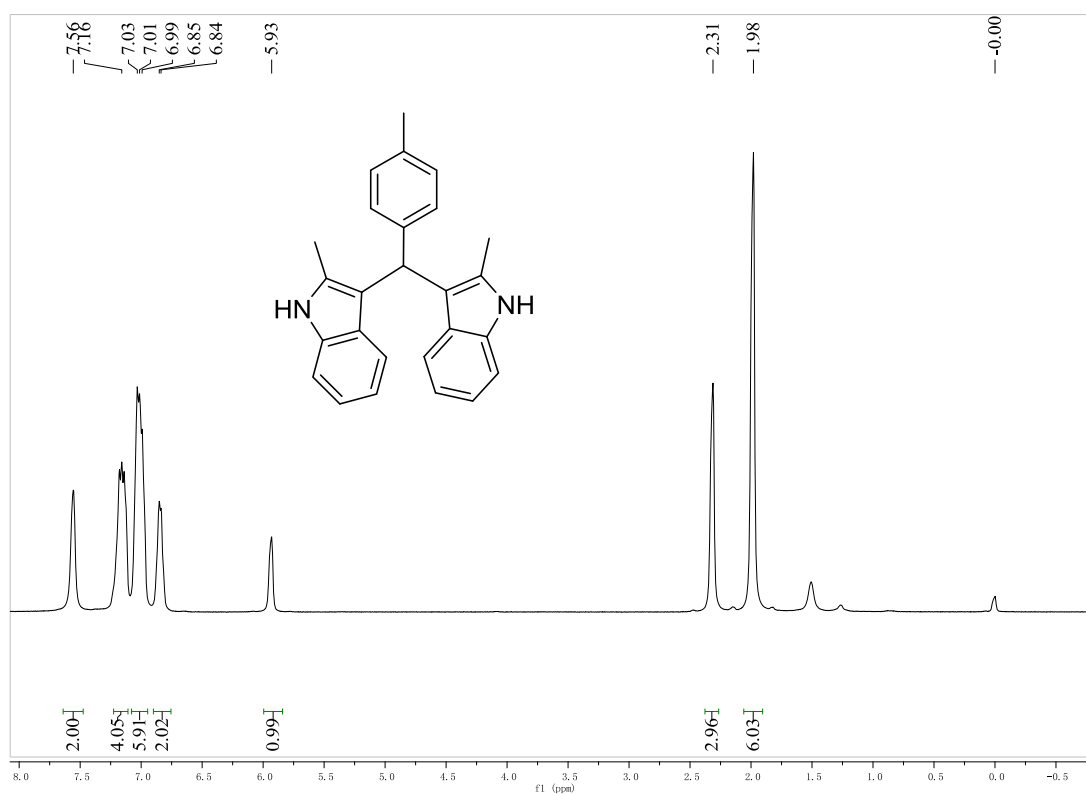
purple solid, 71%, m.p. 187-190 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.99 (s, 1H, NH), 7.32-7.26 (m, 3H, ArH), 7.25-7.19 (m, 2H, ArH), 7.15-7.04 (m, 4H, ArH), 6.89-6.79 (m, 2H, ArH), 6.35-6.34 (m, 1H, ArH), 5.10 (d, $J = 16.0$ Hz, 1H, CH), 3.79-3.77 (m, 2H, CH_2), 2.97-2.92 (m, 1H, CH), 2.13-2.05 (m, 2H, CH_2), 1.76-1.64 (m, 1H, CH), 1.53-1.48 (m, 1H, CH), 0.88-0.87 (m, 2H, CH_2); ^{13}C NMR (400 MHz, CDCl_3) δ : 217.4, 215.8, 140.9, 139.9, 136.0, 134.7, 134.2, 133.9, 130.1, 129.9, 129.5, 129.3, 128.6, 128.6, 128.2, 128.1, 128.1, 127.8, 127.7, 127.0, 126.9, 125.9, 121.4, 121.4, 119.5, 119.4, 119.4, 119.2, 110.7, 110.6, 107.4, 107.2, 65.3, 48.4, 46.9, 37.3, 36.4, 29.6, 26.4; IR (KBr) ν : 3431, 3244, 3061, 2946, 1857, 1768, 1645, 1600, 1548, 1431, 1360, 1331, 1266, 1146, 862, 792 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{29}\text{H}_{23}\text{Cl}_2^{35}\text{NO}_2$ ($[\text{M}+\text{Na}]^+$): 510.0998, found: 510.0998. $\text{C}_{29}\text{H}_{23}\text{Cl}_2^{37}\text{NO}_2$ ($[\text{M}+\text{Na}]^+$): 512.0969, found: 512.0976.

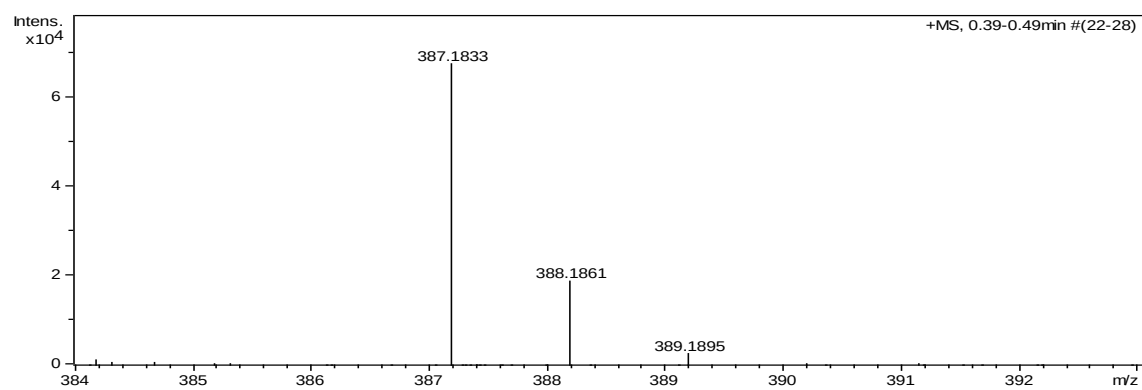
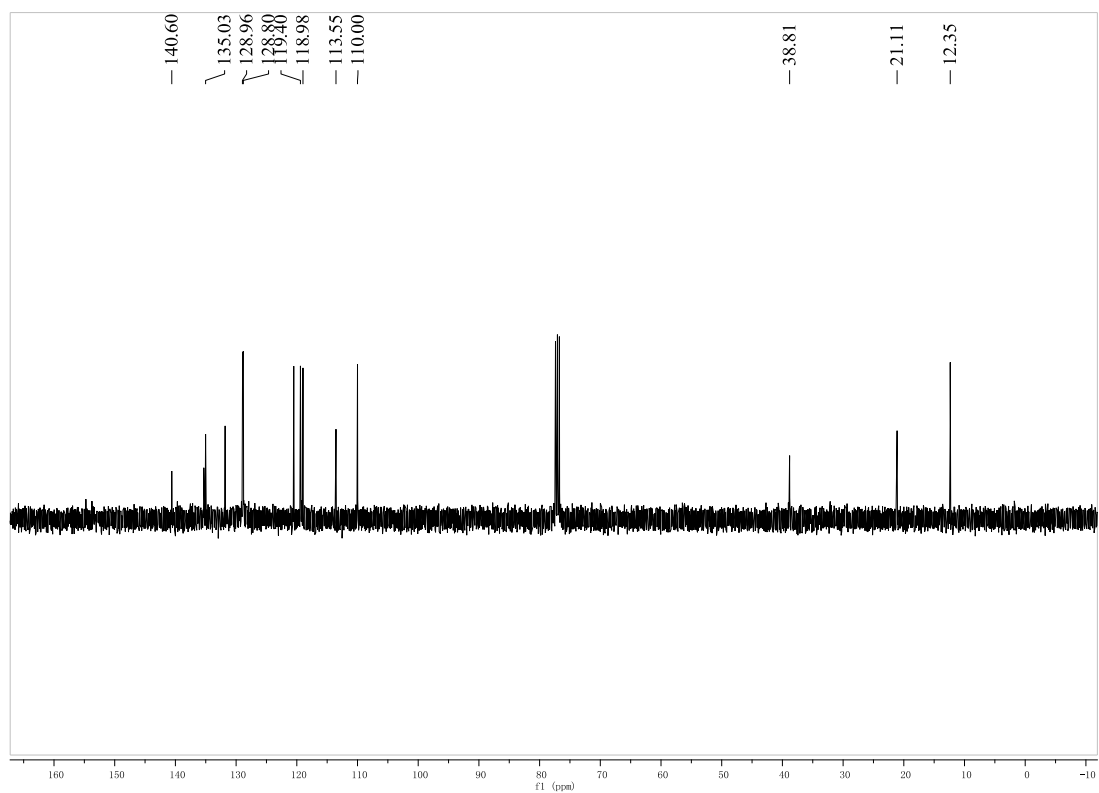




3,3'-(*p*-Tolylmethylene)bis(2-methyl-1H-indole) (6a):

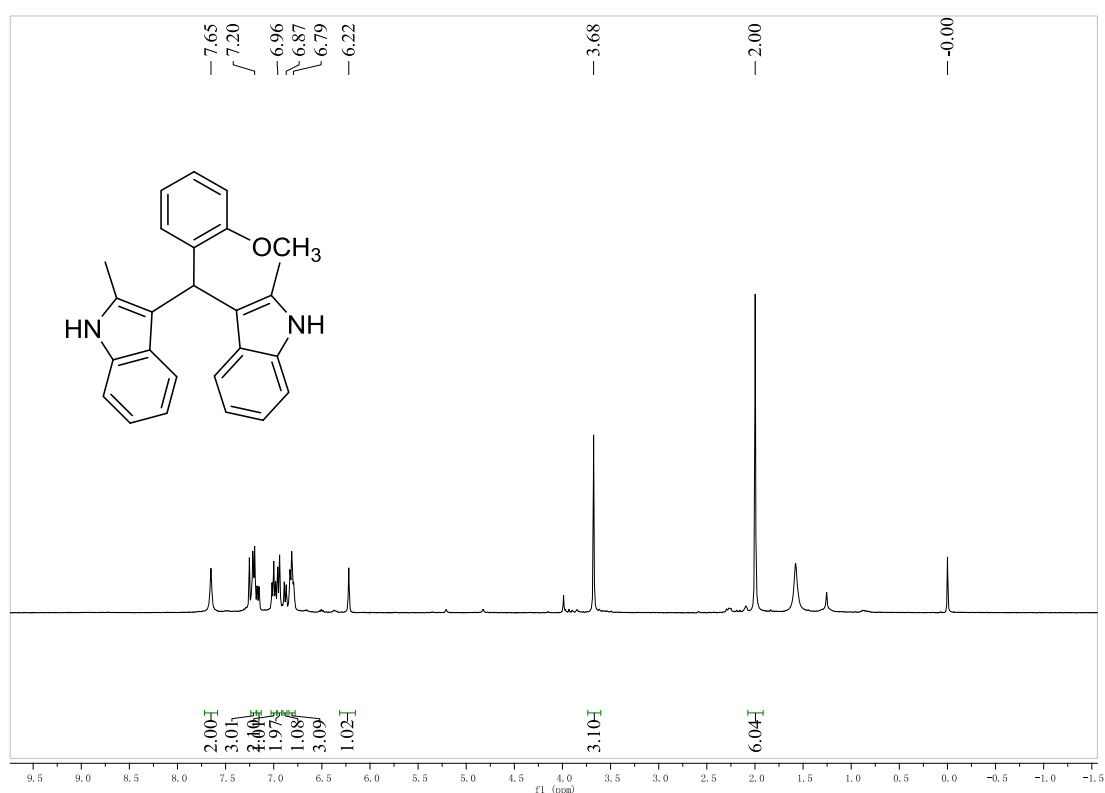
purple solid, 75%, m.p. 176-178 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.56 (s, 2H, NH), 7.17-7.14 (m, 4H, ArH), 7.03-6.99 (m, 6H, ArH), 6.85-6.84 (m, 2H, ArH), 5.93 (s, 1H, CH), 2.31 (s, 3H, CH_3), 1.98 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 140.5, 135.3, 135.0, 131.8, 128.9, 128.9, 128.8, 120.5, 119.4, 118.9, 113.5, 110.0, 38.8, 21.1, 12.3; IR (KBr) ν : 3400, 2918, 1547, 1526, 1348, 1287, 1260, 750 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{26}\text{H}_{24}\text{N}_2$ ($[\text{M}+\text{Na}]^+$): 387.1832, found: 387.1833.

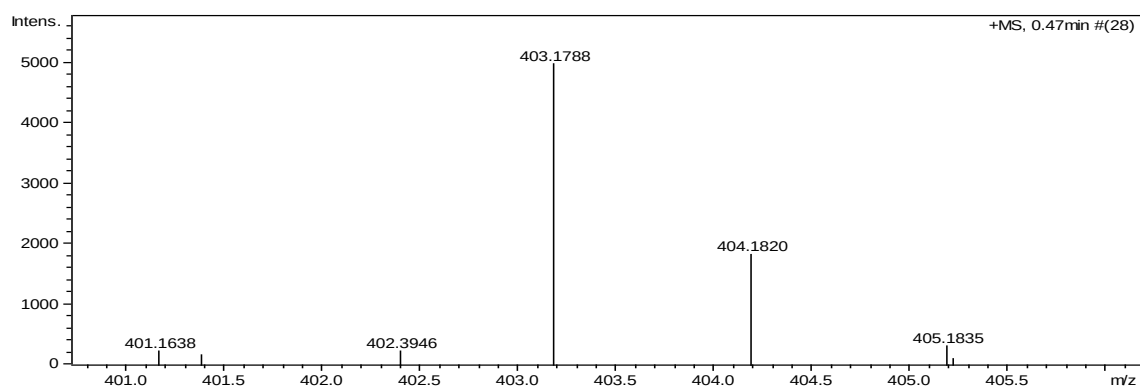
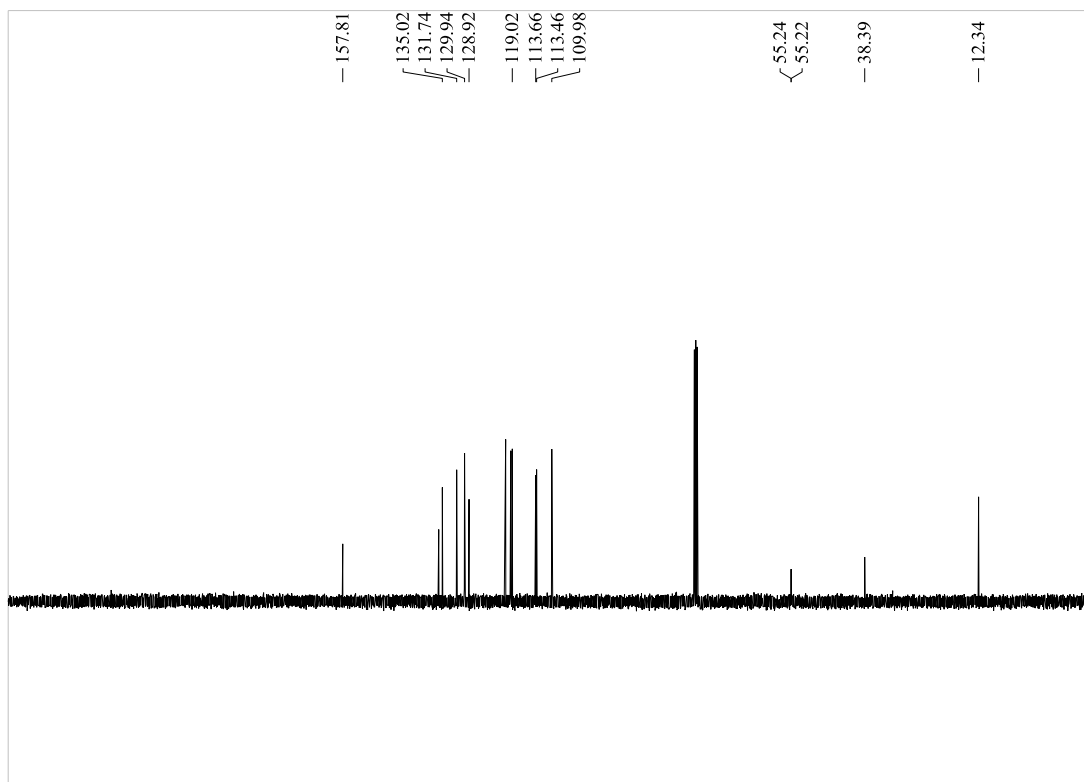




3,3'-((2-Methoxyphenyl)methylene)bis(2-methyl-1H-indole) (6b):

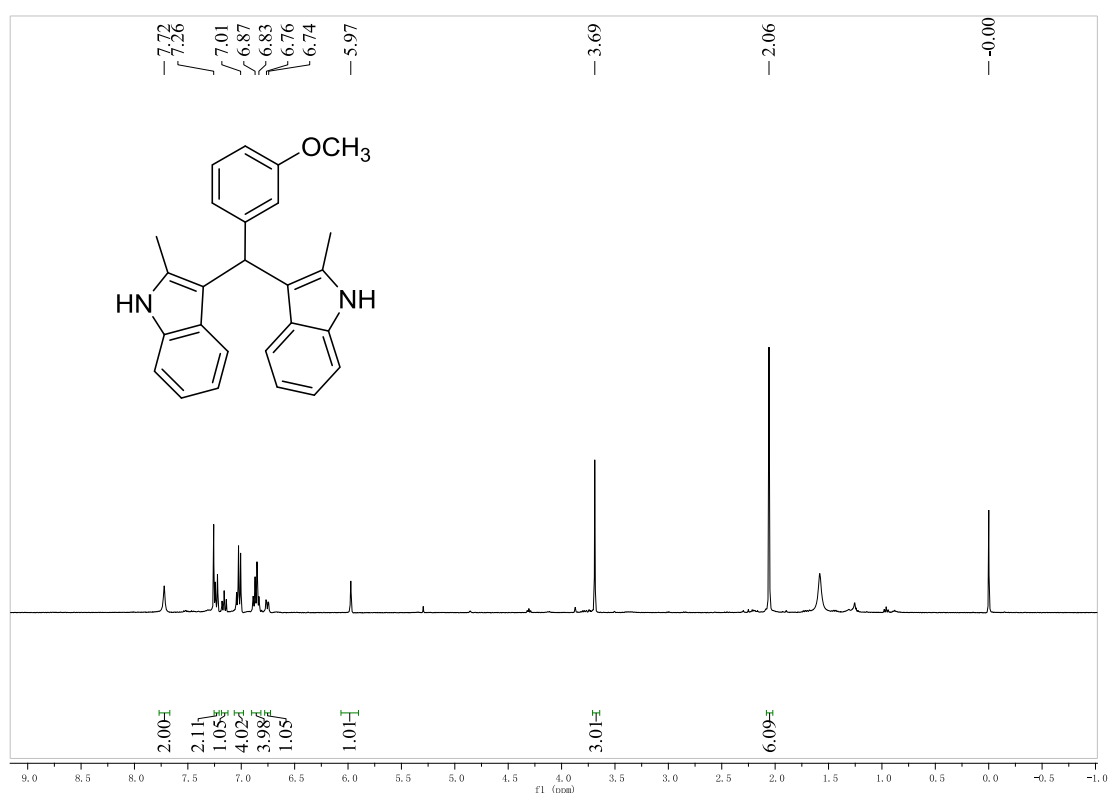
purple solid, 72%, m.p. 185-187 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.66 (s, 2H, NH), 7.20 (d, $J = 7.6$ Hz, 3H, ArH), 7.16 (d, $J = 7.6$ Hz, 1H, ArH), 7.00 (t, $J = 7.6$ Hz, 2H, ArH), 6.94 (d, $J = 8.0$ Hz, 2H, ArH), 6.88 (d, $J = 8.0$ Hz, 1H, ArH), 6.81 (d, $J = 7.6$ Hz, 3H, ArH), 6.22 (s, 1H, CH), 3.68 (s, 3H, OCH_3), 2.00 (s, 3H, CH_3), 1.99 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 157.8, 135.8, 135.0, 131.7, 129.9, 128.9, 120.5, 119.3, 119.0, 113.6, 113.4, 109.9, 55.2, 55.2, 38.3, 12.3; IR (KBr) ν : 3412, 3056, 2921, 2850, 1616, 1455, 1339, 1288, 1250, 744 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{26}\text{H}_{24}\text{N}_2\text{O}$ ($[\text{M}+\text{Na}]^+$): 403.1781, found: 403.1788.

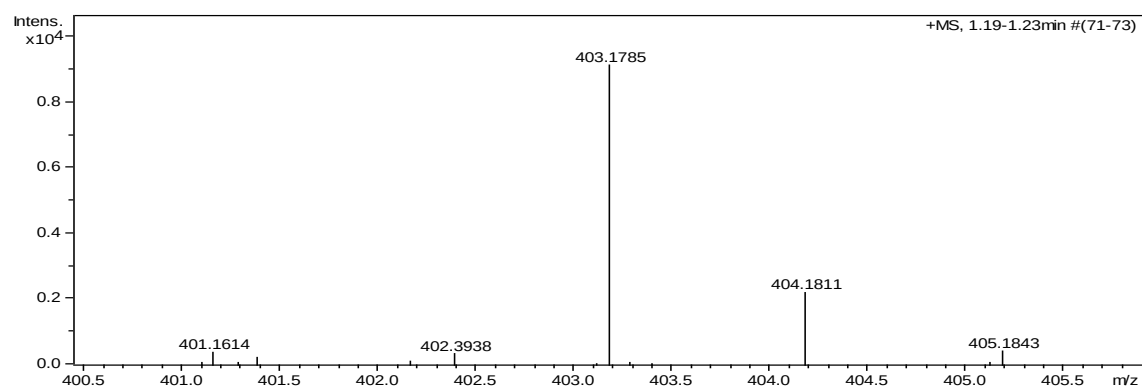
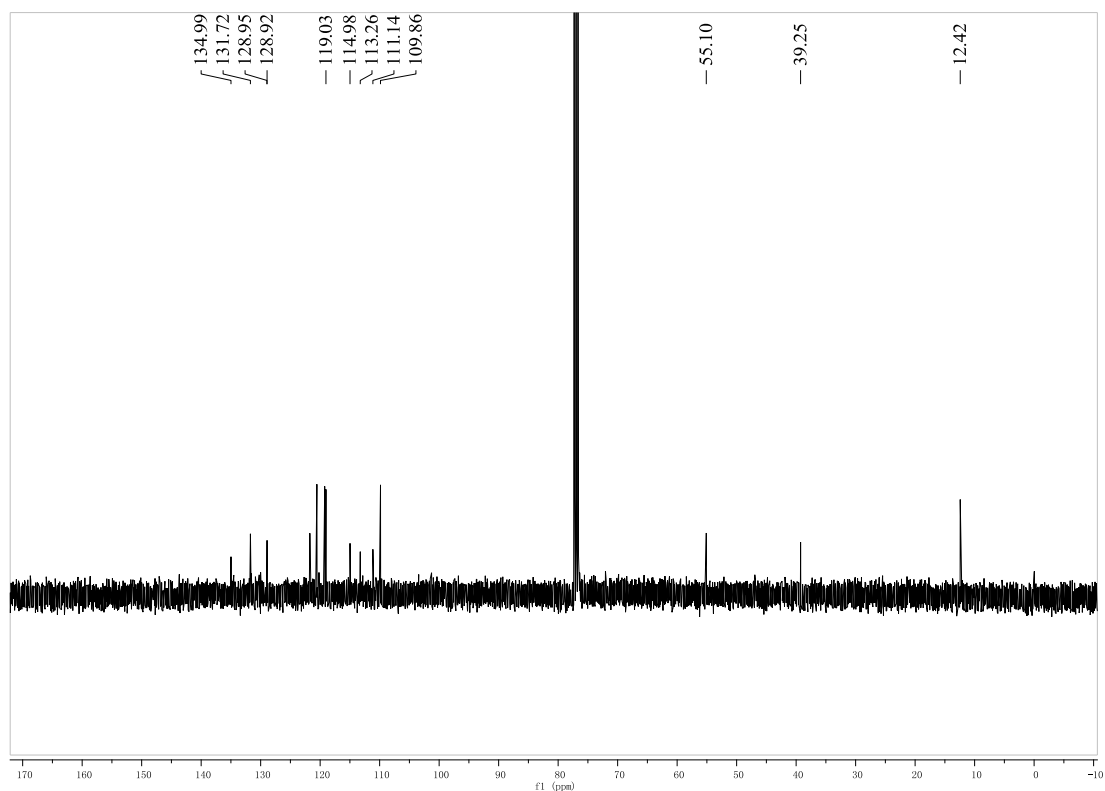




3,3'-((3-Methoxyphenyl)methylene)bis(2-methyl-1H-indole) (6c):

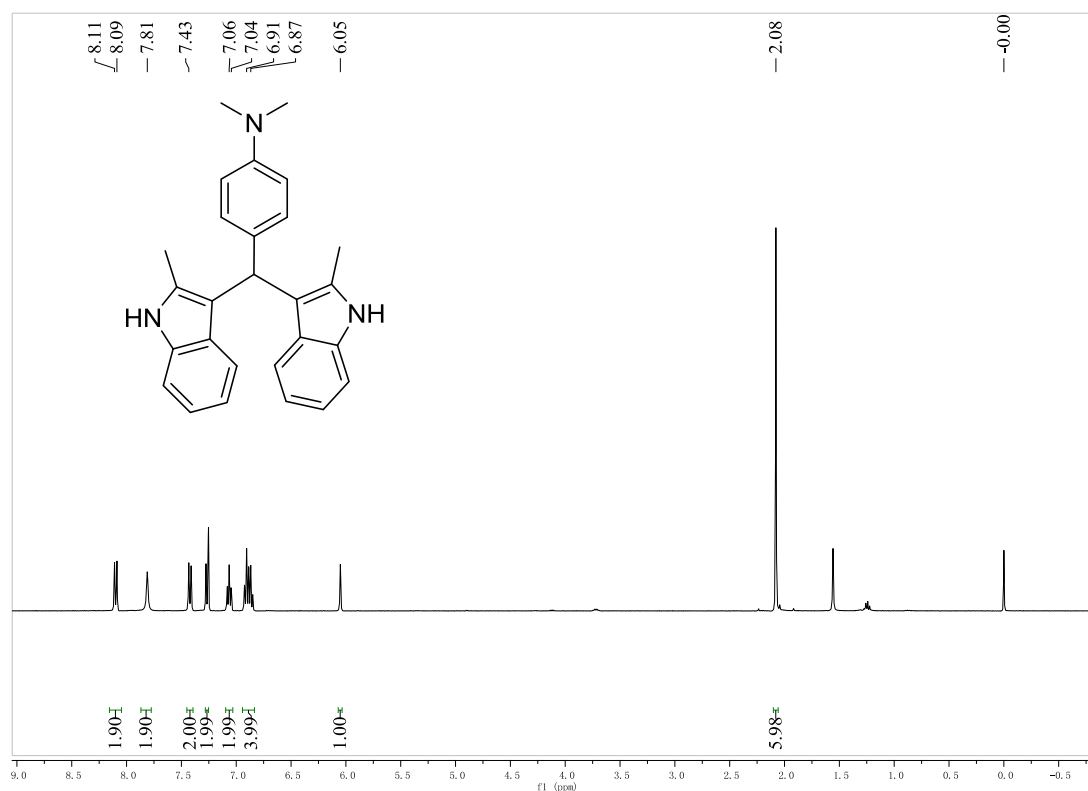
purple solid, 71%, m.p. 179-181 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.72 (s, 2H, NH), 7.23 (d, J = 8.0 Hz, 2H, ArH), 7.16 (t, J = 8.0 Hz, 1H, ArH), 7.03 (t, J = 7.6 Hz, 4H, ArH), 6.86 (q, J = 7.2 Hz, 4H, ArH), 6.75 (dd, J_1 = 8.0 Hz, J_2 = 2.4 Hz, 1H, ArH), 5.98 (s, 1H, CH), 3.69 (s, 3H, OCH_3), 2.06 (s, 3H, CH_3), 2.05 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 134.9, 131.7, 128.9, 128.9, 121.7, 120.5, 119.2, 119.0, 114.9, 113.2, 111.1, 109.8, 55.1, 39.2, 12.4; IR (KBr) ν : 3410, 3048, 2917, 2844, 1618, 1431, 1333, 1271, 1250, 745 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{26}\text{H}_{24}\text{N}_2\text{O}$ ($[\text{M}+\text{Na}]^+$): 403.1781, found: 403.1785.

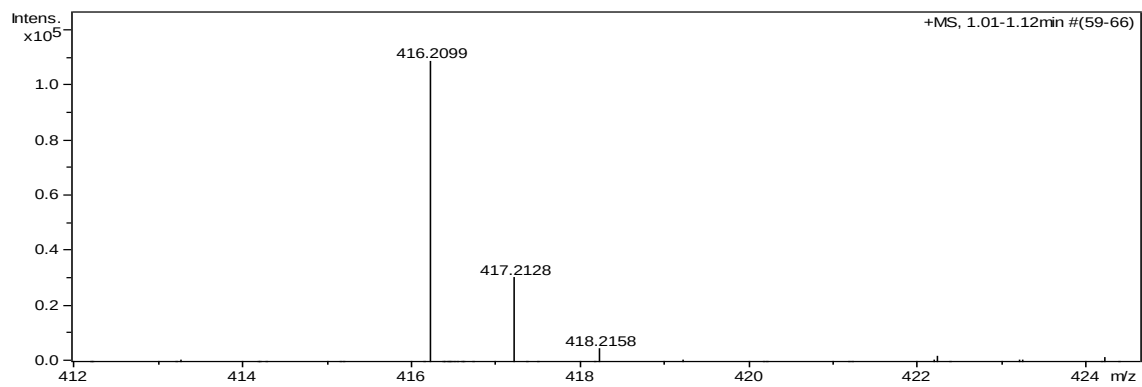
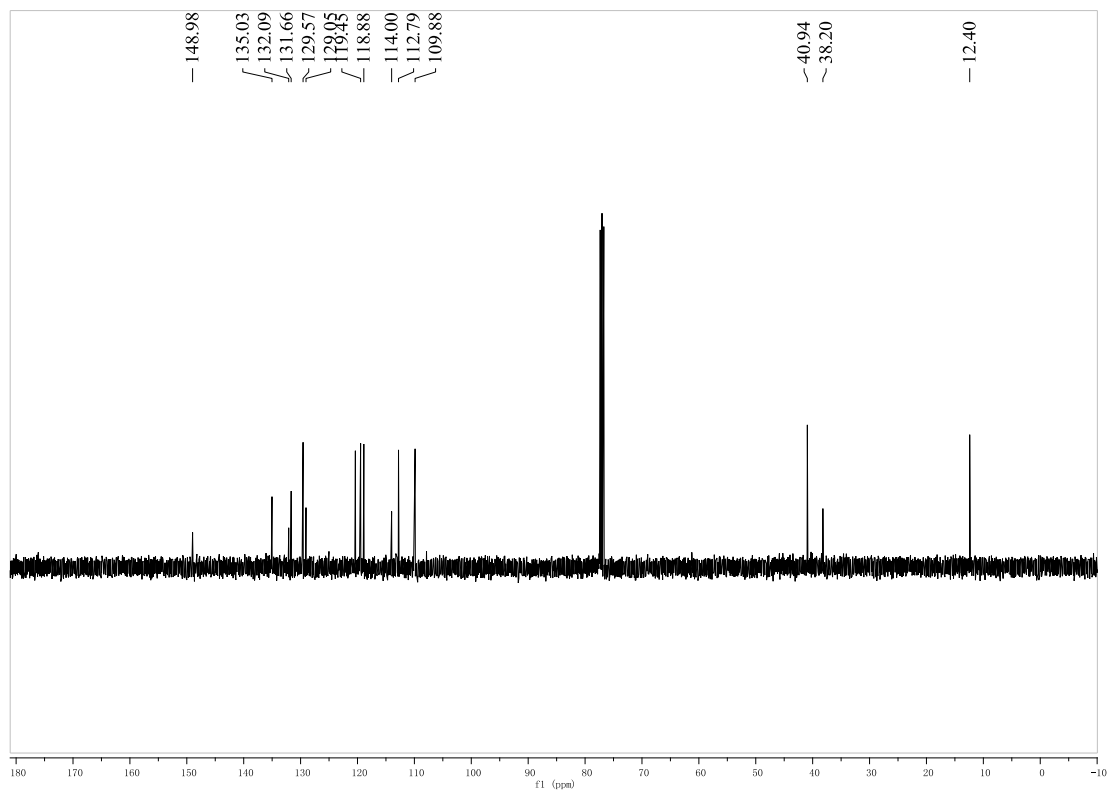




4-(Bis(2-methyl-1H-indol-3-yl)methyl)-N,N-dimethylaniline (6d):

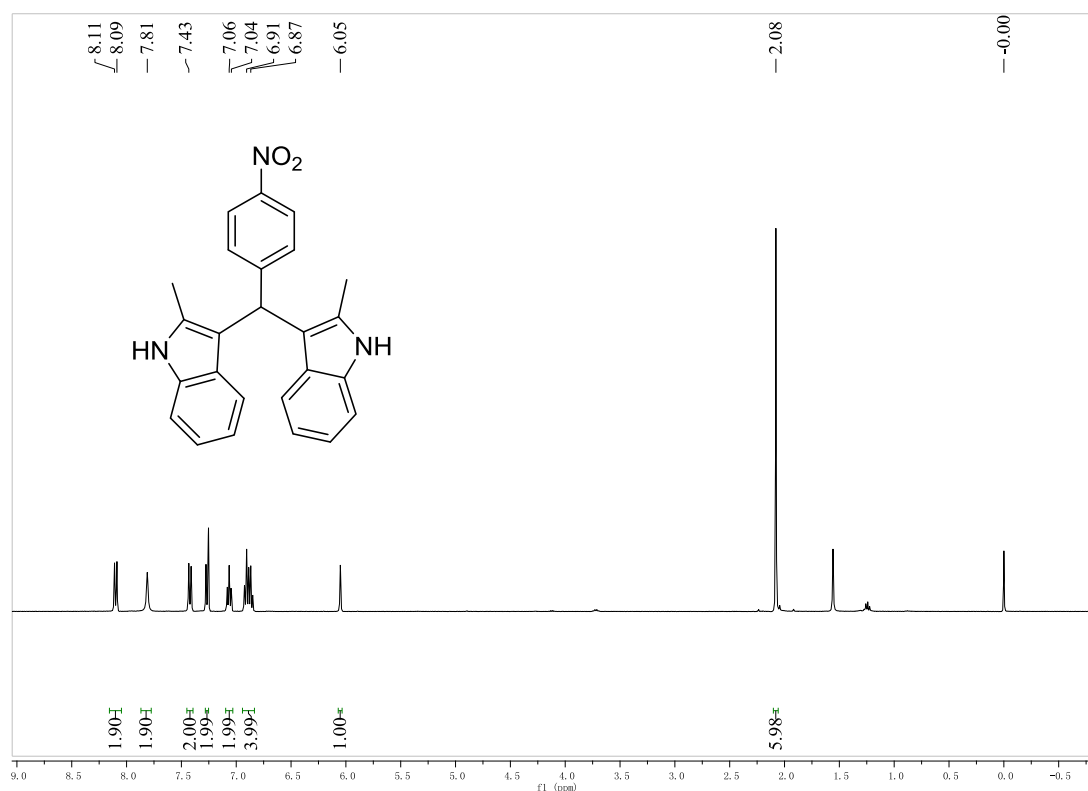
purple solid, 70%, m.p. 184-186 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.61 (s, 2H, NH), 7.18 (d, J = 8.0 Hz, 2H, ArH), 7.10 (d, J = 8.0 Hz, 2H, ArH), 7.04-6.99 (m, 4H, ArH), 6.84 (t, J = 7.2 Hz, 2H, ArH), 6.64 (d, J = 8.4 Hz, 2H, ArH), 5.90 (s, 1H, CH), 2.89 (s, 3H, CH_3), 2.88 (s, 3H, CH_3), 2.01 (s, 3H, CH_3), 2.00 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 148.9, 135.0, 132.0, 131.6, 129.5, 129.0, 120.3, 119.4, 118.8, 114.0, 112.7, 109.8, 40.9, 38.2, 12.3; IR (KBr) ν : 3387, 2918, 2771, 1275, 1270, 1260, 760, 750 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{27}\text{H}_{27}\text{N}_3$ ($[\text{M}+\text{Na}]^+$): 416.2097, found: 416.2099.

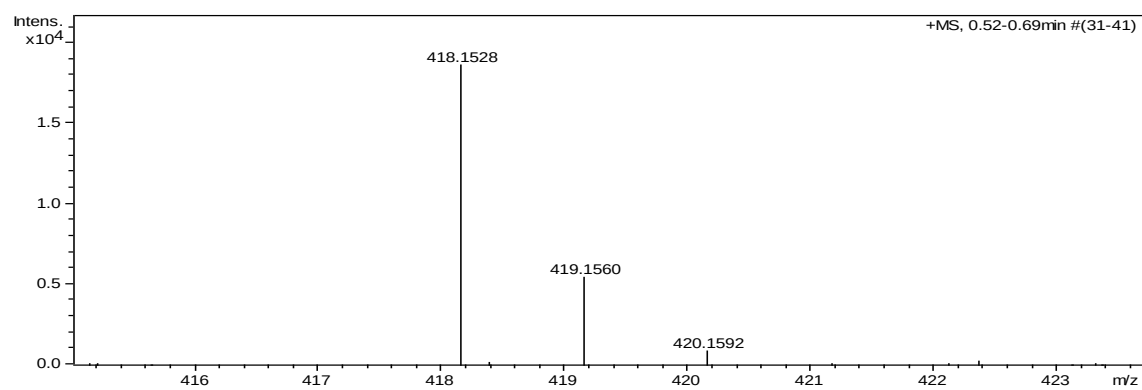
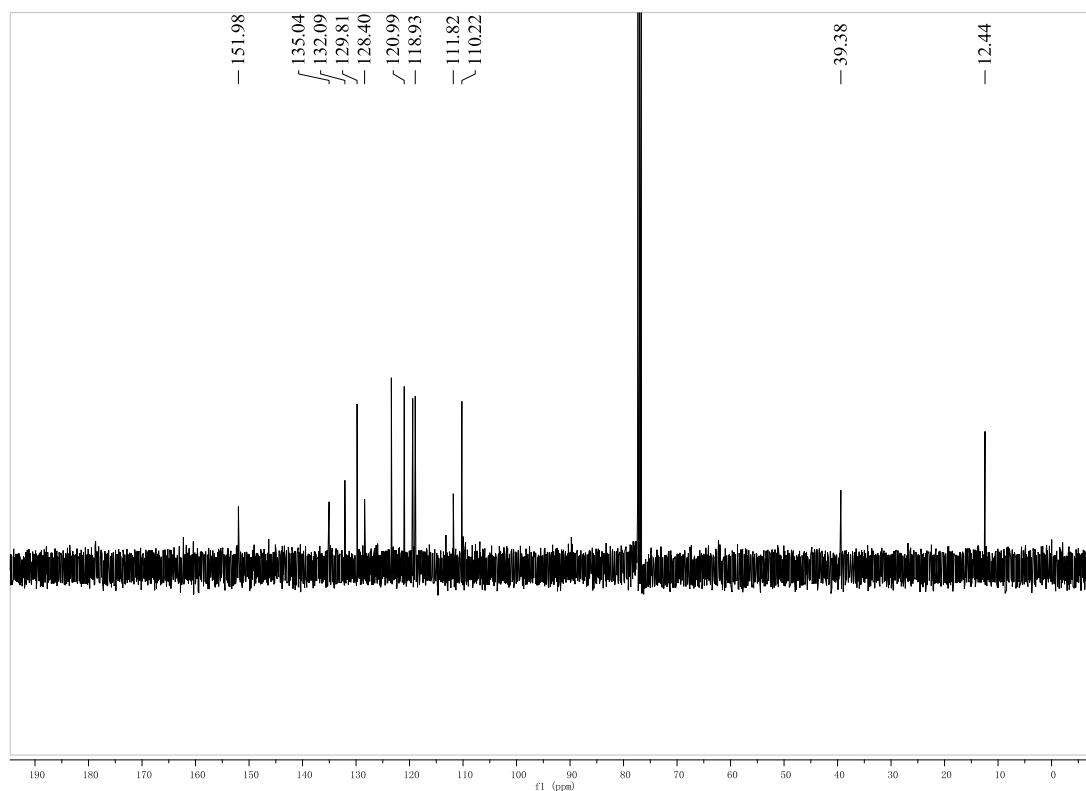




3,3'-((4-Nitrophenyl)methylene)bis(2-methyl-1H-indole) (6e):

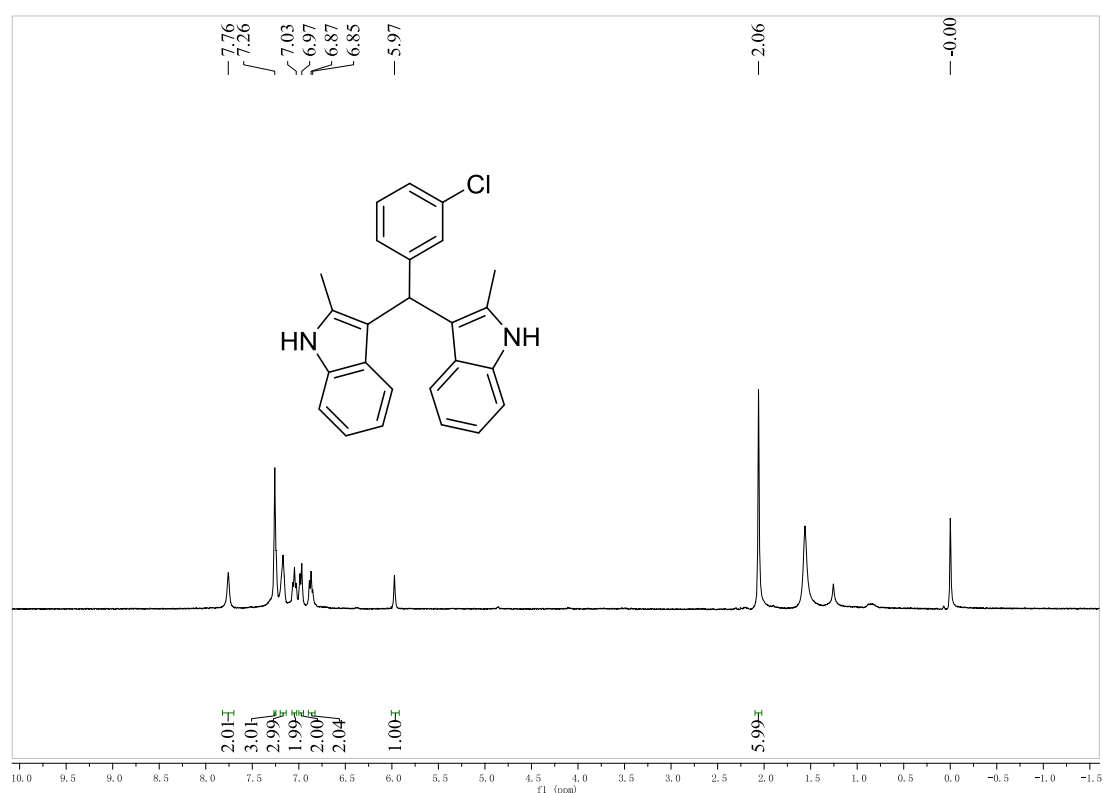
purple solid, 65%, m.p. 187-189 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.10 (d, $J = 8.8$ Hz, 2H, ArH), 7.81 (s, 2H, NH), 7.42 (d, $J = 8.4$ Hz, 2H, ArH), 7.26 (d, $J = 8.4$ Hz, 2H, ArH), 7.06 (t, $J = 8.0$ Hz, 2H, ArH), 6.93-6.87 (m, 4H, ArH), 6.05 (s, 1H, CH), 2.08 (s, 3H, CH_3), 2.07 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 151.9, 135.0, 132.0, 129.8, 128.4, 123.4, 120.9, 119.4, 118.9, 111.8, 110.2, 39.3, 12.4; IR (KBr) ν : 3393, 2919, 2878, 1645, 1548, 1525, 1459, 1345, 749 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{25}\text{H}_{21}\text{N}_3\text{O}_2$ ($[\text{M}+\text{Na}]^+$): 418.1526, found: 418.1528.

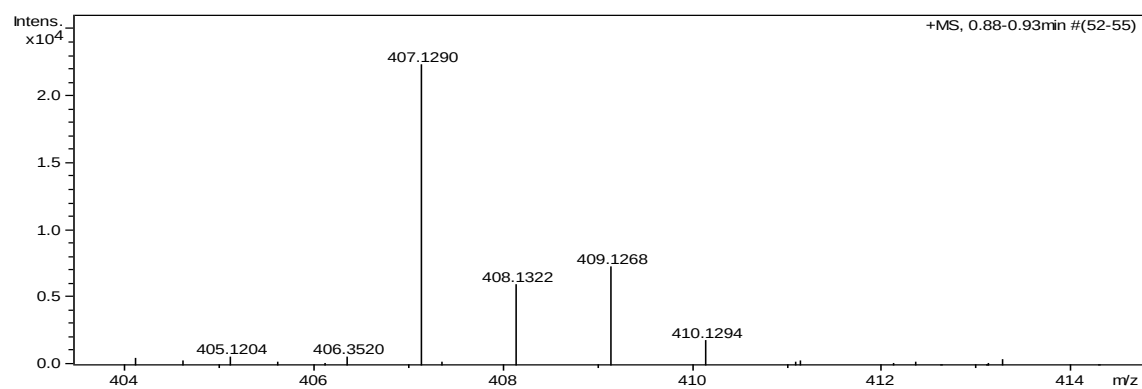
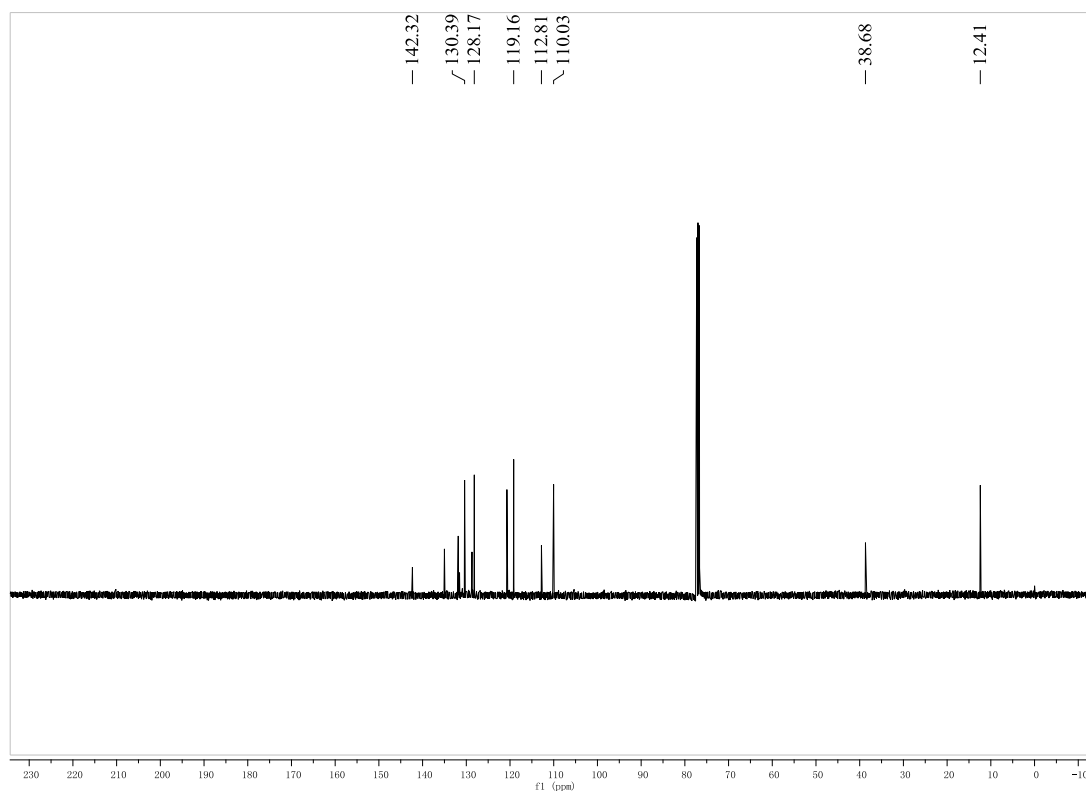




3,3'-((3-Chlorophenyl)methylene)bis(2-methyl-1H-indole) (6f):

purple solid, 70%, m.p. 175-177 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.76 (s, 2H, NH), 7.26-7.24 (m, 3H, ArH), 7.19-7.17 (m, 3H, ArH), 7.05 (t, $J = 7.2$ Hz, 2H, ArH), 6.97 (d, $J = 8.0$ Hz, 2H, ArH), 6.89-6.85 (m, 2H, ArH), 5.97 (s, 1H, CH), 2.06 (s, 3H, CH_3), 2.05 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 142.3, 135.0, 131.8, 130.3, 128.7, 128.1, 120.7, 119.1, 119.1, 112.8, 110.0, 38.6, 12.4; IR (KBr) ν : 3412, 2925, 2853, 1617, 1474, 1337, 1281, 1251, 787, 744, 787 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{25}\text{H}_{21}\text{Cl}^{35}\text{N}_2$ ($[\text{M}+\text{Na}]^+$): 407.1285, found: 407.1290. $\text{C}_{25}\text{H}_{21}\text{Cl}^{37}\text{N}_2$ ($[\text{M}+\text{Na}]^+$): 409.1256, found: 409.1268.





3,3'-((4-Chlorophenyl)methylene)bis(2-methyl-1H-indole) (6g):

purple solid, 76%, m.p. 170-172 °C; ^1H NMR (400 MHz, CDCl_3) δ : 7.74 (s, 2H, NH), 7.24 (d, J = 8.8 Hz, 3H, ArH), 7.20-7.18 (m, 3H, ArH), 7.04 (t, J = 7.2 Hz, 2H, ArH), 6.96 (d, J = 7.6 Hz, 2H, ArH), 6.86 (t, J = 7.6 Hz, 2H, ArH), 5.95 (s, 1H, CH), 2.05 (s, 3H, CH_3), 2.04 (s, 3H, CH_3); ^{13}C NMR (400 MHz, CDCl_3) δ : 142.3, 135.0, 131.8, 130.3, 128.7, 128.1, 120.7, 119.1, 112.8, 110.0, 38.6, 12.4; IR (KBr) ν : 3400, 2917, 2780, 1523, 1318, 1260, 749.8 cm^{-1} ; MS (m/z): HRMS (ESI) Calcd. for $\text{C}_{25}\text{H}_{21}\text{Cl}^{35}\text{N}_2$ ($[\text{M}+\text{Na}]^+$): 407.1285, found: 407.1287. $\text{C}_{25}\text{H}_{21}\text{Cl}^{37}\text{N}_2$ ($[\text{M}+\text{Na}]^+$): 409.1256, found: 409.1267.

