

Supporting Information

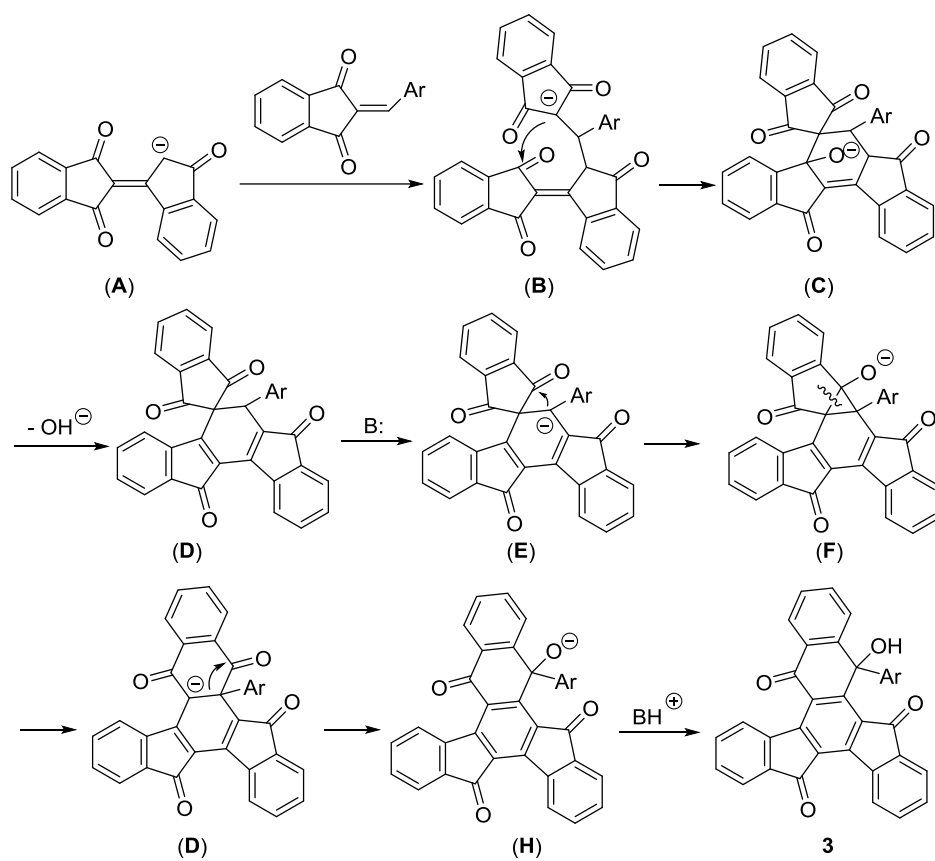
Domino Reaction of Bindone and 1,3-Dipolarophiles for Synthesis of Diverse Spiro and Fused Indeno[1,2-*a*]fluorenes

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Scheme s1 Proposed reaction mechanism for the compounds **7**

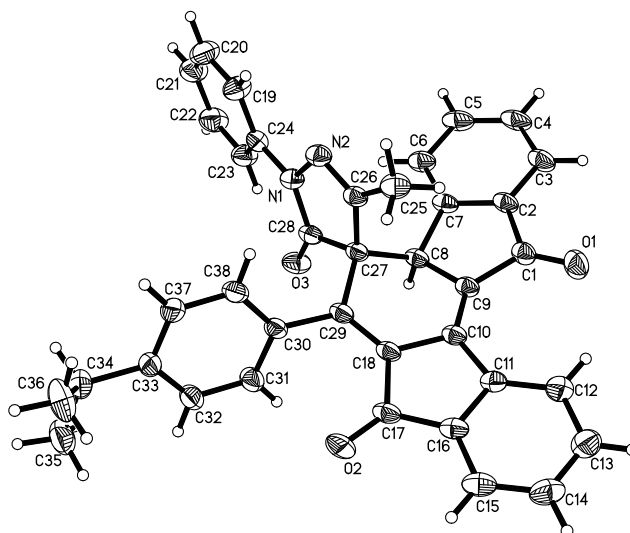


Fig. s1 ORTEP drawing (30%) of the crystal structure of **5e**

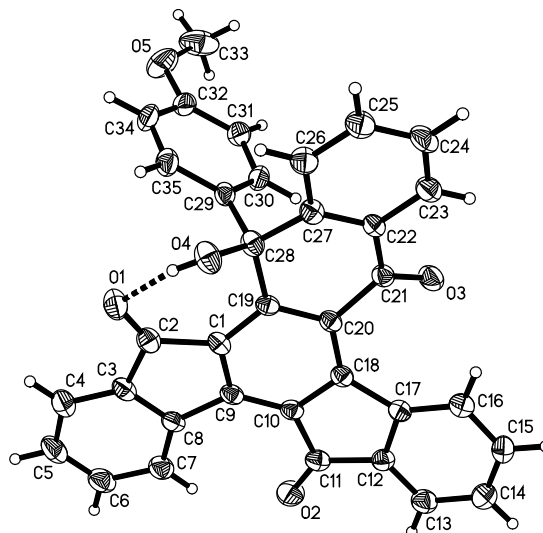


Fig. s2 ORTEP drawing (30%) of the crystal structure of **7b**

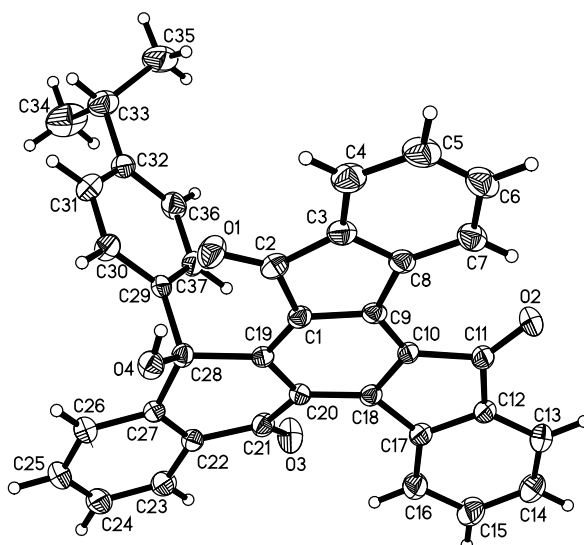


Fig. s3 ORTEP drawing (30%) of the crystal structure of **7g**

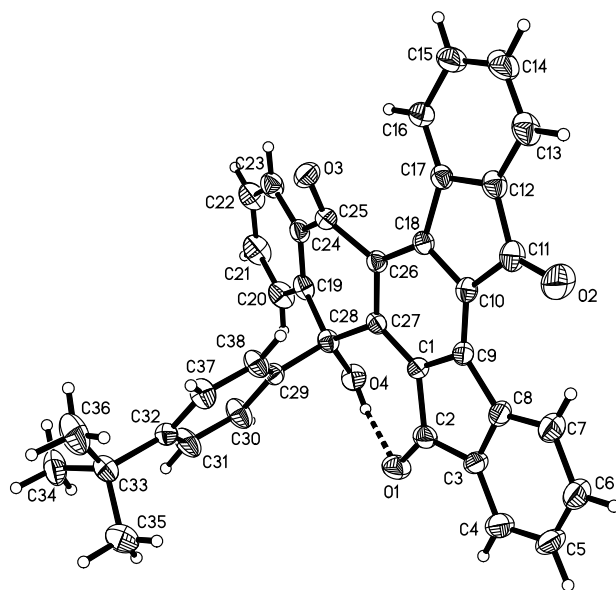


Fig. s4 ORTEP drawing (30%) of the crystal structure of **7h**

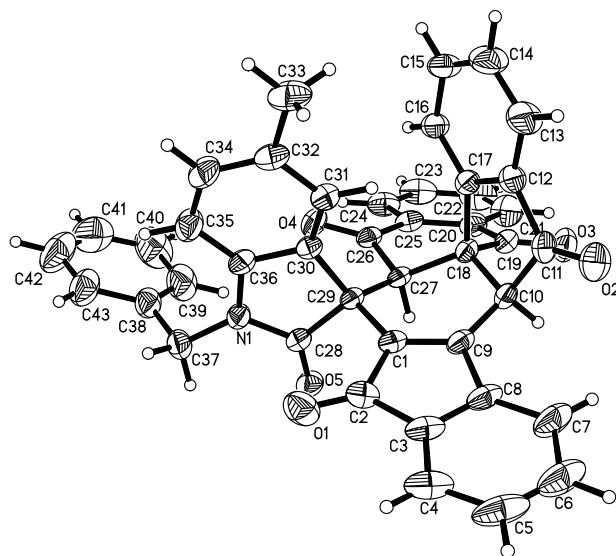


Fig. s5 ORTEP drawing (30%) of the crystal structure of **8b**

Table S1 The single crystal data of compounds **3b**, **5a**, **5e**

Phase	3b	5a	5e
Empirical formula	C ₃₇ H ₂₃ N ₃ O ₄	C ₃₅ H ₂₂ N ₂ O ₃	C ₃₈ H ₂₈ N ₂ O ₃
Formula weight	573.58	518.54	560.62
Temperature/K	296(2) K	296(2) K	296(2) K
Wavelength/ Å	0.71073	0.71073	0.71073
Crystal system	Orthorhombic	Monoclinic	Monoclinic
Space group	Pbca	P2(1)	P2(1)
<i>a</i> / Å	17.4162(16)	10.5877(14)	9.7066(6)
<i>b</i> / Å	11.9045(11)	12.8107(16)	12.5713(7)
<i>c</i> / Å	34.111(3)	18.985(3)	23.8206(14)
α (°)	90	90	90
β (°)	90	91.436(5)	100.881(2)
γ (°)	90	90	90
<i>V</i> (Å ³)	7072.2(11)	2574.2(6)	2854.4(3)
<i>Z</i>	8	4	4
Calculated density (g·cm ⁻³)	1.077	1.338	1.305
Absorption coefficient (mm ⁻¹)	0.071	0.086	0.083
<i>F</i> (000)	2384	1080	1176
θ range / (°)	2.157 to 25.998	2.146 to 25.992	2.137 to 25.998
Limiting indices	-21<= <i>h</i> <=18, -14<= <i>k</i> <=12, - 42<= <i>l</i> <=42	-13<= <i>h</i> <=12, -13<= <i>k</i> <=15, -23<= <i>l</i> <=23	-11<= <i>h</i> <=10, -15<= <i>k</i> <=15, -28<= <i>l</i> <=29
Reflections collected/unique	55584 / 6945 [<i>R</i> (int) = 0.1433]	21391 / 5023 [<i>R</i> (int) = 0.0377]	27056 / 5589 [<i>R</i> (int) = 0.0436]
Completeness to theta	99.8 %	99.2 %	99.6 %
Max. and min. transmission	0.7456 and 0.6451	0.7456 and 0.6747	0.7455 and 0.6107
Refinement method	Full-matrix least-squares on <i>F</i> ²	Full-matrix least-squares on <i>F</i> ²	Full-matrix least-squares on <i>F</i> ²
Data/restraints/parameters	6945 / 7 / 399	5023 / 0 / 362	5589 / 0 / 391
Goodness-of-fit on <i>F</i> ²	1.023	1.034	1.026
Final <i>R</i> indices[<i>I</i> >2sigma(<i>I</i>)]	<i>R</i> 1 = 0.0859, <i>wR</i> 2 = 0.2212	<i>R</i> 1 = 0.0465, <i>wR</i> 2 = 0.0943	<i>R</i> 1 = 0.0469, <i>wR</i> 2 = 0.1061
<i>R</i> indices (all data)	<i>R</i> 1 = 0.1964, <i>wR</i> 2 = 0.2761	<i>R</i> 1 = 0.0820, <i>wR</i> 2 = 0.1090	<i>R</i> 1 = 0.0807, <i>wR</i> 2 = 0.1197
Largest diff. peak and hole /(e · Å ⁻³)	0.279 and -0.164	0.202 and -0.167	0.177 and -0.156

Table S2 The single crystal data of compounds **7a**, **7b**, **7g**

Phase	7a	7b	7g
Empirical formula	C ₃₅ H ₂₀ O ₄	C ₃₅ H ₂₀ O ₅	C ₃₇ H ₂₄ O ₄
Formula weight	504.51	520.51	532.56
Temperature/K	273(2) K	296(2) K	296(2) K
Wavelength/ Å	0.71073	0.71073	0.71073
Crystal system	Triclinic	Monoclinic	Triclinic
Space group	P-1	P2(1)	P-1
<i>a</i> / Å	7.992(3)	8.5047(11)	8.2970(4)
<i>b</i> / Å	8.725(3)	27.831(4)	12.8462(7)
<i>c</i> / Å	17.432(6)	10.6483(15)	13.7948(7)
α (°)	87.327(11)	90	114.9926(16)
β (°)	79.388(12)	107.629(4)	100.881(2)
γ (°)	89.684(11)	90	93.9003(16)
<i>V</i> (Å ³)	1193.4(8)	2402.1(6)	100.7507(18)
<i>Z</i>	2	4	2
Calculated density (g·cm ⁻³)	1.404	1.439	1.370
Absorption coefficient (mm ⁻¹)	0.091	0.096	0.088
<i>F</i> (000)	524	1080	556
θ range / (°)	2.573 to 24.997	2.136 to 25.998	2.533 to 25.999
Limiting indices	-9<= <i>h</i> <=9, -10<= <i>k</i> <=10, 0<= <i>l</i> <=20	-10<= <i>h</i> <=10, -28<= <i>k</i> <=34, -13<= <i>l</i> <=12	-9<= <i>h</i> <=10, -15<= <i>k</i> <=15, - 17<= <i>l</i> <=16
Reflections collected/unique	3142 / 3142 [<i>R</i> (int) = ?]	23093 / 4711 [<i>R</i> (int) = 0.1165]	18439 / 5053 [<i>R</i> (int) = 0.0424]
Completeness to theta	89.8 %	99.8 %	99.7 %
Max. and min. transmission	0.982 and 0.978	0.7456 and 0.6772	0.7456 and 0.6842
Refinement method	Full-matrix least-squares on <i>F</i> ²	Full-matrix least-squares on <i>F</i> ²	Full-matrix least-squares on <i>F</i> ²
Data/restraints/parameters	3142 / 1 / 354	4711 / 0 / 363	5053 / 0 / 373
Goodness-of-fit on <i>F</i> ²	0.984	1.010	1.021
Final <i>R</i> indices[<i>I</i> >2σ(<i>I</i>)]	<i>R</i> 1 = 0.0651, <i>wR</i> 2 = 0.1094	<i>R</i> 1 = 0.0659, <i>wR</i> 2 = 0.1091	<i>R</i> 1 = 0.0512, <i>wR</i> 2 = 0.1148
<i>R</i> indices (all data)	<i>R</i> 1 = 0.1546, <i>wR</i> 2 = 0.1271	<i>R</i> 1 = 0.1941, <i>wR</i> 2 = 0.1460	<i>R</i> 1 = 0.0936, <i>wR</i> 2 = 0.1336
Largest diff. peak and hole /(e · Å ⁻³)	0.278 and -0.229	0.237 and -0.175	0.251 and -0.181

Table S3 The single crystal data of compounds **7h**, **8a**, **8b**

Phase	7h	8a	8b
Empirical formula	C ₃₈ H ₂₆ O ₄	C ₄₂ H ₅ NO ₅	C ₄₃ H ₂₇ NO ₅
Formula weight	546.59	623.63	637.65
Temperature/K	296(2) K	296(2) K	296(2) K
Wavelength/ Å	0.71073	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic	Monoclinic
Space group	P2(1)/c	P2(1)	P2(1)
<i>a</i> / Å	18.978(2)	12.9685(9)	13.3642(11)
<i>b</i> / Å	12.4830(13)	13.8484(11)	14.9517(12)
<i>c</i> / Å	11.7028(12)	20.7097(16)	20.5922(15)
α (°)	90	90	90
β (°)	94.907(3)	93.367(2)	90.279(2)
γ (°)	90	90	90
<i>V</i> (Å ³)	2762.2(5)	3712.9(5)	4114.6(6)
<i>Z</i>	4	4	4
Calculated density (g·cm ⁻³)	1.314	1.116	1.029
Absorption coefficient (mm ⁻¹)	0.084	0.073	0.067
<i>F</i> (000)	1144	1296	1328
θ range / (°)	2.154 to 25.999	1.970 to 25.997	2.044 to 26.000
Limiting indices	-20<= <i>h</i> <=23, -15<= <i>k</i> <=15, -14<= <i>l</i> <=13	-15<= <i>h</i> <=15, -14<= <i>k</i> <=17, -25<= <i>l</i> <=23	-16<= <i>h</i> <=16, -18<= <i>k</i> <=16, -25<= <i>l</i> <=24
Reflections collected/unique	22520 / 5377 [<i>R</i> (int) = 0.0858]	34303 / 7270 [<i>R</i> (int) = 0.0692]	33607 / 8083 [<i>R</i> (int) = 0.1086]
Completeness to theta	99.0 %	99.8 %	99.6 %
Max. and min. transmission	0.7456 and 0.6101	0.7456 and 0.6399	0.7456 and 0.6410
Refinement method	Full-matrix least-squares on <i>F</i> ²	Full-matrix least-squares on <i>F</i> ²	Full-matrix least-squares on <i>F</i> ²
Data/restraints/parameters	5377 / 84 / 414	7270 / 0 / 433	8083 / 0 / 443
Goodness-of-fit on <i>F</i> ²	1.006	1.034	0.975
Final <i>R</i> indices[<i>I</i> >2σ(<i>I</i>)]	<i>R</i> 1 = 0.0588, <i>wR</i> 2 = 0.1190	<i>R</i> 1 = 0.0595, <i>wR</i> 2 =	<i>R</i> 1 = 0.0622, <i>wR</i> 2 = 0.1425
<i>R</i> indices (all data)	<i>R</i> 1 = 0.1616, <i>wR</i> 2 = 0.1569	<i>R</i> 1 = 0.1350, <i>wR</i> 2 =	<i>R</i> 1 = 0.1722, <i>wR</i> 2 = 0.1831
Largest diff. peak and hole /(e · Å ⁻³)	0.187 and -0.222	0.159 and -0.211	0.174 and -0.154

Experimental section

1. General procedure for the reactions of bindone and isatylidene malononitriles: A mixture of bindane (1.0 mmol), isatylidene malononitrile (1.0 mmol) and triethylamine (2.0 mmol) in dry dichloromethane (20.0 mL) was stirred at room temperature for twelve hours. After removing the solvent, the residue was recrystallized in alcohol to give the pure product **3a-3j** for analysis.

3'-amino-1''-benzyl-1,2'',3,9'-tetraoxo-1,3-dihydro-9'H-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carbonitrile (3a): yellow solid, 0.514 g, 92%, m.p. 292-294 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 8.38-8.25 (m, 4H), 7.55 (d, *J* = 7.6 Hz, 2H), 7.45 (d, *J* = 7.2 Hz, 1H), 7.39-7.10 (m, 8H), 6.92 (d, *J* = 8.0 Hz, 1H), 6.54 (s, 2H), 5.94 (d, *J* = 7.2 Hz, 1H), 5.08 (d, *J* = 16.0 Hz, 1H), 5.03 (d, *J* = 16.0 Hz, 1H); ¹³C{¹H} NMR (100 MHz, DMSO-*d*₆) δ: 195.5, 195.3, 191.8, 174.8, 152.0, 150.1, 143.7, 143.4, 142.4, 140.1, 139.0, 138.8, 136.2, 135.2, 133.2, 130.7, 130.5, 129.9, 128.9, 127.8, 127.7, 125.9, 125.6, 124.0, 123.6, 120.6, 117.2, 110.3, 77.6, 61.6, 49.8, 44.1; HRMS (ESI-TOF) Calcd. For C₃₆H₂₂N₃O₄ ([M+H]⁺): 560.1605, found: 560.1618.

3'-amino-1''-benzyl-5''-methyl-1,2'',3,9'-tetraoxo-1,3-dihydro-9'H-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carbonitrile (3b): yellow solid, 0.532 g, 93%, m.p. 291-293 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 8.37-8.27 (m, 4H), 7.54 (d, *J* = 6.8 Hz, 2H), 7.38-7.30 (m, 5H), 7.24 (t, *J* = 8.0 Hz, 1H), 7.18 (t, *J* = 7.6 Hz, 1H), 7.08 (d, *J* = 8.0 Hz, 1H), 6.79 (d, *J* = 8.0 Hz, 1H), 6.52 (s, 2H), 5.94 (d, *J* = 7.2 Hz, 1H), 5.05 (d, *J* = 16.0 Hz, 1H), 5.00 (d, *J* = 16.0 Hz, 1H), 2.27 (s, 3H); ¹³C{¹H} NMR (100 MHz, DMSO-*d*₆) δ: 195.5, 195.3, 191.8, 174.67, 151.8, 150.0, 143.7, 142.3, 141.0, 140.1, 139.0, 138.8, 136.3, 135.2, 133.2, 132.6, 130.7, 130.6, 130.0, 129.9, 128.9, 127.8, 127.6, 125.9, 125.6, 124.5, 124.0, 120.6, 117.2, 110.1, 77.8, 61.6, 49.8, 44.1, 21.2; HRMS (ESI-TOF) Calcd. For C₃₇H₂₄N₃O₄ ([M+H]⁺): 574.1761, found: 574.1771.

3'-amino-1''-benzyl-5''-chloro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'H-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carbonitrile (3c): yellow solid, 0.539 g, 91%, m.p. 293-295 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 8.38-8.26 (m, 4H), 7.55-7.47 (m, 3H), 7.40-7.29 (m, 5H), 7.25 (t, *J* = 7.6 Hz, 1H), 7.19 (t, *J* = 7.6 Hz, 1H), 6.96 (d, *J* = 7.6 Hz, 1H), 6.68 (s, 2H), 5.97 (d, *J* = 7.2 Hz, 1H), 5.09 (d, *J* = 16.0 Hz, 1H), 5.05 (d, *J* = 16.0 Hz, 1H); ¹³C{¹H} NMR (100 MHz, DMSO-*d*₆) δ: 195.3, 195.2, 191.7, 174.5, 152.4, 150.6, 143.8, 142.3, 142.2, 139.9, 139.2, 138.8, 135.9, 135.3, 132.6, 132.5, 130.8, 129.9, 129.8, 129.0, 127.9, 127.7, 127.6, 126.0, 125.7, 124.2, 124.1, 120.8, 117.0, 112.0, 76.6, 61.6, 49.8, 44.1; HRMS (ESI-TOF) Calcd. For C₃₆H₂₁ClN₃O₄ ([M+H]⁺):

594.1215, found: 594.1214.

3'-amino-1''-benzyl-5''-fluoro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'H-dispiro[indene-2,4'-

fluorene-1',3''-indoline]-2'-carbonitrile (3d): yellow solid, 0.553 g, 96%, m.p. 233-235 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 8.39-8.25 (m, 4H), 7.55 (d, *J* = 7.2 Hz, 2H), 7.40-7.15 (m, 8H), 6.94 (s, 1H), 6.65 (s, 2H), 5.97 (d, *J* = 7.2 Hz, 1H), 5.09 (d, *J* = 16.0 Hz, 1H), 5.04 (d, *J* = 16.0 Hz, 1H); ¹³C{¹H} NMR (100 MHz, DMSO-*d*₆) δ: 195.3, 195.2, 191.6, 174.7, 159.9 (d, *J* = 238.4 Hz), 152.3, 150.5, 143.8, 142.2, 139.9, 139.6, 139.0 (d, *J* = 30.5 Hz), 136.0, 135.2, 132.6, 132.4, 132.3, 130.8, 129.8, 128.9, 127.9, 127.7, 126.0, 125.7, 124.2, 120.8, 117.0, 116.3 (d, *J* = 23.6 Hz), 111.8, 111.4 (d, *J* = 8.0 Hz), 76.8, 61.6, 56.5, 50.0, 44.2, 19.0; HRMS (ESI-TOF) Calcd. For C₃₆H₂₁FN₃O₄ ([M+H]⁺): 578.1511, found: 578.1519.

3'-amino-1''-butyl-5''-methyl-1,2'',3,9'-tetraoxo-1,3-dihydro-9'H-dispiro[indene-2,4'-

fluorene-1',3''-indoline]-2'-carbonitrile (3e): yellow solid, 0.495 g, 92%, m.p. 288-290 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 8.36-8.26 (m, 4H), 7.30 (d, *J* = 7.2 Hz, 1H), 7.24-7.14 (m, 4H), 7.08 (d, *J* = 8.0 Hz, 1H), 6.45 (s, 2H), 5.91 (d, *J* = 7.6 Hz, 1H), 3.78-3.74 (m, 2H), 2.30 (s, 3H), 1.71-1.64 (m, 2H), 1.48-1.42 (m, 2H), 0.94 (t, *J* = 7.6 Hz, 3H); ¹³C{¹H} NMR (100 MHz, DMSO-*d*₆) δ: 195.6, 195.4, 191.7, 174.2, 151.6, 149.7, 143.6, 142.3, 141.4, 140.1, 138.9, 138.7, 135.1, 133.4, 132.1, 130.6, 130.5, 130.1, 129.9, 125.9, 125.5, 124.5, 123.9, 120.5, 117.0, 109.5, 77.9, 61.5, 49.6, 29.4, 21.2, 19.9, 14.2; HRMS (ESI-TOF) Calcd. For C₃₄H₂₆N₃O₄ ([M+H]⁺): 540.1918, found: 540.1934.

3'-amino-1''-butyl-5''-chloro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'H-dispiro[indene-2,4'-

fluorene-1',3''-indoline]-2'-carbonitrile (3f): yellow solid, 0.503 g, 90%, m.p. 210-212 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 8.38-8.25 (m, 4H), 7.47-7.45 (m, 2H), 7.33 (d, *J* = 7.2 Hz, 1H), 7.29-7.15 (m, 3H), 6.62 (s, 2H), 5.95 (d, *J* = 7.2 Hz, 1H), 3.84-3.77 (m, 2H), 1.72-1.64 (m, 2H), 1.50-1.40 (m, 2H), 0.95 (t, *J* = 7.2 Hz, 3H); ¹³C{¹H} NMR (100 MHz, DMSO-*d*₆) δ: 195.3, 195.2, 191.6, 174.1, 152.2, 150.2, 143.8, 142.8, 142.2, 139.9, 139.1, 138.8, 135.2, 132.6, 132.6, 130.8, 129.8, 127.2, 125.9, 125.7, 124.1, 124.0, 120.7, 116.8, 111.5, 76.7, 61.6, 49.6, 29.4, 19.9, 14.1; HRMS (ESI-TOF) Calcd. For C₃₃H₂₃ClN₃O₄ ([M+H]⁺): 560.1372, found: 560.1381.

3'-amino-1''-butyl-5''-fluoro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'H-dispiro[indene-2,4'-

fluorene-1',3''-indoline]-2'-carbonitrile (3g): yellow solid, 0.472 g, 87%, m.p. 280-282 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 8.38-8.24 (m, 4H), 7.32 (d, *J* = 7.2 Hz, 1H), 7.28-7.21 (m, 4H), 7.17

(t, $J = 7.2$ Hz, 1H), 6.58 (s, 2H), 5.95 (d, $J = 7.6$ Hz, 1H), 3.82-3.78 (m, 2H), 1.72-1.65 (m, 2H), 1.50-1.41 (m, 2H), 0.95 (t, $J = 7.6$ Hz, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO- d_6) δ : 195.4, 195.3, 191.5, 174.2, 159.1 (d, $J = 237.6$ Hz), 152.1, 150.2, 143.7, 142.1, 140.0, 139.9, 138.9 (d, $J = 30.3$ Hz), 135.1, 132.7, 132.4, 132.3, 130.7, 129.8, 125.9, 125.7 (d, $J = 32.5$ Hz), 120.7, 116.8, 116.3 (d, $J = 22.8$ Hz), 111.6, 111.4, 110.9 (d, $J = 7.6$ Hz), 76.9, 61.5, 49.8, 29.4, 19.9, 14.2.; HRMS (ESI-TOF) Calcd. For $\text{C}_{33}\text{H}_{23}\text{FN}_3\text{O}_4$ ($[\text{M}+\text{H}]^+$): 544.1667, found: 544.1682.

Ethyl 3'-amino-1''-benzyl-5''-methyl-1,2'',3,9'-tetraoxo-1,3-dihydro-9'*H*-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carboxylate (3h): purple solid, 0.527 g, 85%, m.p. 281-283 °C; ^1H NMR (400 MHz, DMSO- d_6) δ : 8.40-8.29 (m, 4H), 7.65 (d, $J = 7.2$ Hz, 2H), 7.49 (s, 1H), 7.41 (t, $J = 6.0$ Hz, 2H), 7.33-7.28 (m, 3H), 7.22-7.12 (m, 3H), 7.01 (d, $J = 8.0$ Hz, 1H), 6.84 (d, $J = 7.6$ Hz, 1H, NH), 5.90 (d, $J = 7.2$ Hz, 1H, NH), 5.03 (d, $J = 15.6$ Hz, 1H), 4.84 (d, $J = 15.6$ Hz, 1H), 3.82-3.77 (m, 1H), 3.46-3.42 (m, 1H), 2.23 (s, 3H), 0.53 (t, $J = 7.2$ Hz, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO- d_6) δ : 196.3, 195.9, 191.9, 176.9, 167.9, 153.3, 147.8, 143.6, 142.7, 142.5, 139.9, 139.1, 138.9, 137.2, 134.9, 133.2, 131.2, 130.3, 129.8, 128.8, 128.7, 128.7, 127.8, 125.8, 125.6, 123.7, 123.6, 120.2, 109.0, 94.3, 62.3, 59.5, 49.8, 46.2, 21.2, 13.9, 9.1; HRMS (ESI-TOF) Calcd. For $\text{C}_{39}\text{H}_{28}\text{N}_2\text{NaO}_6$ ($[\text{M}+\text{Na}]^+$): 643.1840, found: 643.1842.

Ethyl 3'-amino-1''-benzyl-5''-chloro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'*H*-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carboxylate (3i): purple solid, 0.524 g, 82%, m.p. 234-236 °C; ^1H NMR (400 MHz, DMSO- d_6) δ : 8.80 (d, $J = 7.2$ Hz, 1H), 8.42 (s, 1H), 7.57-7.55 (m, 2H), 7.48 (t, $J = 7.6$ Hz, 1H), 7.34-7.28 (m, 2H), 7.22-7.04 (m, 5H), 6.92 (t, $J = 7.6$ Hz, 2H), 6.67 (d, $J = 8.4$ Hz, 1H), 6.35 (d, $J = 7.2$ Hz, 1H), 6.34 (s, 2H), 4.80 (d, $J = 16.0$ Hz, 1H), 4.40 (d, $J = 16.0$ Hz, 1H), 3.92-3.86 (m, 1H), 3.67-3.62 (m, 1H), 0.65 (t, $J = 7.2$ Hz, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO- d_6) δ : 183.7, 180.7, 168.9, 167.8, 166.9, 151.5, 145.3, 142.4, 142.1, 140.0, 138.7, 135.9, 130.5, 130.1, 130.0, 128.7, 127.2, 126.3, 125.6, 125.2, 125.1, 124.0, 121.9, 118.7, 111.5, 110.1, 99.1, 68.5, 59.4, 58.8, 46.2, 13.9, 9.1; HRMS (ESI-TOF) Calcd. For $\text{C}_{38}\text{H}_{26}\text{ClN}_2\text{O}_6$ ($[\text{M}+\text{H}]^+$): 641.1474, found: 641.1485.

Ethyl 3'-amino-1''-benzyl-5''-fluoro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'*H*-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carboxylate (3j): purple solid, 0.549 g, 88%, m.p. 230-232 °C; ^1H NMR (400 MHz, DMSO- d_6) δ : 8.80 (d, $J = 7.2$ Hz, 1H), 8.40 (s, 1H), 7.55 (d, $J = 7.6$ Hz, 1H), 7.47 (t, $J = 7.2$ Hz, 1H), 7.35-7.27 (m, 2H), 7.22-7.03 (m, 6H), 6.92 (t, $J = 7.6$ Hz, 2H), 6.65-6.62 (m,

1H), 6.37 (s, 2H), 6.36 (d, $J = 7.6$ Hz, 1H), 4.80 (d, $J = 16.0$ Hz, 1H), 4.39 (d, $J = 16.0$ Hz, 1H), 3.92-3.86 (m, 1H), 3.64-3.60 (m, 1H), 0.64 (t, $J = 7.2$ Hz, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO- d_6) δ : 183.7, 180.7, 169.1, 167.9, 166.9, 157.8 (d, $J = 234.9$ Hz), 151.4, 145.3, 142.5, 140.1, 139.4, 138.8, 136.1, 130.3 (d, $J = 24.9$ Hz), 130.1, 129.9, 128.6, 127.1, 126.3, 125.1, 124.1, 121.9, 118.6, 115.0 (d, $J = 22.7$ Hz), 114.9, 113.1 (d, $J = 23.7$ Hz), 111.5, 99.2, 68.5, 60.1, 59.3, 58.9, 46.2, 13.9, 9.1; HRMS (ESI-TOF) Calcd. For $\text{C}_{38}\text{H}_{25}\text{FN}_2\text{NaO}_6$ ($[\text{M}+\text{H}]^+$): 647.1589, found: 647.1585.

2. General procedure for the reactions of bindone and 4-arylidene-pyrazol-3-ones: A mixture of bindone (1.0 mmol), 4-arylidene-pyrazol-3-one (1.0 mmol), and DABCO (2.0 mmol) in DCM (20 mL) was stirred at room temperature for 24 hours. After removing the solvent by rotatory evaporating at reduced pressure, the resulting residue was purified by column chromatography on silica gel with ethyl acetate and petroleum ether (1:4) as eluent to afford the desired products **5a-5j**.

3'-methyl-1',6-diphenyl-7H-spiro[indeno[1,2-a]fluorene-5,4'-pyrazole]-5',7,12(1'H,4bH)-

trione (5a): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, yellow solid, 0.383 g, 74%, m.p. 264-266 °C; ^1H NMR (400 MHz, CDCl_3) δ : **major isomer:** 9.55 (d, $J = 8.0$ Hz, 1H), 8.01 (d, $J = 6.0$ Hz, 1H), 7.87 (t, $J = 7.6$ Hz, 2H), 7.67 (t, $J = 7.6$ Hz, 1H), 7.58 (t, $J = 8.0$ Hz, 2H), 7.54-7.53 (m, 2H), 7.38 (t, $J = 7.6$ Hz, 6H), 7.25-7.16 (m, 3H), 5.15 (s, 1H), 1.76 (s, 3H); **ratio of major/minor = 15:1**; $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ : 189.7, 187.1, 173.3, 161.4, 157.2, 145.9, 145.2, 140.6, 140.3, 140.2, 139.3, 138.9, 137.3, 137.0, 135.8, 135.7, 135.5, 134.9, 134.0, 133.4, 132.5, 132.3, 130.0, 129.8, 129.3, 129.3, 129.2, 129.1, 129.0, 128.8, 128.7, 128.1, 126.3, 126.0, 125.7, 124.9, 124.7, 124.5, 123.9, 123.7, 119.5, 119.4, 65.1, 45.7, 43.8, 15.5, 15.0; MS (m/z): HRMS (ESI-TOF) Calcd. For $\text{C}_{35}\text{H}_{22}\text{N}_2\text{NaO}_3$ ($[\text{M}+\text{Na}]^+$): 541.1523, found: 541.1525.

3'-methyl-1'-phenyl-6-(p-tolyl)-7H-spiro[indeno[1,2-a]fluorene-5,4'-pyrazole]-

5',7,12(1'H,4bH)-trione (5b): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, yellow solid, 0.414 g, 78%, m.p. 240-242 °C; ^1H NMR (400 MHz, CDCl_3) δ : **major isomer:** 9.54 (d, $J = 8.0$ Hz, 1H), 8.01-7.99 (m, 1H), 7.88-7.85 (m, 2H), 7.68-7.63 (m, 3H), 7.54-7.51 (m, 2H), 7.40 (t, $J = 8.0$ Hz, 2H), 7.23 (d, $J = 7.6$ Hz, 1H), 7.17 (s, 5H), 5.13 (s, 1H), 2.34 (s, 3H), 1.76 (s, 3H); **ratio of major/minor = 14:1**; $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ : 189.7, 188.1, 187.0, 173.5, 157.6, 155.6, 146.6, 146.3, 145.2, 144.3, 140.4, 140.2, 140.1, 139.3, 137.6, 137.1, 135.7, 135.6, 135.4, 134.9, 133.8, 132.5, 132.2, 130.6, 129.3, 129.1, 129.0, 128.9, 128.8, 126.1, 125.9, 125.6,

124.7, 124.6, 123.9, 123.7, 119.5, 119.4, 65.2, 47.1, 46.0, 43.8, 21.5, 21.4, 15.6, 15.0; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C₃₆H₂₄N₂NaO₃ ([M+Na]⁺): 555.1679, found: 555.1682.

6-(4-methoxyphenyl)-3'-methyl-1'-phenyl-7H-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5c): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, yellow solid, 0.438 g, 80%, m.p. 268-270 °C; ¹H NMR (400 MHz, CDCl₃) δ: **major isomer:** 9.55 (d, *J* = 8.0 Hz, 1H), 8.01-7.99 (m, 1H), 7.86 (t, *J* = 8.0 Hz, 2H), 7.67 (d, *J* = 8.0 Hz, 3H), 7.53-7.51 (m, 2H), 7.41 (t, *J* = 8.0 Hz, 2H), 7.27 (s, 1H), 7.24 (d, *J* = 8.0 Hz, 2H), 7.18-7.17 (m, 1H), 6.88 (d, *J* = 8.4 Hz, 2H), 5.12 (s, 1H), 3.81 (s, 3H), 1.75 (s, 3H); **ratio of major/minor = 13:1**; ¹³C{¹H} NMR (100 MHz, CDCl₃) δ: 189.7, 189.3, 187.3, 187.0, 173.6, 168.0, 162.0, 161.1, 161.0, 158.0, 146.6, 146.2, 145.1, 144.2, 141.2, 140.5, 140.4, 140.0, 139.2, 138.9, 138.8, 138.0, 137.0, 135.7, 135.5, 135.4, 134.8, 133.6, 133.3, 132.5, 132.2, 129.3, 129.2, 129.1, 129.0, 128.8, 126.0, 125.9, 125.8, 125.6, 125.5, 124.8, 124.7, 124.6, 124.0, 123.9, 123.6, 119.5, 119.3, 113.6, 113.4, 65.1, 63.8, 55.2, 55.2, 46.2, 43.9, 15.6, 15.0; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C₃₆H₂₄N₂NaO₄ ([M+Na]⁺): 571.1628, found: 571.1631.

6-(3-methoxyphenyl)-3'-methyl-1'-phenyl-7H-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5d): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, yellow solid, 0.471 g, 86%, m.p. 245-247 °C; ¹H NMR (400 MHz, CDCl₃) δ: **major isomer:** 9.54 (d, *J* = 8.0 Hz, 1H), 8.01-7.99 (m, 1H), 7.88-7.85 (m, 2H), 7.68 (d, *J* = 7.2 Hz, 1H), 7.63 (d, *J* = 8.0 Hz, 2H), 7.55-7.52 (m, 2H), 7.39 (t, *J* = 8.0 Hz, 2H), 7.32-7.28 (m, 1H), 7.25-7.22 (m, 2H), 7.17 (d, *J* = 7.6 Hz, 1H), 6.93-6.91 (m, 1H), 6.83 (s, 1H), 5.13 (s, 1H), 3.73 (s, 3H), 1.75 (s, 3H); **minor isomer:** 9.55 (d, *J* = 8.0 Hz, 1H), 7.96 (s, 1H), 7.82 (d, *J* = 7.6 Hz, 2H), 7.65 (s, 2H), 7.58 (s, 1H), 7.55 (s, 1H), 7.50 (s, 1H), 6.99-6.96 (m, 2H), 4.86 (s, 1H), 3.62 (s, 3H), 2.17 (s, 3H); **ratio of major/minor = 10:1**; ¹³C{¹H} NMR (100 MHz, CDCl₃) δ: 189.7, 186.9, 173.2, 159.1, 157.3, 145.6, 145.1, 140.3, 140.2, 139.2, 137.3, 137.0, 135.8, 135.5, 134.6, 134.0, 132.5, 129.3, 129.1, 129.0, 126.3, 125.9, 124.7, 124.5, 123.9, 119.4, 119.2, 115.8, 65.0, 55.2, 45.8, 15.6; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C₃₆H₂₄N₂NaO₄ ([M+Na]⁺): 571.1628, found: 571.1631.

6-(4-isopropylphenyl)-3'-methyl-1'-phenyl-7H-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5e): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, yellow solid, 0.397 g, 71%, m.p. 241-243 °C; ¹H NMR (400 MHz, CDCl₃) δ: **major isomer:** 9.54 (d, *J* = 8.4 Hz, 1H), 8.01-8.00 (m, 1H), 7.88-7.85 (m, 2H), 7.68-7.51 (m, 6H), 7.38 (t, *J* = 7.6 Hz, 2H),

7.25-7.21 (m, 5H), 5.13 (s, 1H), 2.96-2.86 (m, 1H), 1.75 (s, 3H), 1.22 (s, 3H), 1.20 (s, 3H); **minor isomer**: 9.56 (d, $J = 8.4$ Hz, 1H), 7.96 (d, $J = 7.6$ Hz, 1H), 7.82 (d, $J = 7.2$ Hz, 2H), 7.46 (t, $J = 7.6$ Hz, 3H), 7.32 (t, $J = 7.6$ Hz, 3H), 7.15 (d, $J = 7.6$ Hz, 2H), 4.85 (s, 1H), 2.09 (s, 3H), 1.29 (s, 3H), 1.28 (s, 3H); **ratio of major/minor = 4:1**; $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ : 189.7, 187.1, 173.4, 157.4, 151.2, 150.7, 146.5, 145.2, 140.4, 140.2, 139.2, 137.5, 136.9, 135.7, 135.6, 135.4, 134.8, 133.6, 132.5, 132.2, 130.8, 129.2, 129.1, 128.9, 128.8, 126.2, 126.1, 126.0, 125.6, 124.7, 124.6, 123.9, 123.6, 119.8, 119.4, 65.1, 45.6, 43.9, 33.9, 23.8, 23.6, 15.5, 15.0; MS (m/z): HRMS (ESI-TOF) Calcd. For $\text{C}_{38}\text{H}_{28}\text{N}_2\text{NaO}_3$ ($[\text{M}+\text{Na}]^+$): 583.1992, found: 583.1992.

6-(4-(tert-butyl)phenyl)-3'-methyl-1'-phenyl-7H-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5f): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, yellow solid, 0.436 g, 76%, m.p. 259-261 °C; ^1H NMR (400 MHz, CDCl_3) δ : **major isomer**: 9.55 (d, $J = 8.0$ Hz, 1H), 8.02-7.99 (m, 1H), 7.89-7.85 (m, 2H), 7.67 (t, $J = 7.2$ Hz, 1H), 7.55-7.53 (m, 2H), 7.48 (t, $J = 8.0$ Hz, 2H), 7.38-7.35 (m, 4H), 7.24-7.18 (m, 4H), 5.13 (s, 1H), 1.75 (s, 3H), 1.28 (s, 9H, 3CH₃); **minor isomer**: 9.56 (d, $J = 8.0$ Hz, 1H), 7.96 (d, $J = 7.6$ Hz, 1H), 7.83-7.81 (m, 1H), 7.63-7.57 (m, 5H), 7.31 (d, $J = 8.4$ Hz, 2H), 7.15 (d, $J = 8.8$ Hz, 1H), 4.85 (s, 1H), 2.09 (s, 3H), 1.34 (s, 9H, 3CH₃); **ratio of major/minor = 9:1**; $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ : 189.7, 187.2, 173.4, 157.4, 152.9, 146.5, 145.3, 140.4, 140.2, 139.2, 137.4, 136.9, 135.7, 135.4, 133.6, 132.5, 130.5, 129.2, 129.1, 128.9, 126.1, 126.0, 125.0, 124.7, 124.6, 123.9, 119.9, 65.1, 45.5, 34.7, 31.1, 15.5; MS (m/z): HRMS (ESI-TOF) Calcd. For $\text{C}_{39}\text{H}_{31}\text{N}_2\text{O}_3$ ($[\text{M}+\text{H}]^+$): 575.2329, found: 575.2331.

6-(4-chlorophenyl)-3'-methyl-1'-phenyl-7H-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5g): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, yellow solid, 0.463 g, 84%, m.p. 277-279 °C; ^1H NMR (400 MHz, CDCl_3) δ : **major isomer**: 9.53 (d, $J = 8.0$ Hz, 1H), 7.89-7.81 (m, 3H), 7.68 (t, $J = 7.6$ Hz, 1H), 7.64-7.62 (m, 2H), 7.58-7.53 (m, 2H), 7.44-7.28 (m, 6H), 7.18-7.14 (m, 2H), 5.12 (s, 1H), 1.75 (s, 3H); **minor isomer**: 9.54 (d, $J = 7.6$ Hz, 1H), 8.01-7.96 (m, 3H), 7.52-7.46 (m, 3H), 7.24 (s, 1H), 4.84 (s, 1H), 2.15 (s, 3H); **ratio of major/minor = 2:1**; $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ : 189.7, 187.0, 173.2, 161.2, 157.3, 145.0, 144.3, 144.1, 141.0, 140.6, 140.3, 139.1, 138.8, 137.8, 137.0, 136.8, 136.2, 136.0, 135.9, 135.6, 135.1, 134.5, 132.6, 132.4, 131.8, 131.6, 129.4, 129.2, 129.1, 128.9, 128.5, 126.4, 126.2, 125.8, 125.3, 125.0, 124.8, 124.5, 124.0, 123.9, 119.4, 119.3, 101.3, 65.0, 45.8, 43.7, 29.7, 15.6, 15.1; MS (m/z): HRMS (ESI-TOF) Calcd. For $\text{C}_{38}\text{H}_{21}\text{ClN}_2\text{NaO}_3$ ($[\text{M}+\text{Na}]^+$): 575.1133, found: 575.1139.

6-(3-chlorophenyl)-3'-methyl-1'-phenyl-7H-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-

5',7,12(1'*H*,4*bH*)-trione (5h): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, yellow solid, 0.419 g, 76%, m.p. 257-259 °C; ¹H NMR (400 MHz, CDCl₃) δ: **major isomer:** 9.53 (d, *J* = 8.0 Hz, 1H), 8.01 (d, *J* = 8.4 Hz, 1H), 7.87 (d, *J* = 7.2 Hz, 2H), 7.68 (t, *J* = 7.2 Hz, 1H), 7.57-7.53 (m, 3H), 7.42-7.29 (m, 5H), 7.24 (d, *J* = 7.6 Hz, 1H), 7.16 (d, *J* = 5.2 Hz, 1H), 5.13 (s, 1H), 1.75 (s, 3H); **minor isomer:** 9.54 (d, *J* = 8.0 Hz, 1H), 7.97 (d, *J* = 7.2 Hz, 1H), 7.90 (s, 1H), 7.82 (d, *J* = 7.6 Hz, 2H), 7.48 (d, *J* = 7.6 Hz, 1H), 4.85 (s, 1H), 2.18 (s, 3H); **ratio of major/minor = 4:1**; ¹³C{¹H} NMR (100 MHz, CDCl₃) δ: 189.7, 186.9, 173.1, 161.0, 157.0, 145.0, 144.2, 144.1, 143.6, 143.4, 141.0, 140.7, 140.3, 140.3, 139.2, 139.1, 138.8, 136.8, 136.7, 136.0, 135.9, 135.6, 135.2, 135.1, 135.0, 134.9, 134.8, 134.7, 134.2, 132.6, 132.4, 130.0, 129.8, 129.7, 129.6, 129.4, 129.3, 129.2, 129.0, 128.9, 126.6, 126.1, 125.9, 125.4, 125.0, 124.8, 124.5, 124.0, 123.8, 123.7, 119.7, 119.5, 64.9, 63.7, 45.6, 43.6, 43.1, 15.5, 15.1; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C₃₅H₂₂ClN₂O₃ ([M+H]⁺): 553.1313, found: 553.1320.

6-(4-bromophenyl)-3'-methyl-1'-phenyl-7H-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-

5',7,12(1'*H*,4*bH*)-trione (5i): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, yellow solid, 0.429 g, 72%, m.p. 275-277 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: **major isomer:** 9.43 (d, *J* = 8.0 Hz, 1H), 8.02-7.95 (m, 2H), 7.82-7.78 (m, 2H), 7.69 (t, *J* = 8.0 Hz, 1H), 7.63-7.56 (m, 5H), 7.47 (t, *J* = 7.6 Hz, 2H), 7.31-7.20 (m, 3H), 7.03 (d, *J* = 7.6 Hz, 1H), 5.17 (s, 1H), 1.72 (s, 3H); **ratio of major/minor = 14:1**; ¹³C{¹H} NMR (100 MHz, DMSO-*d*₆) δ: 189.6, 186.9, 173.4, 158.1, 145.4, 142.8, 140.3, 139.3, 136.9, 136.4, 134.8, 133.3, 133.0, 131.3, 129.7, 128.7, 126.9, 126.6, 124.9, 124.2, 123.3, 119.6, 64.8, 45.5, 15.6; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C₃₅H₂₂BrN₂O₃ ([M+H]⁺): 597.0808, found: 597.0808.

3'-methyl-6-(4-nitrophenyl)-1'-phenyl-7H-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-

5',7,12(1'*H*,4*bH*)-trione (5j): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, yellow solid, 0.461 g, 82%, m.p. 273-275 °C; ¹H NMR (400 MHz, CDCl₃) δ: **major isomer:** 9.53 (d, *J* = 8.0 Hz, 1H), 8.23 (d, *J* = 8.8 Hz, 2H), 8.03-7.99 (m, 1H), 7.92-7.82 (m, 2H), 7.71-7.66 (m, 1H), 7.61-7.50 (m, 5H), 7.40 (t, *J* = 8.0 Hz, 2H), 7.33 (t, *J* = 7.2 Hz, 1H), 7.23-7.13 (m, 2H), 5.16 (s, 1H), 1.78 (s, 3H); **minor isomer:** 7.97 (s, 1H), 7.50 (s, 1H), 4.88 (s, 1H), 2.19 (s, 3H); **ratio of major/minor = 3:1**; ¹³C{¹H} NMR (100 MHz, CDCl₃) δ: 189.6, 187.0, 172.8, 167.6, 160.5, 156.8, 148.3, 144.8, 144.0, 142.0, 141.4, 140.9, 140.8, 140.5, 140.2, 139.9, 139.0, 138.6, 137.0, 136.7, 136.4, 136.3,

136.2, 135.9, 135.5, 135.4, 132.9, 132.6, 131.0, 129.6, 129.5, 129.4, 129.3, 129.2, 129.1, 128.7, 128.1, 127.0, 126.3, 126.1, 125.9, 125.2, 124.9, 124.5, 124.2, 124.0, 123.8, 123.5, 119.3, 119.1, 64.8, 63.8, 45.6, 43.6, 15.6, 15.3; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C₃₅H₂₁N₃NaO₅ ([M+Na]⁺): 586.1373, found: 586.1380.

3. General Procedure for the reaction of bindone and 2-arylidene-1,3-indanediones: A mixture of bindone (1.0 mmol), bindone (1.0 mmol), 2-arylidene-1,3-indanedione (1.0 mmol) and Et₃N (2.0 mmol) in toluene (3.0 mL) in sealed tube was heated 120 °C in oil bath at for twelve hours. After removing the solvent, the residue was recrystallized in ethyl acetate to give the pure product **7a-7h** for analysis.

15-hydroxy-15-(*m*-tolyl)diindeno[1,2-*a*:1',2'-*c*]anthracene-5,10,16(15*H*)-trione [7a]: light yellow solid, 0.413g, 82%, m.p. >300 °C; ¹H NMR (400 MHz, CDCl₃) δ: 9.16 (d, *J* = 7.6 Hz, 1H), 8.30 (d, *J* = 7.6 Hz, 1H), 8.10-8.07 (m, 2H), 7.88 (s, 1H), 7.81 (d, *J* = 7.2 Hz, 1H), 7.66-7.60 (m, 4H), 7.51-7.48 (m, 1H), 7.44-7.40 (m, 2H), 7.28 (s, 1H), 7.14 (s, 1H), 7.10-7.06 (m, 1H), 6.90 (d, *J* = 7.2 Hz, 1H), 2.21 (s, 3H); ¹³C NMR (100 MHz, CDCl₃) δ: 194.4, 191.9, 186.6, 154.4, 150.9, 147.8, 146.7, 145.9, 142.2, 141.3, 138.2, 136.2, 135.6, 135.2, 134.7, 134.3, 131.7, 131.4, 130.7, 130.4, 129.8, 128.5, 128.4, 128.2, 127.8, 127.5, 126.4, 126.0, 125.4, 124.8, 124.2, 121.9, 7.59, 21.6; MS (*m/z*): HRMS (ESI) Calcd. For C₃₅H₂₀NaO₄ ([M+Na]⁺): 527.1254, found: 527.1259.

15-hydroxy-15-(4-methoxyphenyl)diindeno[1,2-*a*:1',2'-*c*]anthracene-5,10,16(15*H*)-trione (7b): light yellow solid, 0.395g, 76%, m.p. 286-288 °C; ¹H NMR (400 MHz, CDCl₃) δ: 9.16 (d, *J* = 8.0 Hz, 1H), 9.29 (d, *J* = 8.0 Hz, 1H), 8.11-8.07 (m, 2H), 7.94 (s, 1H), 7.81 (d, *J* = 7.6 Hz, 1H), 7.65-7.61 (m, 4H), 7.51-7.48 (m, 1H), 7.44-7.41 (m, 2H), 7.10-7.04 (m, 2H), 6.91 (d, *J* = 7.6 Hz, 1H), 6.62 (d, *J* = 10.4 Hz, 1H), 3.71 (s, 3H); ¹³C NMR (100 MHz, CDCl₃) δ: 194.5, 191.9, 186.4, 159.5, 154.1, 151.1, 151.0, 150.7, 147.9, 147.6, 146.4, 145.8, 142.2, 141.3, 136.3, 135.9, 135.6, 135.3, 134.7, 134.3, 131.8, 131.5, 131.5, 130.7, 130.5, 129.8, 129.5, 128.6, 127.9, 127.6, 126.4, 126.1, 124.9, 124.2, 117.3, 112.2, 111.3, 74.5, 55.2; MS (*m/z*): HRMS (ESI) Calcd. For C₃₅H₂₀NaO₅ ([M+Na]⁺): 543.1203, found: 543.1203.

15-hydroxy-15-(3-methoxyphenyl)diindeno[1,2-*a*:1',2'-*c*]anthracene-5,10,16(15*H*)-trione (7c): light yellow solid, 0.338g, 65%, m.p. 290-292 °C; ¹H NMR (400 MHz, CDCl₃) δ: 9.17 (d, *J* =

8.0 Hz ,1H), 8.30 (d, J = 7.6 Hz, 1H), 8.10-8.07 (m, 2H), 7.94 (s, 1H), 7.82 (d, J = 7.2 Hz, 2H), 7.66-7.62 (m, 4H), 7.52-7.48 (m, 1H), 7.45-7.41 (m, 2H), 7.10-7.03 (m, 2H), 6.91 (d, J = 8.0 Hz, 1H), 6.62 (d, J = 8.4 Hz 1H), 3.71 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ : 194.4, 191.8, 186.4, 159.5, 154.1, 151.0, 147.8, 147.6, 146.4, 142.2, 141.2, 136.2, 135.5, 135.2, 134.6, 134.3, 131.7, 131.4, 130.7, 130.4, 129.8, 129.5, 128.5, 127.9, 127.6, 126.4, 126.1, 124.8, 124.2, 117.3, 112.2, 111.2, 74.5, 55.2; MS (m/z): HRMS (ESI) Calcd. For $\text{C}_{35}\text{H}_{20}\text{NaO}_5$ ($[\text{M}+\text{Na}]^+$): 543.1203, found: 543.1197.

15-hydroxy-15-(2-methoxyphenyl)diindeno[1,2-a:1',2'-c]anthracene-5,10,16(15H)-trione (7d):

light yellow solid, 0.411g, 79%, m.p. 297-299°C; ^1H NMR (400 MHz, CDCl_3) δ : 9.17 (d, J = 8.0 Hz ,1H), 8.30 (d, J = 8.0 Hz, 1H), 8.10-8.05 (m, 2H), 7.92 (s, 1H), 7.83 (d, J = 7.2 Hz, 1H), 7.68-7.62 (m, 4H), 7.53-7.49 (m, 1H), 7.46-7.42 (m, 2H), 7.41-7.37 (m, 2H), 6.89-6.84 (m, 2H); ^{13}C NMR (100 MHz, CDCl_3) δ : 194.4, 191.9, 186.4, 159.5, 154.1, 151.0, 147.8, 147.6, 146.4, 142.2, 141.3, 136.3, 135.6, 135.2, 134.7, 134.3, 131.8, 131.5, 131.4, 130.7, 130.5, 129.8, 129.5, 128.6, 127.9, 127.6, 126.4, 126.1, 124.9, 124.2, 117.3, 112.2, 111.2, 74.5, 55.2; MS (m/z): HRMS (ESI) Calcd. For $\text{C}_{35}\text{H}_{20}\text{NaO}_5$ ($[\text{M}+\text{Na}]^+$): 543.1203, found: 543.1211.

15-(4-fluorophenyl)-15-hydroxydiindeno[1,2-a:1',2'-c]anthracene-5,10,16(15H)-trione (7e):

light yellow solid, 0.340g, 67%, m.p. 285-287°C; ^1H NMR (400 MHz, CDCl_3) δ : 9.17 (d, J = 8.0 Hz ,1H), 8.3 (d, J = 8.0 Hz , 1H), 8.10-8.05 (m, 2H), 7.92 (s, 1H), 7.83 (d, J = 7.2 Hz, 1H), 7.68-7.62 (m, 4H), 7.53-7.49 (m, 1H), 7.46-7.42 (m, 2H), 7.41-7.37 (m, 2H), 6.89-6.84 (m, 2H); ^{13}C NMR (100 MHz, CDCl_3) δ : 194.5, 191.9, 186.4, 159.5, 154.1, 151.1, 147.7 (d, J = 26.3 Hz), 146.4, 142.2, 141.3, 136.3, 135.6, 135.2, 134.7, 134.3, 131.6, 131.5, 130.7, 129.8, 129.5, 128.6, 127.9, 127.6, 126.4, 126.1, 124.9, 124.2, 117.3, 112.2, 111.3, 74.6, 55.1, 29.7; MS (m/z): HRMS (ESI) Calcd. For $\text{C}_{34}\text{H}_{17}\text{FNaO}_4$ ($[\text{M}+\text{Na}]^+$): 531.1003, found: 532.1020.

15-(3-fluorophenyl)-15-hydroxydiindeno[1,2-a:1',2'-c]anthracene-5,10,16(15H)-trione (7f):

light yellow solid, 0.360g, 71%, m.p. 286-287°C; ^1H NMR (400 MHz, CDCl_3) δ : 9.17 (d, J = 8.0 Hz, 1H), 8.30 (d, J = 8.0 Hz, 1H), 8.11-8.06 (m, 2H), 7.97 (s, 1H), 7.83 (d, J = 7.6 Hz ,1H), 7.69-7.63 (m, 4H), 7.53-7.50 (m, 1H), 7.47-7.42 (m, 2H), 7.23 (s, 1H), 7.15-7.06 (m, 2H), 6.82-6.77(m, 1H); ^{13}C NMR (100 MHz, CDCl_3) δ : 194.5, 191.7, 186.2, 162.6 (d, J = 245.7 Hz), 153.4, 151.3, 148.7, 148.6, 147.8, 145.9, 142.1, 141.2, 136.4, 135.5, 135.3, 134.6, 134.5, 131.7, 131.6, 130.7, 130.6, 130.1 (d, J = 8.2 Hz), 129.6, 128.6, 128.5, 128.2, 127.6, 126.4, 126.2, 124.9, 124.2, 120.5,

114.4 (d, $J = 21.0$ Hz), 112.3 (d, $J = 23.1$ Hz), 74.2, 73.0; MS (m/z): HRMS (ESI) Calcd. For $C_{34}H_{17}FNaO_4$ ($[M+Na]^+$): 531.1003, found: 531.1016.

15-hydroxy-15-(4-isopropylphenyl)diindeno[1,2-a:1',2'-c]anthracene-5,10,16(15H)-trione

(7g): light yellow solid, 0.276g, 52%, m.p. 294-296°C; 1H NMR (400 MHz, $CDCl_3$) δ : 9.18 (d, $J = 7.2$ Hz, 1H), 8.32 (d, $J = 7.6$ Hz, 1H), 8.11-8.07 (m, 2H), 7.94 (s, 1H), 7.83 (d, $J = 7.2$ Hz, 1H), 7.66-7.62 (m, 4H), 7.53-7.49 (m, 1H), 7.45-7.40 (m, 2H), 7.31 (d, $J = 7.6$ Hz, 2H), 7.02 (d, $J = 8.0$ Hz, 2H), 2.78-2.71 (m, 1H, CH), 1.10 (d, $J = 6.8$ Hz, 6H); ^{13}C NMR (100 MHz, $CDCl_3$) δ : 194.4, 191.8, 186.6, 154.6, 150.9, 147.8, 147.7, 146.9, 143.2, 142.2, 141.2, 136.1, 135.6, 135.2, 134.7, 134.3, 131.7, 131.4, 130.7, 130.3, 129.8, 128.5, 127.7, 127.6, 126.6, 126.4, 126.0, 124.8, 124.7, 124.1, 74.6, 33.4, 23.7, 23.6; MS (m/z): HRMS (ESI) Calcd. For $C_{37}H_{24}NaO_4$ ($[M+Na]^+$): 555.1567, found: 555.1573.

15-(4-(tert-butyl)phenyl)-15-hydroxydiindeno[1,2-a:1',2'-c]anthracene-5,10,16(15H)-trione

(7h): light yellow solid, 0.317g, 58 %, m.p. >300 °C; 1H NMR (400 MHz, $CDCl_3$) δ : 9.12 (d, $J = 8.8$ Hz, 1H), 8.28 (d, $J = 7.6$ Hz, 1H), 8.10-8.05 (m, 2H), 7.92 (s, 1H), 7.79 (d, $J = 6.8$ Hz, 1H), 7.65-7.55 (m, 4H), 7.50-7.39 (m, 3H), 7.31-7.26 (m, 2H), 7.17 (d, $J = 8.4$ Hz, 2H), 1.17 (s, 9H, 3CH₃); ^{13}C NMR (100 MHz, $CDCl_3$) δ : 194.3, 191.7, 186.6, 154.6, 150.9, 149.9, 147.7, 146.9, 142.7, 142.1, 141.2, 136.1, 135.5, 135.1, 134.6, 134.3, 131.6, 131.4, 131.3, 130.7, 130.3, 129.8, 128.5, 127.7, 127.5, 126.4, 126.0, 125.5, 124.7, 124.4, 124.1, 74.6, 34.3, 31.1; MS (m/z): HRMS (ESI) Calcd. For $C_{38}H_{26}NaO_4$ ($[M+Na]^+$): 569.1723, found: 569.1713.

4. General Procedure for the reaction of 1,3-indanedione and isatins: A mixture of 1,3-indanedione (1.0 mmol), isatin (1.0 mmol) and triethylamine (2.0 mmol) in toluene (10.0 mL) was heated in oil bath at 90 °C for sixteen hours. After removing the solvent, the residue was subjected to column chromatography with a mixture of ethyl acetate and light petroleum (V/V = 1:4) as eluent to give the pure product **8a-8h** for analysis.

1'-benzyl-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-

2',5,10,12,17-pentaone (8a): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, white solid, 0.454 g, 73%, m.p. 291-293 °C; 1H NMR (400 MHz, $CDCl_3$) δ : 8.15 (d, $J = 7.6$ Hz, 1H), 8.10 (d, $J = 3.6$ Hz, 1H), 8.00 (d, $J = 7.6$ Hz, 1H), 7.94 (d, $J = 7.6$ Hz, 1H), 7.87-7.77 (m, 2H), 7.65 (d, $J = 7.6$ Hz, 2H), 7.54-7.42 (m, 5H), 7.35-7.30 (m, 2H), 7.23 (s, 1H), 7.00-6.93 (m, 2H), 6.72 (d, $J = 8.0$ Hz, 1H), 6.28 (t, $J = 7.6$ Hz, 1H), 5.27 (d, $J = 16.0$ Hz, 1H), 5.19 (s, 1H), 5.04 (d, $J = 16.0$ Hz, 1H), 4.85

(d, $J = 7.6$ Hz, 1H), 4.66 (s, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ : 197.5, 193.0, 192.6, 177.0, 152.5, 149.5, 145.0, 141.7, 135.8, 135.4, 135.3, 135.0, 134.9, 133.7, 132.3, 132.0, 130.6, 130.2, 129.4, 129.0, 128.7, 128.5, 127.4, 127.2, 126.8, 126.6, 125.2, 123.7, 122.4, 121.3, 109.6, 59.9, 57.1, 52.0, 49.2, 45.2; HRMS (ESI-TOF) Calcd. For $\text{C}_{42}\text{H}_5\text{NNaO}_5$ ($[\text{M}+\text{Na}]^+$): 646.1625, found: 646.1629.

1'-benzyl-5'-methyl-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8b): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, white solid, 0.515 g, 81%, m.p. 293-295 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.15 (d, $J = 7.6$ Hz, 1H), 8.11 (d, $J = 3.6$ Hz, 1H), 8.00 (d, $J = 7.6$ Hz, 1H), 7.96 (d, $J = 7.6$ Hz, 1H), 7.84-7.79 (m, 2H), 7.65 (d, $J = 7.6$ Hz, 2H), 7.56-7.48 (m, 2H), 7.43 (t, $J = 7.6$ Hz, 2H), 7.33-7.26 (m, 3H), 7.24 (d, $J = 7.6$ Hz, 1H), 6.96 (d, $J = 8.0$ Hz, 1H), 6.79 (d, $J = 8.0$ Hz, 1H), 6.60 (d, $J = 8.0$ Hz, 1H), 5.25 (d, $J = 16.0$ Hz, 1H), 5.20 (s, 1H), 5.02 (d, $J = 16.0$ Hz, 1H), 4.65 (s, 1H), 4.60 (s, 1H), 1.65 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ : 197.6, 193.0, 192.6, 192.5, 176.8, 152.3, 149.6, 142.5, 141.7, 135.9, 135.3, 135.3, 135.2, 135.0, 133.7, 132.3, 130.6, 130.3, 130.1, 129.3, 129.0, 128.8, 128.6, 127.5, 127.4, 127.3, 127.1, 126.8, 126.5, 126.4, 123.6, 122.4, 109.3, 59.9, 57.0, 52.0, 49.2, 45.2, 20.6; HRMS (ESI-TOF) Calcd. For $\text{C}_{43}\text{H}_{27}\text{NNaO}_5$ ($[\text{M}+\text{Na}]^+$): 660.1781, found: 660.1785.

1'-benzyl-5'-chloro-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8c): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, white solid, 0.519 g, 79%, m.p. 298-300 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.16-8.11 (m, 2H), 8.01-7.95 (m, 2H), 7.88-7.79 (m, 2H), 7.64 (d, $J = 7.2$ Hz, 2H), 7.59 (t, $J = 7.2$ Hz, 1H), 7.51 (t, $J = 7.2$ Hz, 1H), 7.44 (t, $J = 7.6$ Hz, 2H), 7.39-7.34 (m, 2H), 7.30 (d, $J = 7.6$ Hz, 1H), 7.23 (s, 1H), 6.97-6.93 (m, 2H), 6.62 (d, $J = 8.4$ Hz, 1H), 5.26 (d, $J = 16.0$ Hz, 1H), 5.20 (s, 1H), 5.02 (d, $J = 16.0$ Hz, 1H), 4.66 (s, 1H), 4.64 (d, $J = 8.4$ Hz, 1H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ : 197.1, 192.6, 192.3, 176.6, 153.1, 149.0, 143.5, 141.5, 135.7, 135.4, 135.4, 135.2, 135.2, 134.8, 133.8, 132.4, 131.3, 130.8, 130.5, 129.6, 129.1, 128.9, 128.8, 128.4, 127.5, 127.4, 127.2, 126.8, 126.7, 126.5, 126.0, 123.8, 122.5, 110.4, 59.8, 57.1, 52.1, 49.1, 45.3; HRMS (ESI-TOF) Calcd. For $\text{C}_{42}\text{H}_4\text{ClNNaO}_5$ ($[\text{M}+\text{Na}]^+$): 680.1235, found: 680.1241.

1'-benzyl-5'-fluoro-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8d): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, white solid, 0.493 g, 77%, m.p. 297-299 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.16 (d, $J = 7.6$ Hz, 1H), 8.12 (d, $J = 7.6$ Hz, 1H), 8.00 (d, $J = 7.2$ Hz, 1H), 7.96 (d, $J = 7.6$ Hz, 1H), 7.88-7.80 (m, 2H),

7.64 (d, $J = 7.2$ Hz, 2H), 7.57 (t, $J = 7.6$ Hz, 1H), 7.51 (t, $J = 7.6$ Hz, 1H), 7.45 (t, $J = 7.6$ Hz, 2H), 7.36 (t, $J = 7.6$ Hz, 2H), 7.29 (d, $J = 6.8$ Hz, 1H), 7.23 (d, $J = 6.8$ Hz, 1H), 6.96 (d, $J = 8.0$ Hz, 1H), 6.69 (t, $J = 7.6$ Hz, 1H), 6.63-6.60 (m, 1H), 5.27 (d, $J = 16.0$ Hz, 1H), 5.20 (s, 1H), 5.01 (d, $J = 16.0$ Hz, 1H), 4.66 (s, 1H), 4.49 (d, $J = 9.2$ Hz, 1H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ : 197.2, 192.7, 192.3, 176.7, 159.0, 153.0, 149.1, 141.5, 141.0, 135.6, 135.5, 135.4, 135.2, 134.8, 133.8, 132.4, 131.4, 130.6, 130.5, 129.5, 129.1, 128.9, 128.7, 127.5, 127.4, 127.2, 126.8, 126.5, 123.8, 122.5, 114.6, 113.6, 59.8, 57.1, 52.1, 49.4, 45.3; HRMS (ESI-TOF) Calcd. For $\text{C}_{42}\text{H}_{44}\text{FNNaO}_5$ ($[\text{M}+\text{Na}]^+$): 664.1531, found: 664.1540.

1'-butyl-5'-methyl-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8e): ethyl acetate and petroleum ether ($v/v=1:4$) as the eluent, white solid, 0.518 g, 86%, m.p. 299-301 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.09 (t, $J = 7.2$ Hz, 2H), 7.99-7.94 (m, 2H), 7.84-7.76 (m, 2H), 7.55 (t, $J = 7.6$ Hz, 1H), 7.49 (t, $J = 7.6$ Hz, 1H), 7.32 (t, $J = 8.0$ Hz, 1H), 7.23-7.19 (m, 2H), 6.93 (t, $J = 7.6$ Hz, 2H), 6.84 (d, $J = 8.0$ Hz, 1H), 5.17 (s, 1H), 4.58 (d, $J = 7.2$ Hz, 2H), 3.95-3.88 (m, 1H), 3.85-3.78 (m, 1H), 1.94-1.85 (m, 2H), 1.69 (s, 3H), 1.60-1.55 (m, 2H), 1.05 (t, $J = 7.2$ Hz, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ : 197.7, 192.9, 192.6, 192.5, 176.1, 152.2, 149.7, 142.8, 141.8, 135.4, 135.3, 135.2, 135.0, 134.9, 133.6, 132.3, 132.2, 130.6, 130.0, 129.8, 129.2, 129.0, 128.8, 127.5, 127.3, 126.7, 126.6, 126.5, 123.5, 122.3, 108.1, 59.9, 56.9, 52.0, 49.0, 40.8, 29.0, 20.6, 20.4, 13.9; HRMS (ESI-TOF) Calcd. For $\text{C}_{40}\text{H}_{30}\text{NNaO}_5$ ($[\text{M}+\text{H}]^+$): 604.2118, found: 604.2118.

1'-butyl-5'-chloro-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8f): ethyl acetate and petroleum ether ($v/v=1:4$) as the eluent, white solid, 0.417 g, 67%, m.p. 286-288 °C; ^1H NMR (400 MHz, CDCl_3) δ : 8.11 (d, $J = 7.6$ Hz, 2H), 7.97 (t, $J = 8.0$ Hz, 2H), 7.86-7.77 (m, 2H), 7.59 (t, $J = 7.6$ Hz, 1H), 7.50 (t, $J = 7.6$ Hz, 1H), 7.39 (t, $J = 7.6$ Hz, 1H), 7.25-7.21 (m, 1H), 7.11 (d, $J = 7.6$ Hz, 1H), 7.02 (t, $J = 7.2$ Hz, 1H), 6.92 (d, $J = 7.6$ Hz, 1H), 6.87 (d, $J = 8.4$ Hz, 1H), 5.17 (s, 1H), 4.65 (s, 1H), 4.55 (s, 1H), 3.95-3.78 (m, 2H), 1.92-1.85 (m, 2H), 1.58-1.53 (m, 2H), 1.05 (t, $J = 7.6$ Hz, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ : 197.2, 192.8, 192.6, 192.3, 175.9, 152.9, 149.0, 143.8, 141.6, 135.7, 135.4, 135.2, 135.1, 134.8, 133.8, 132.4, 131.4, 130.7, 130.5, 129.5, 129.1, 128.4, 127.1, 126.7, 126.7, 126.1, 123.7, 122.4, 109.2, 59.7, 57.0, 52.1, 49.0, 40.9, 28.9, 20.4, 13.8; HRMS (ESI-TOF) Calcd. For $\text{C}_{39}\text{H}_{24}\text{ClNNaO}_5$ ($[\text{M}+\text{Na}]^+$): 646.1392, found: 646.1395.

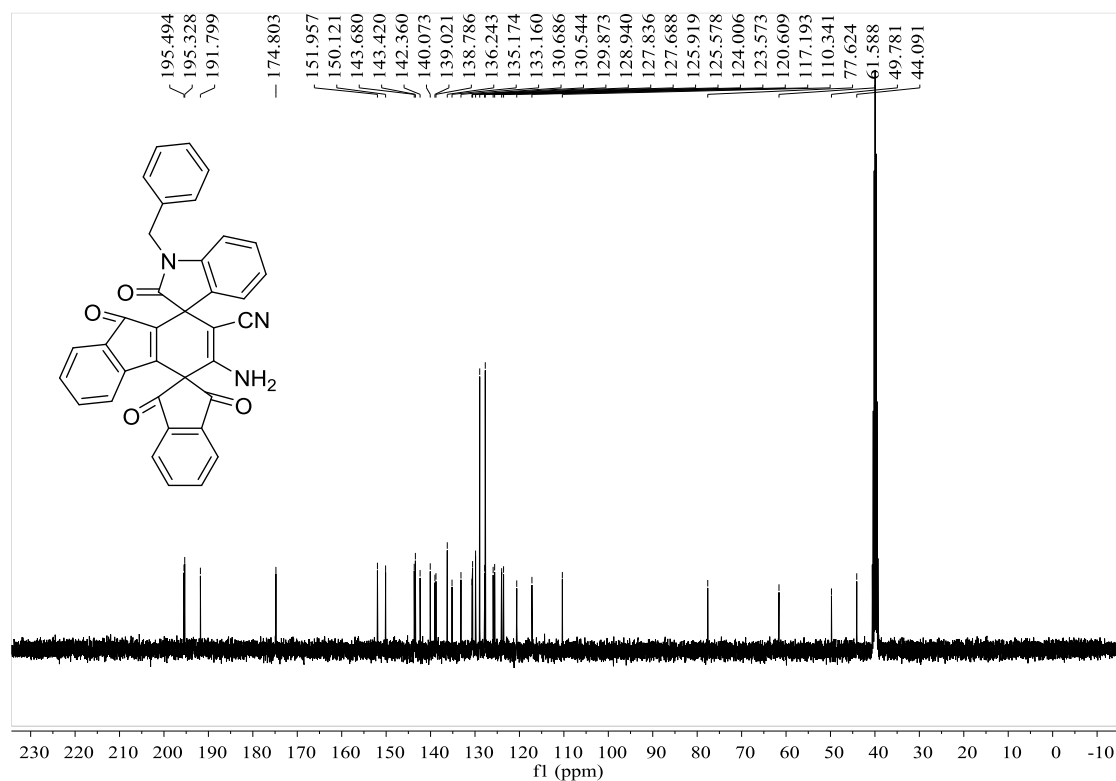
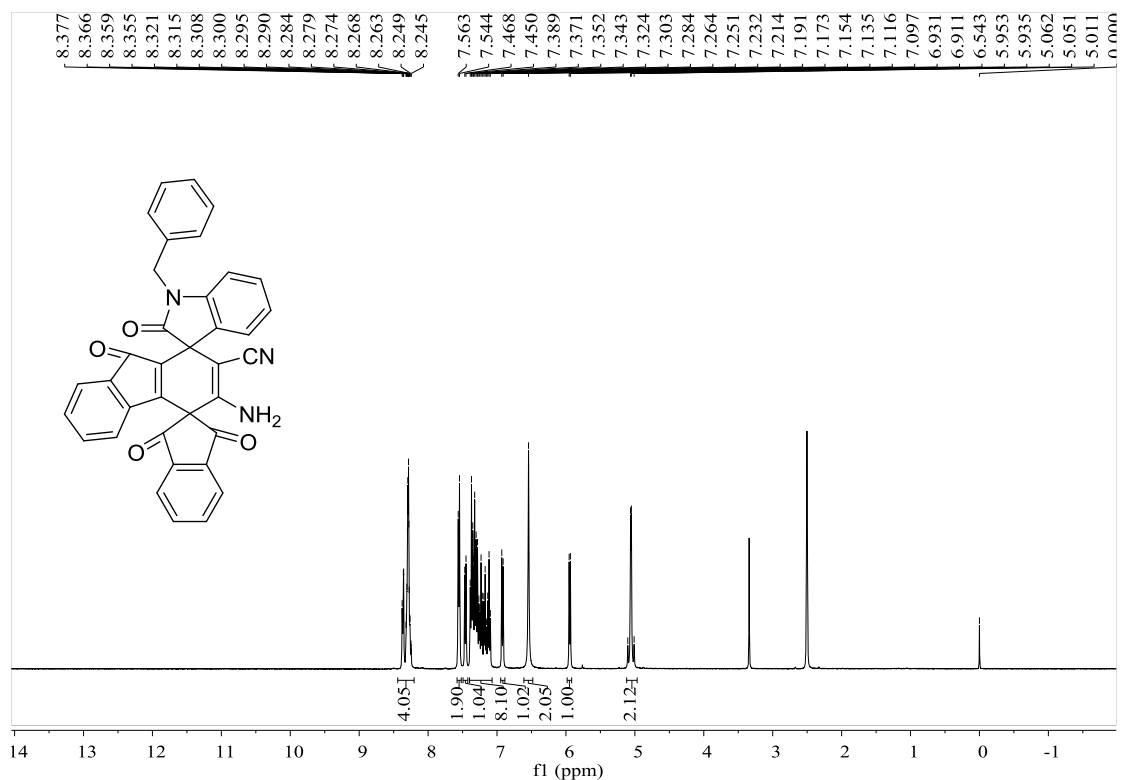
1'-butyl-5'-fluoro-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-

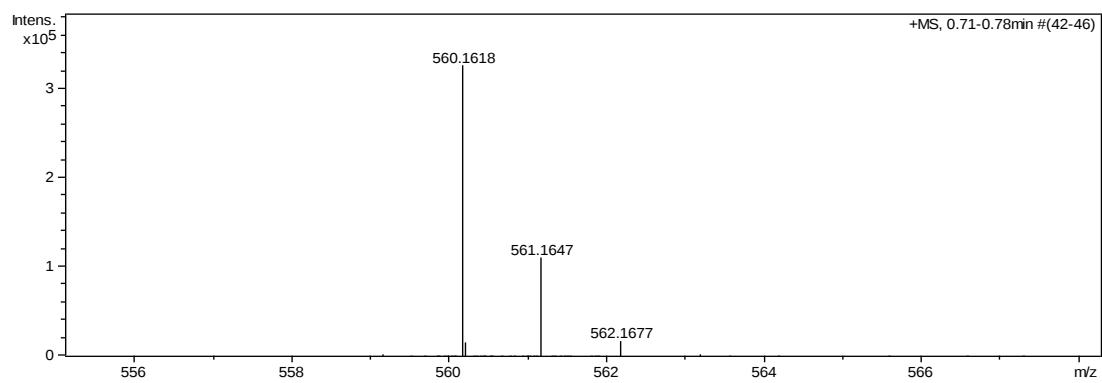
indoline]-2',5,10,12,17-pentaone (8g): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, white solid, 0.370 g, 61%, m.p. 241-243 °C; ¹H NMR (400 MHz, CDCl₃) δ: 8.10 (d, *J* = 7.6 Hz, 2H), 7.97 (t, *J* = 7.6 Hz, 2H), 7.85-7.78 (m, 2H), 7.57 (t, *J* = 7.6 Hz, 1H), 7.49 (t, *J* = 7.2 Hz, 1H), 7.37 (t, *J* = 7.6 Hz, 1H), 7.24-7.21 (m, 2H), 6.94 (d, *J* = 8.0 Hz, 1H), 6.88-6.81 (m, 2H), 5.17 (s, 1H), 4.58 (s, 1H), 4.49 (d, *J* = 7.2 Hz, 1H), 3.95-3.88 (m, 1H), 3.85-3.78 (m, 1H), 1.93-1.84 (m, 2H), 1.60-1.56 (m, 1H), 1.55-1.51 (m, 1H), 1.05 (t, *J* = 7.6 Hz, 3H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ: 197.3, 192.8, 192.6, 192.4, 175.9, 157.7 (d, *J* = 238.4 Hz), 152.8, 149.2, 141.6, 141.2, 135.6, 135.4, 135.2, 135.1, 134.8, 133.7, 132.3, 131.5, 130.5, 130.5, 129.5, 129.1, 127.2, 126.7, 123.7, 122.4, 114.7 (d, *J* = 22.9 Hz), 113.7 (d, *J* = 26.5 Hz), 108.6 (d, *J* = 8.1 Hz), 59.8, 57.0, 52.1, 49.3, 40.9, 28.9, 20.4, 13.8; HRMS (ESI-TOF) Calcd. For C₃₉H₂₆FNNaO₅ ([M+Na]⁺): 630.1687, found: 630.1693.

5'-chloro-1'-methyl-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-

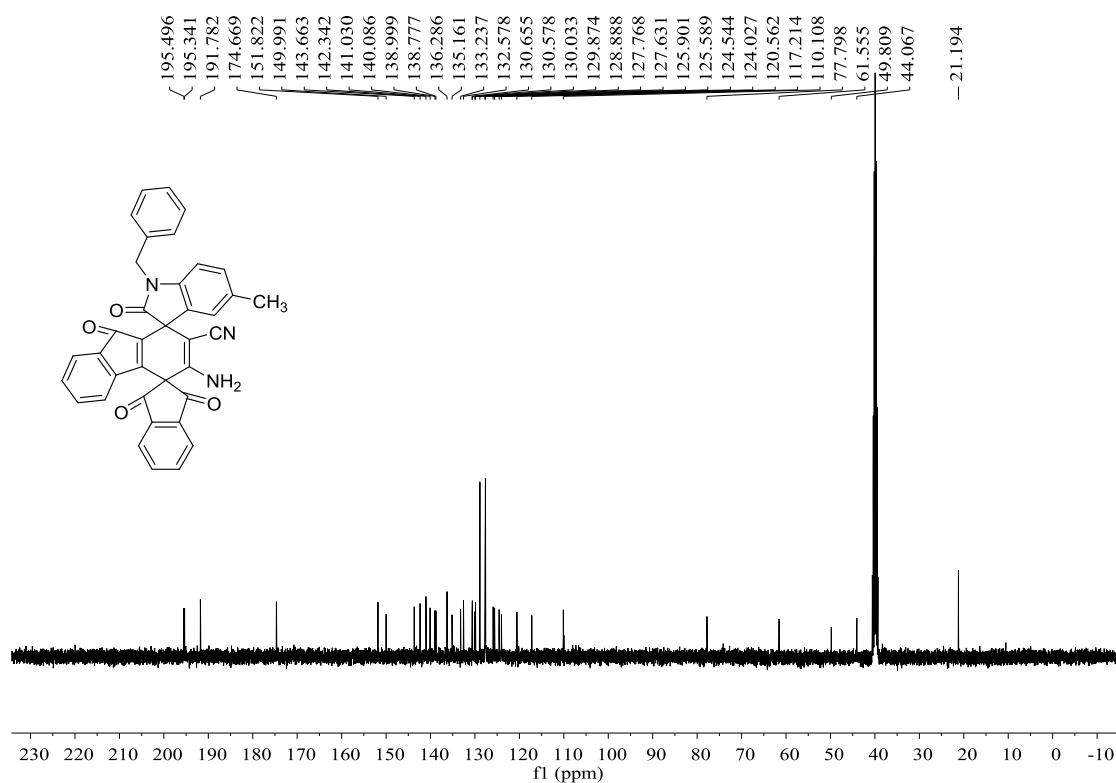
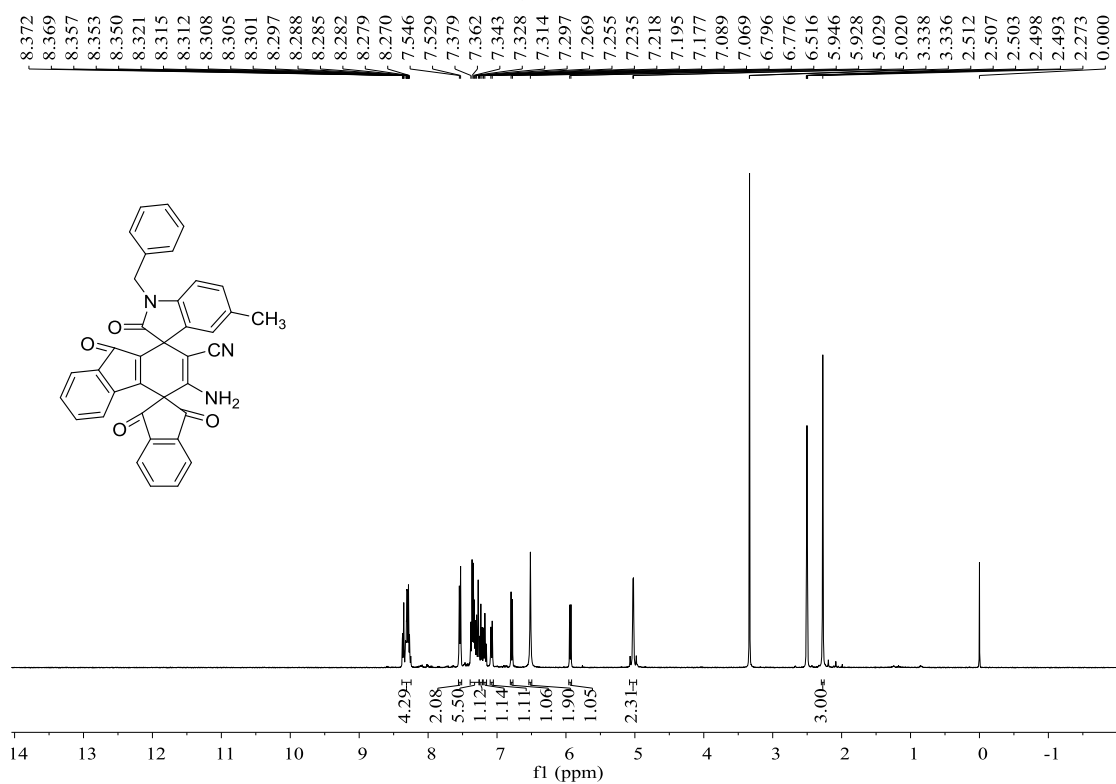
indoline]-2',5,10,12,17-pentaone (8h): ethyl acetate and petroleum ether (v/v=1:4) as the eluent, white solid, 0.447 g, 77%, m.p. >300 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 8.10-8.05 (m, 2H), 7.98-7.92 (m, 4H), 7.69 (t, *J* = 7.6 Hz, 1H), 7.63 (t, *J* = 7.6 Hz, 1H), 7.57 (t, *J* = 7.6 Hz, 1H), 7.35 (t, *J* = 7.6 Hz, 1H), 7.25-7.21 (m, 2H), 7.11 (d, *J* = 7.6 Hz, 1H), 6.83 (d, *J* = 8.0 Hz, 1H), 5.25 (s, 1H), 5.11 (s, 1H), 4.53 (s, 1H), 3.30 (s, 3H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ: 198.2, 193.7, 193.1, 192.4, 176.0, 154.1, 150.0, 144.9, 141.6, 136.1, 134.8, 134.6, 132.6, 131.5, 130.3, 130.1, 129.5, 128.8, 128.7, 127.3, 126.9, 125.7, 125.2, 124.1, 59.6, 56.3, 53.2, 48.9, 27.3; HRMS (ESI-TOF) Calcd. For C₃₆H₂₁ClNO₅ ([M+H]⁺): 582.1103, found: 582.1109.

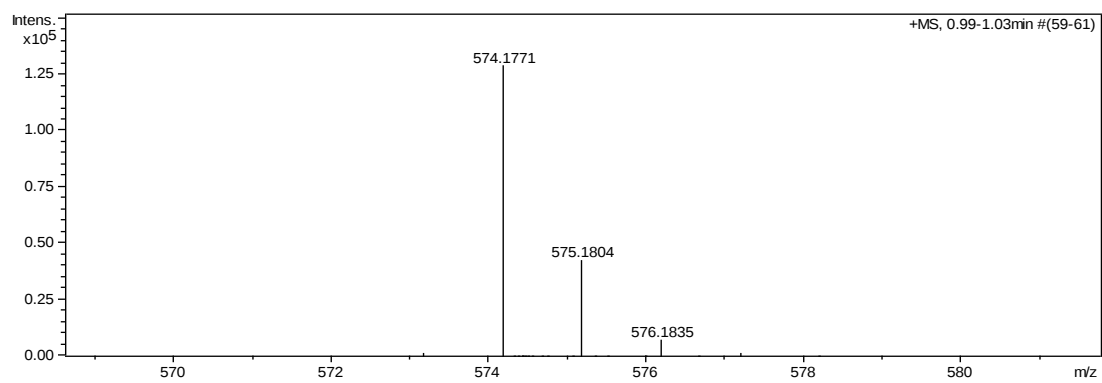
3'-amino-1''-benzyl-1,2'',3,9'-tetraoxo-1,3-dihydro-9'H-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carbonitrile (3a):



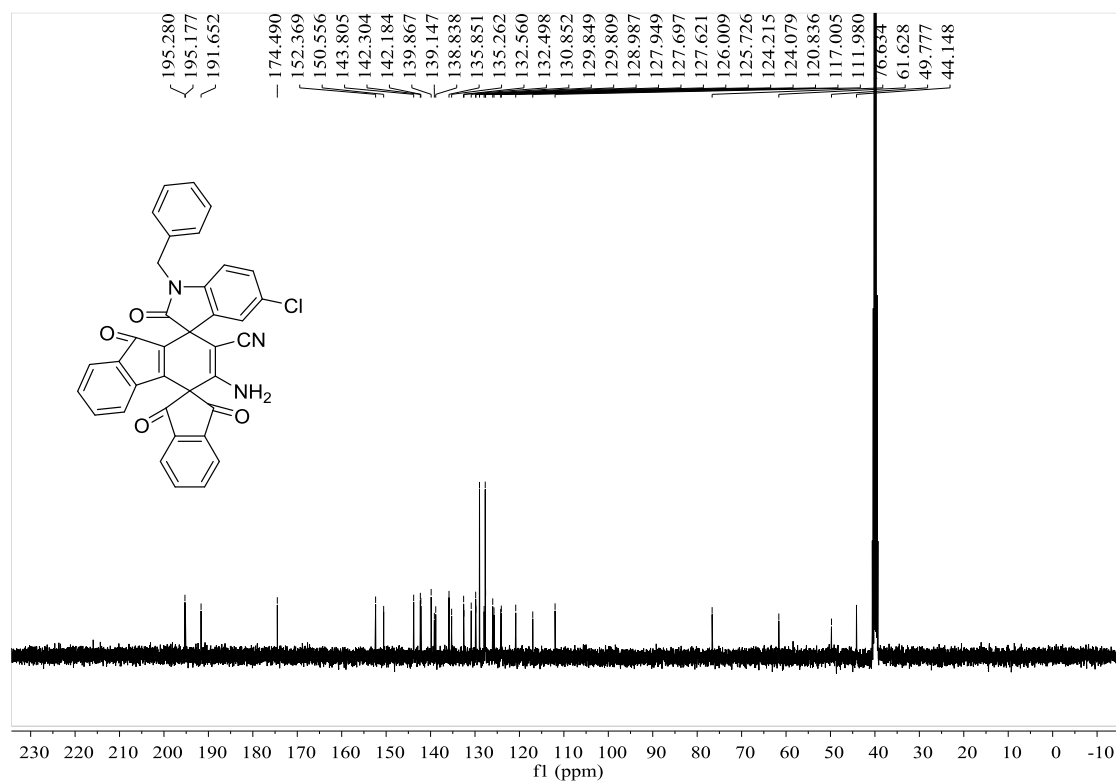
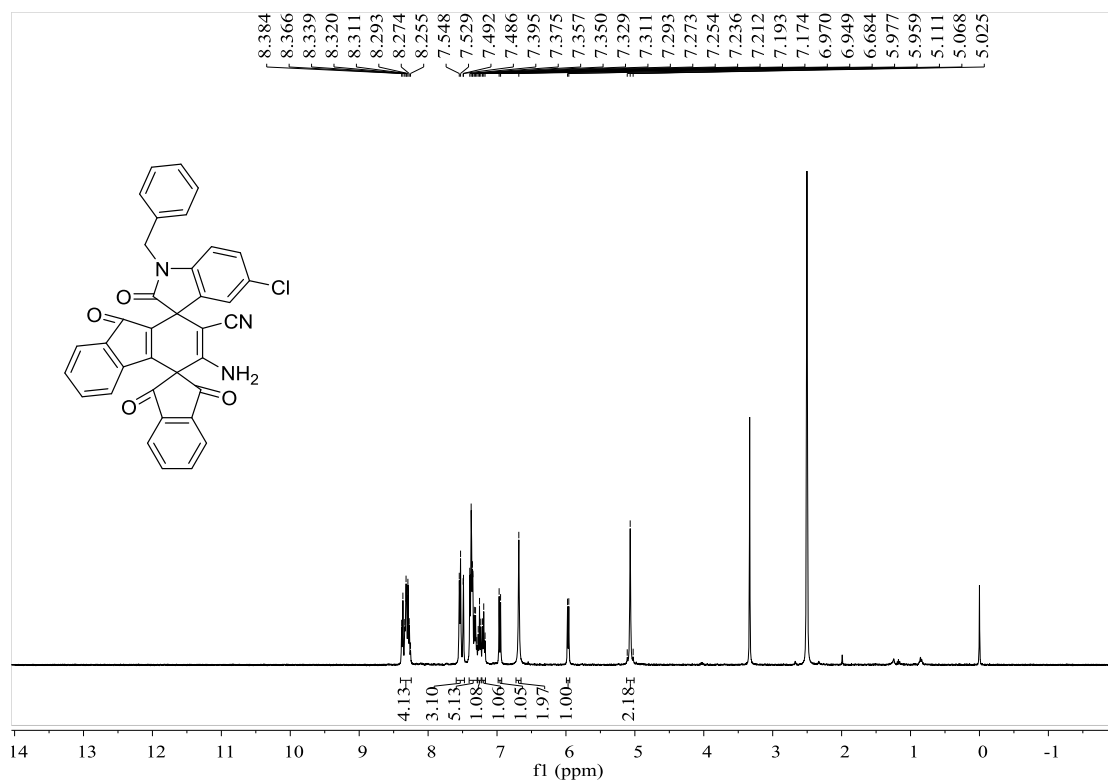


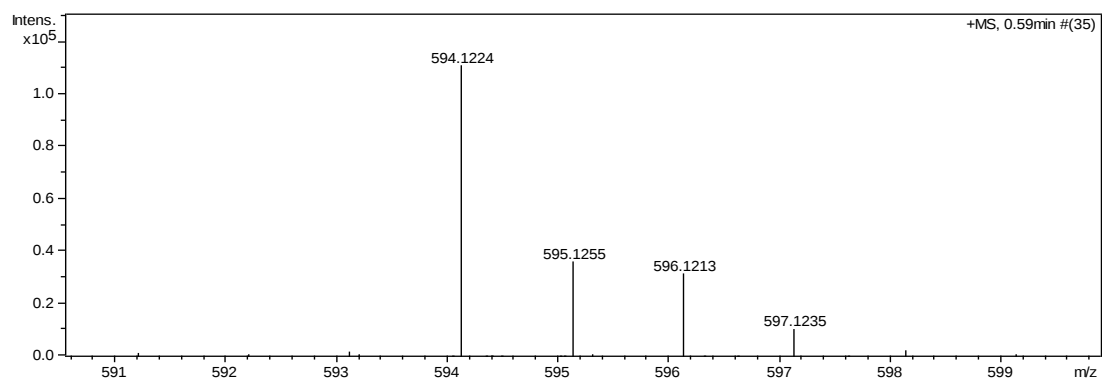
3'-amino-1''-benzyl-5''-methyl-1,2'',3,9'-tetraoxo-1,3-dihydro-9'*H*-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carbonitrile (3b):



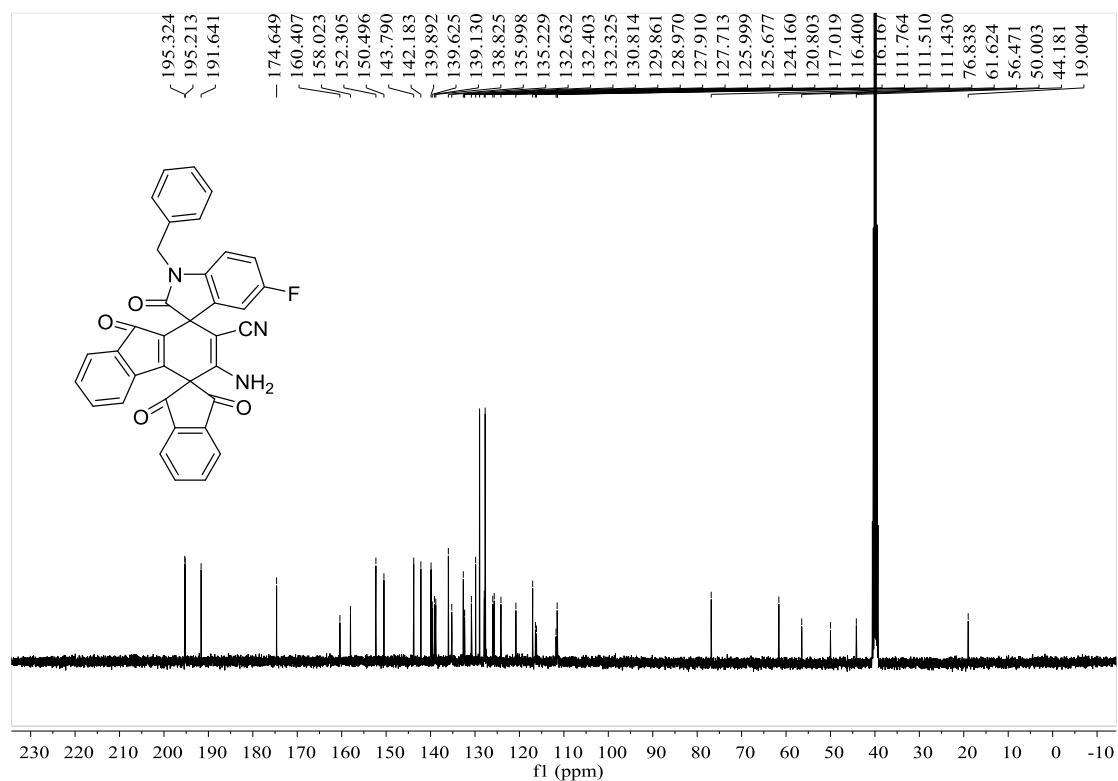
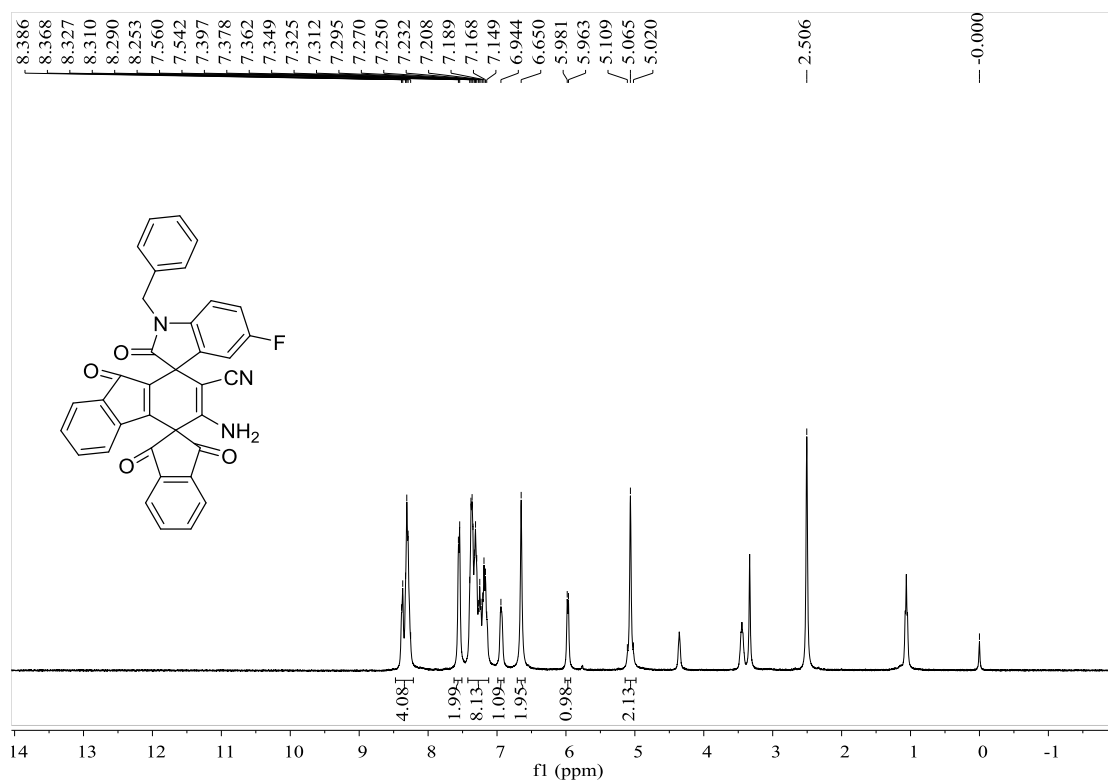


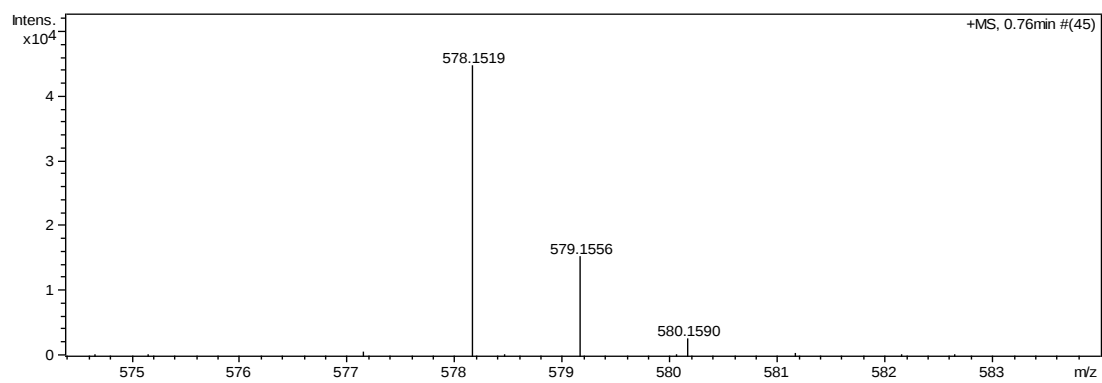
3'-amino-1''-benzyl-5''-chloro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'H-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carbonitrile (3c):



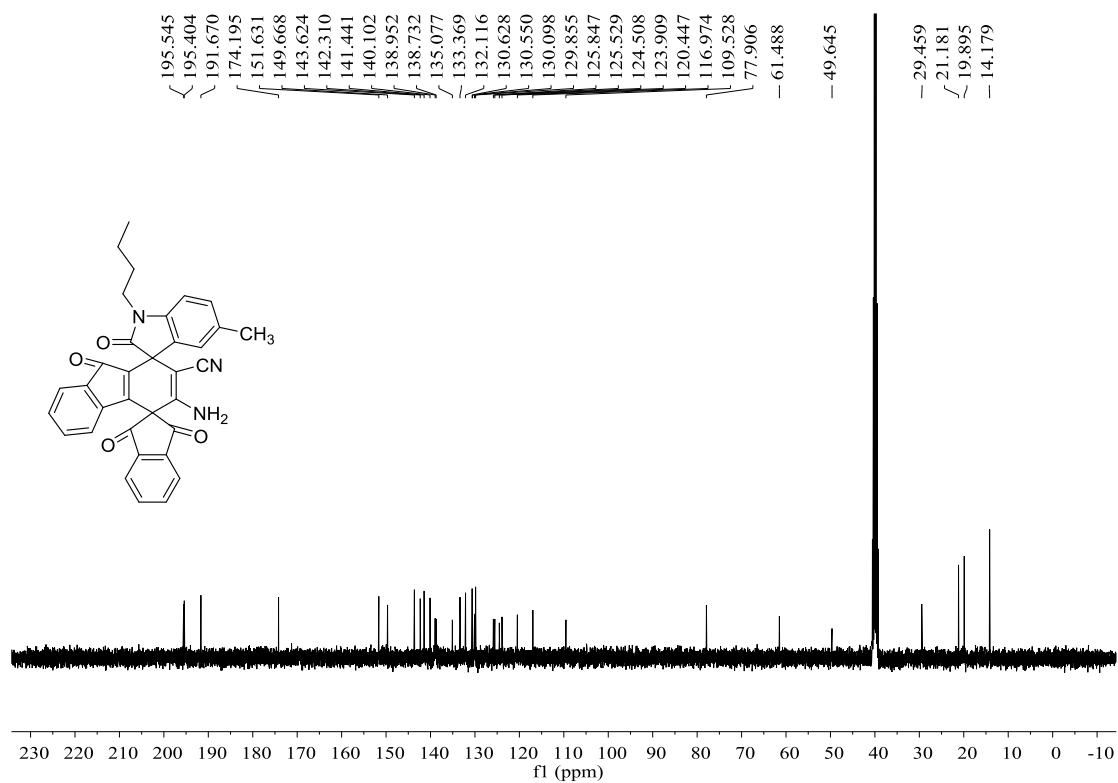
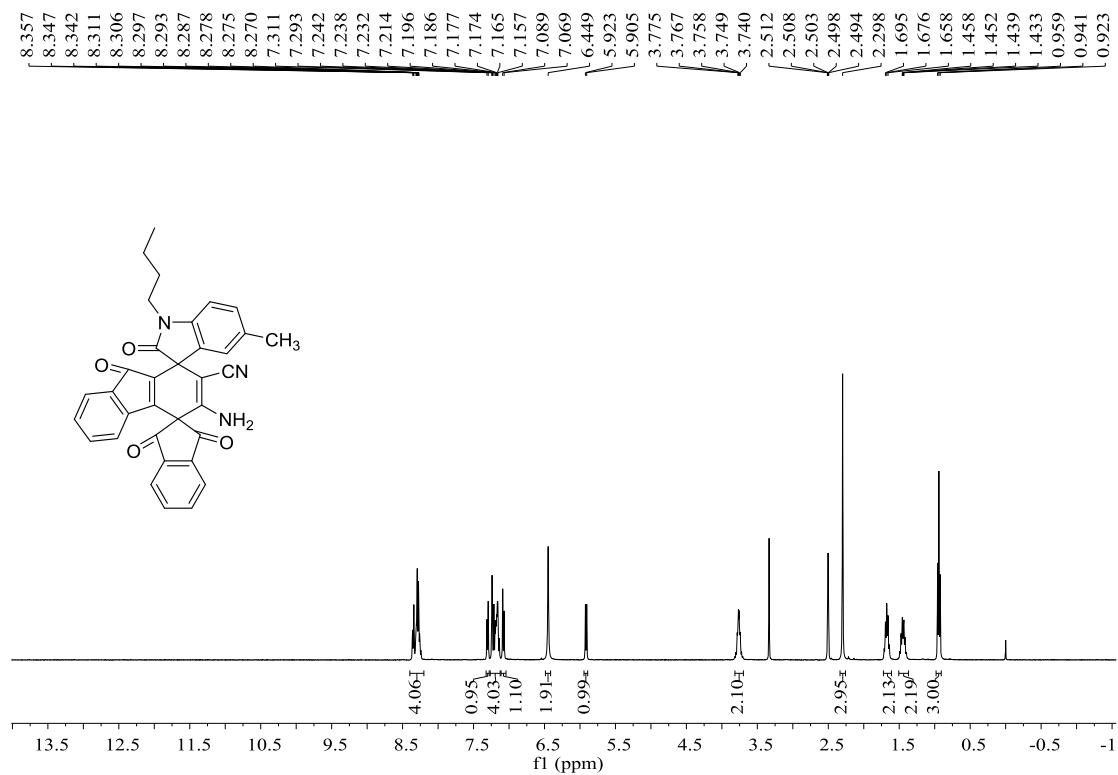


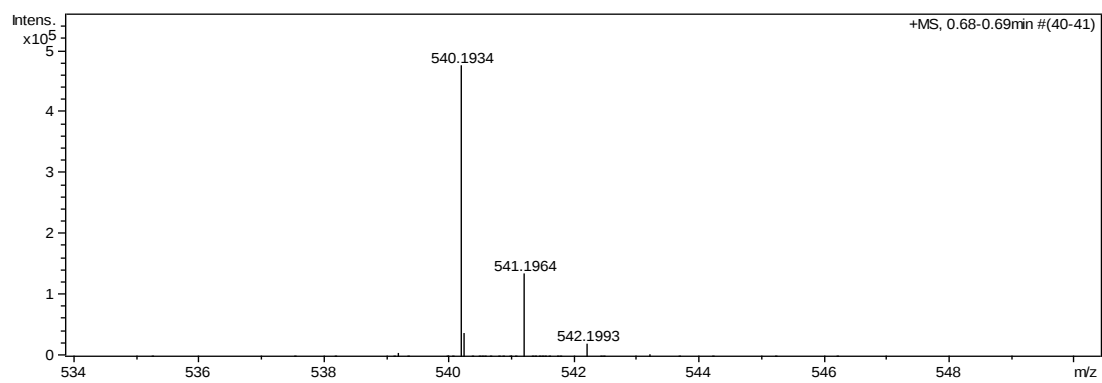
3'-amino-1''-benzyl-5''-fluoro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'H-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carbonitrile (3d):



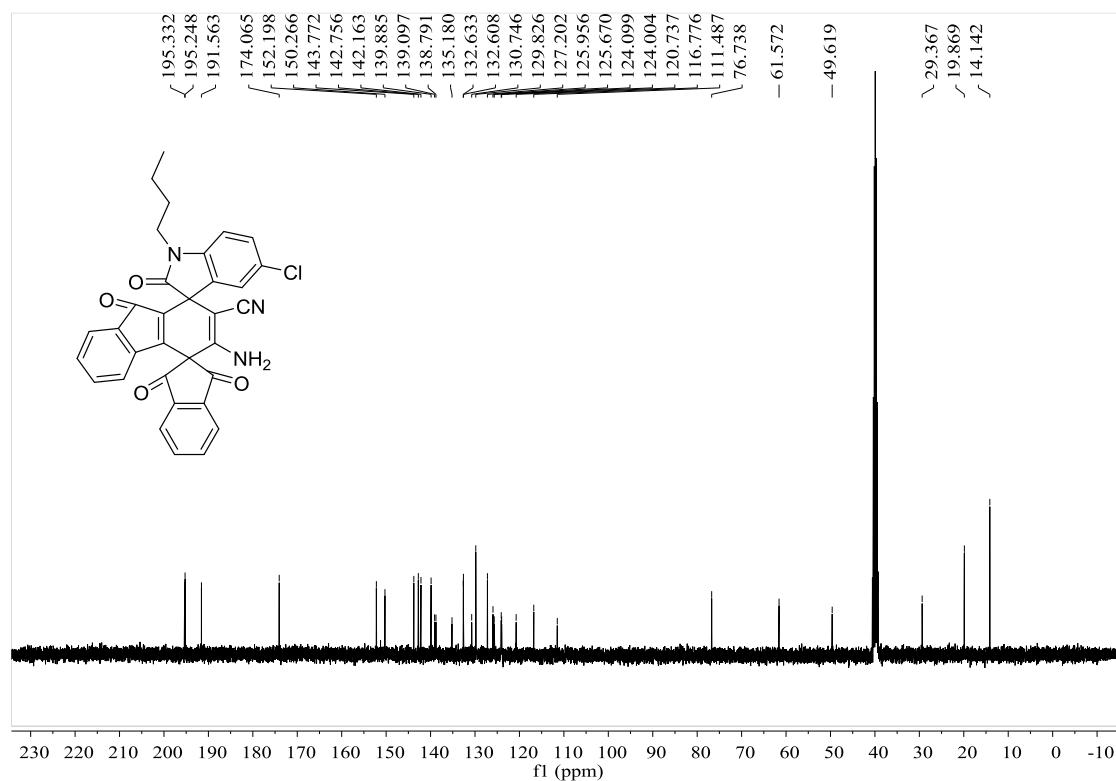
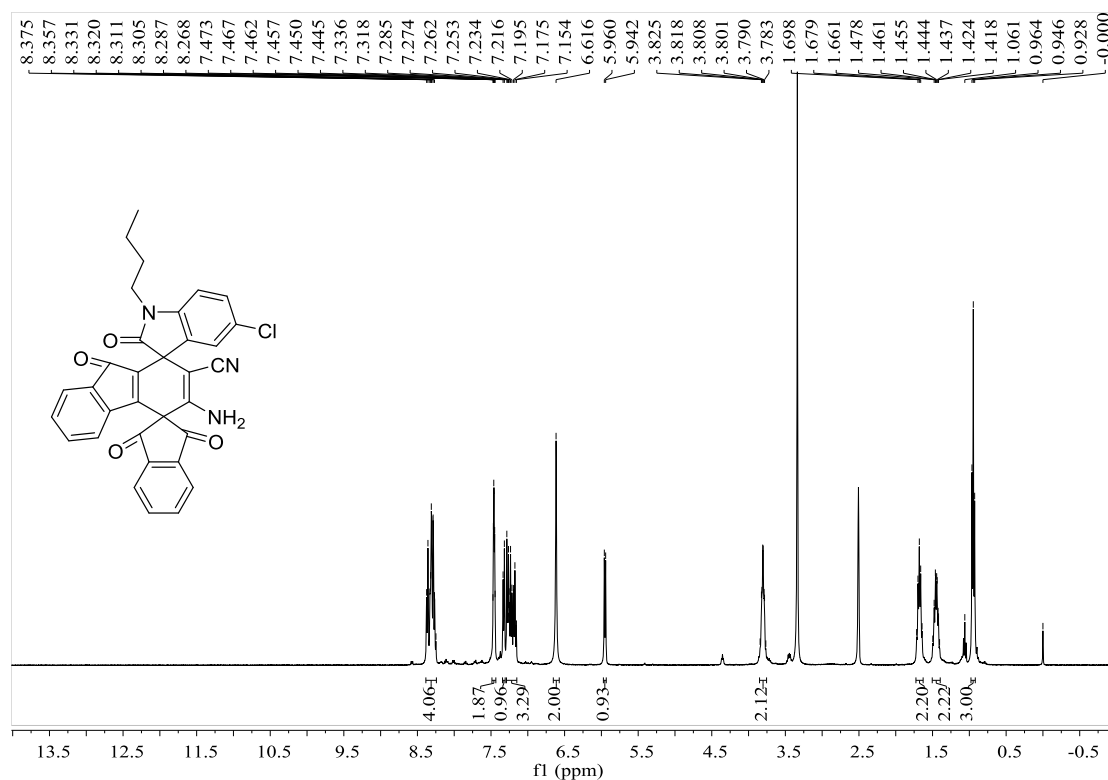


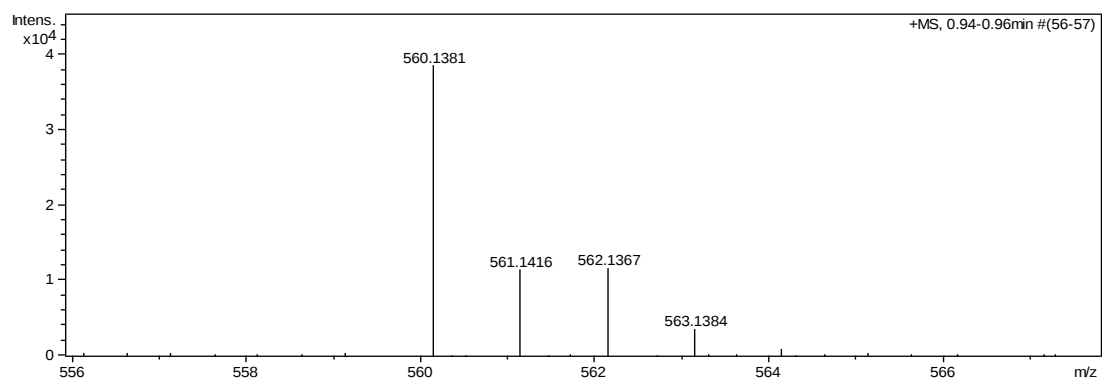
3'-amino-1''-butyl-5''-methyl-1,2'',3,9'-tetraoxo-1,3-dihydro-9'*H*-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carbonitrile (3e):



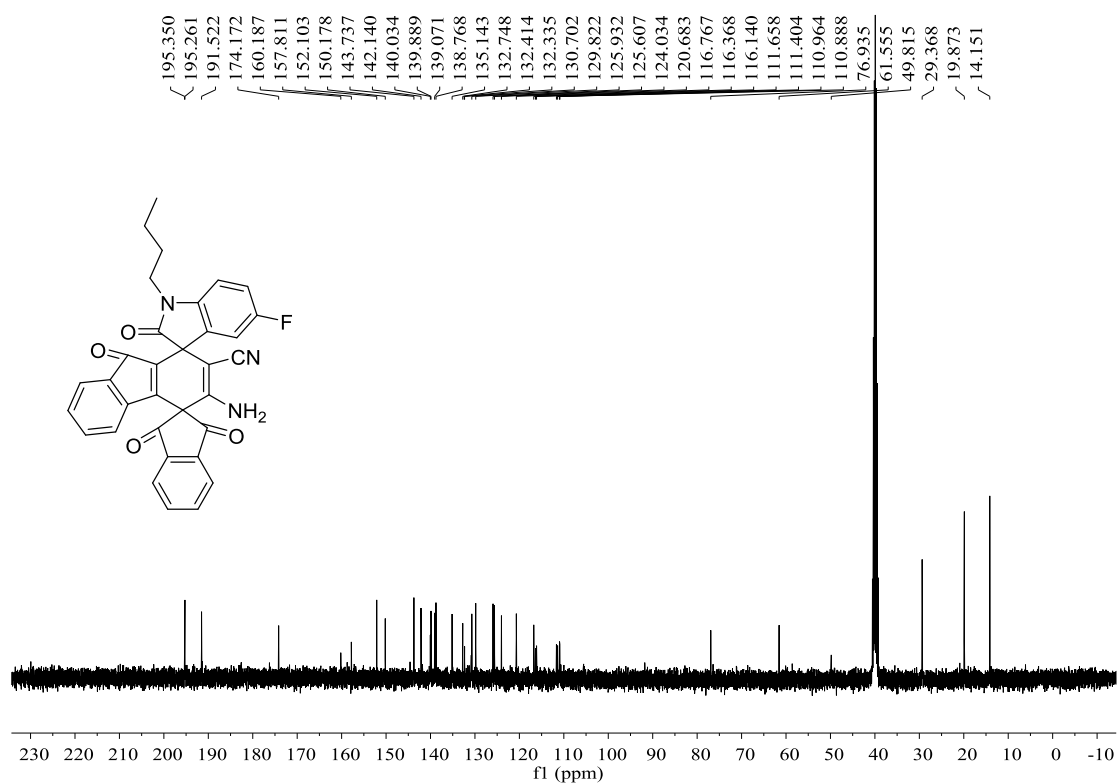
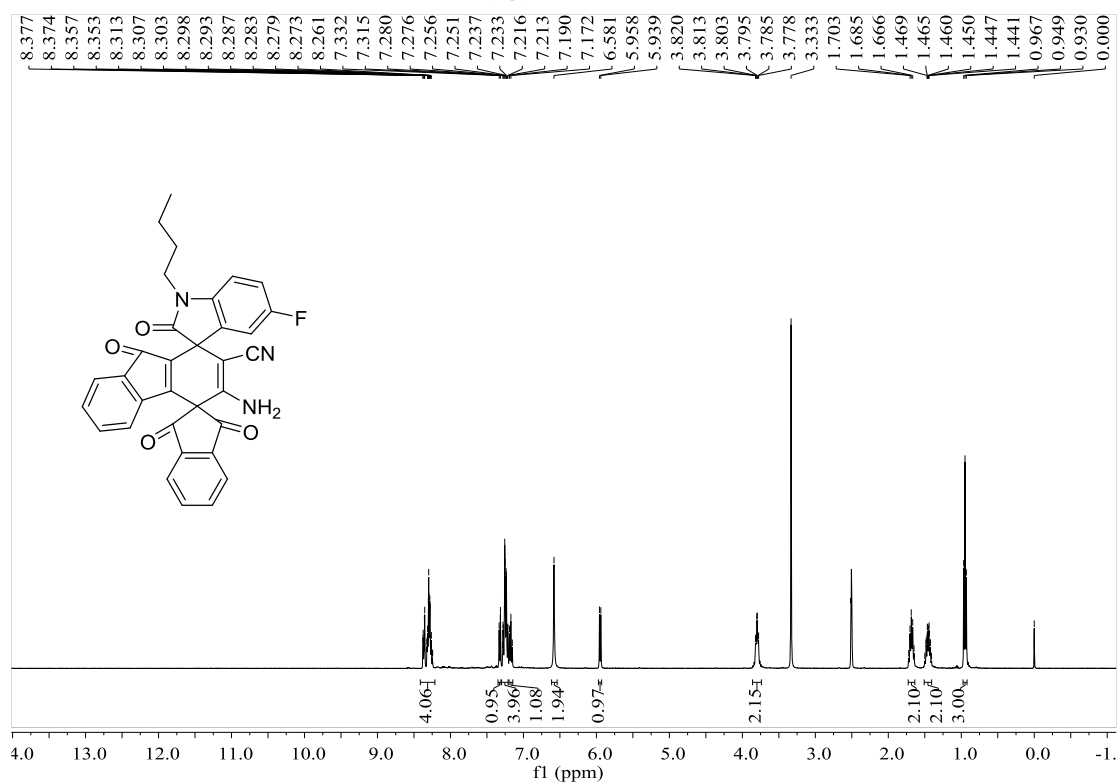


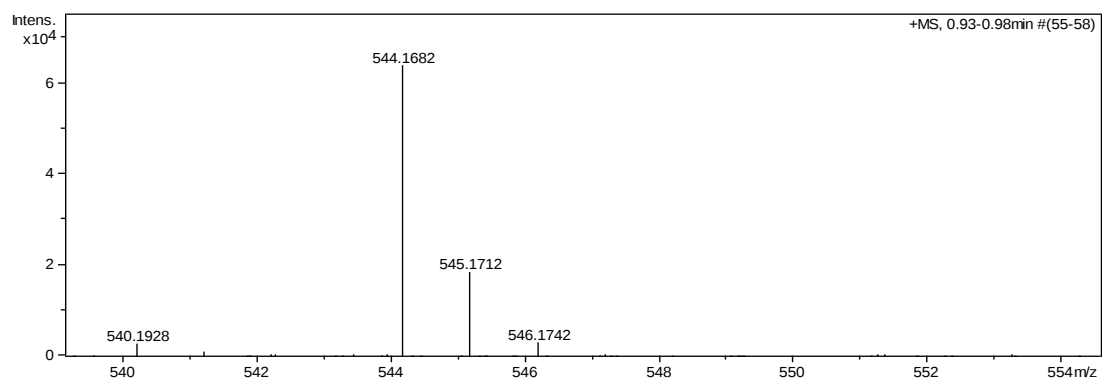
3'-amino-1''-butyl-5''-chloro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'*H*-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carbonitrile (3f):



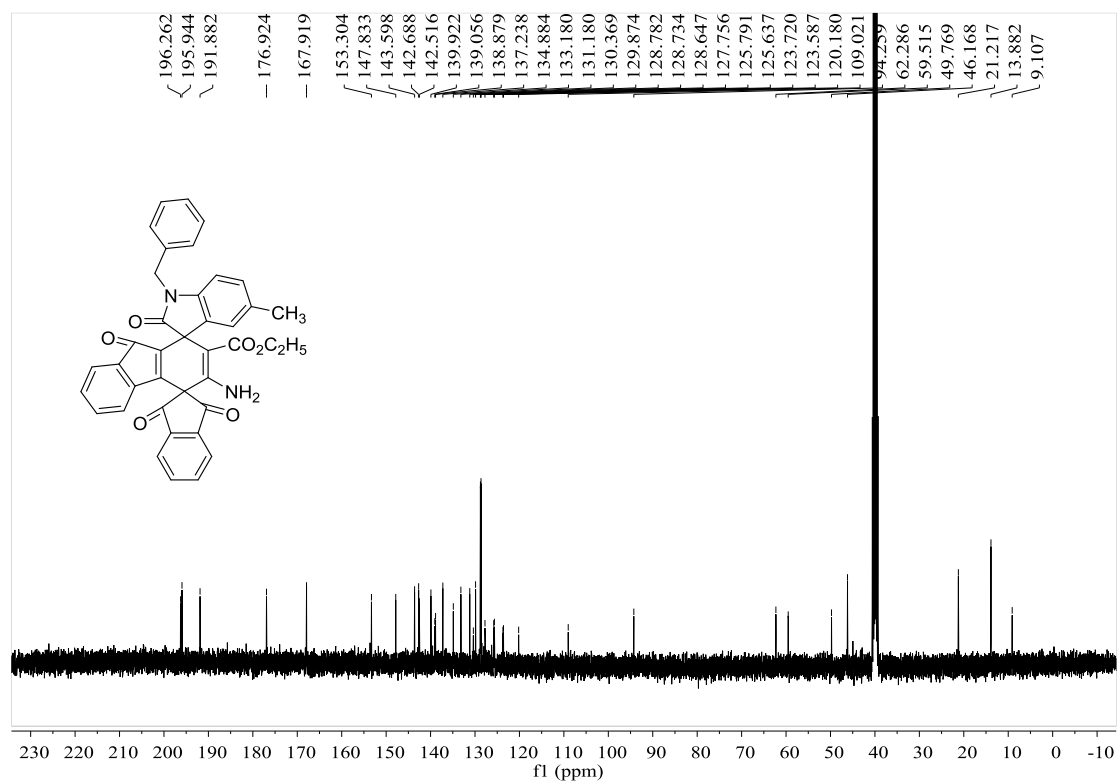
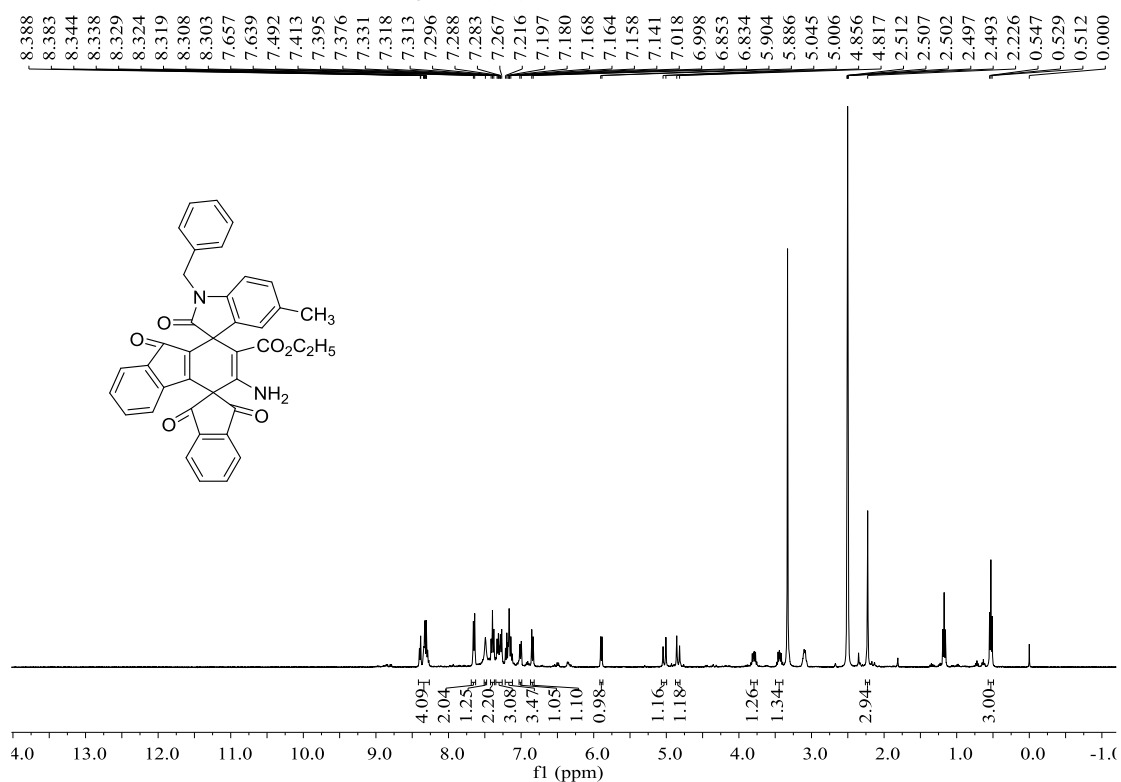


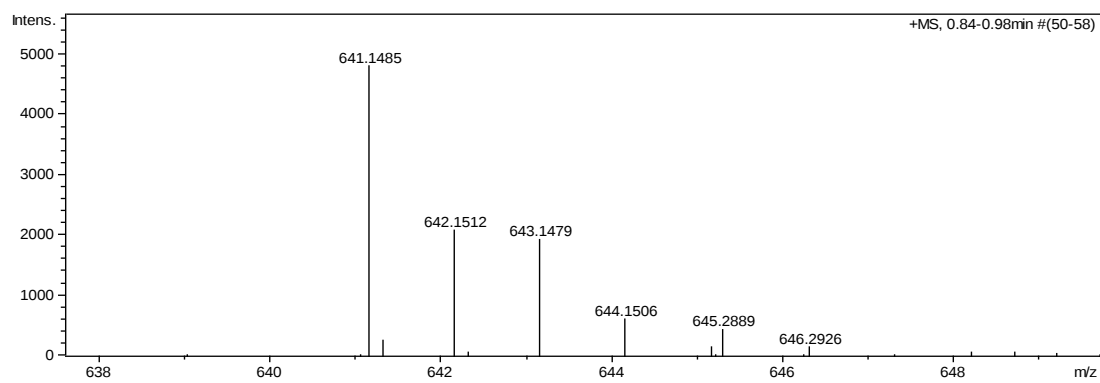
3'-amino-1''-butyl-5''-fluoro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'H-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carbonitrile (3g):



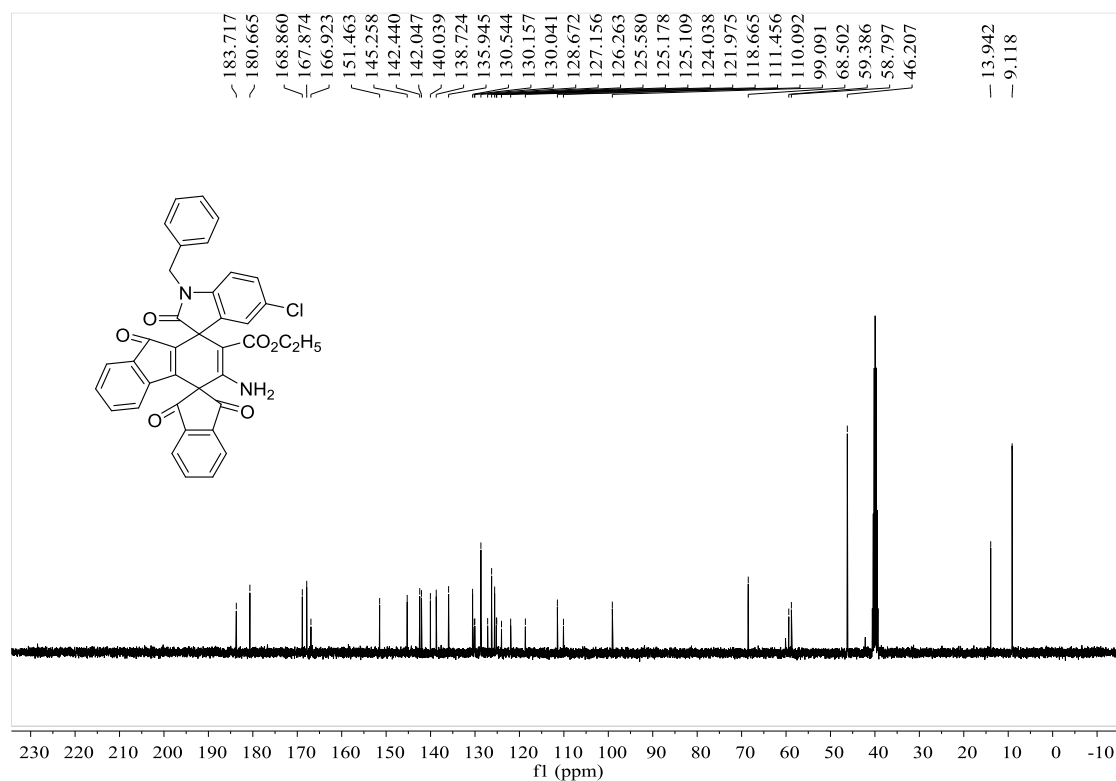
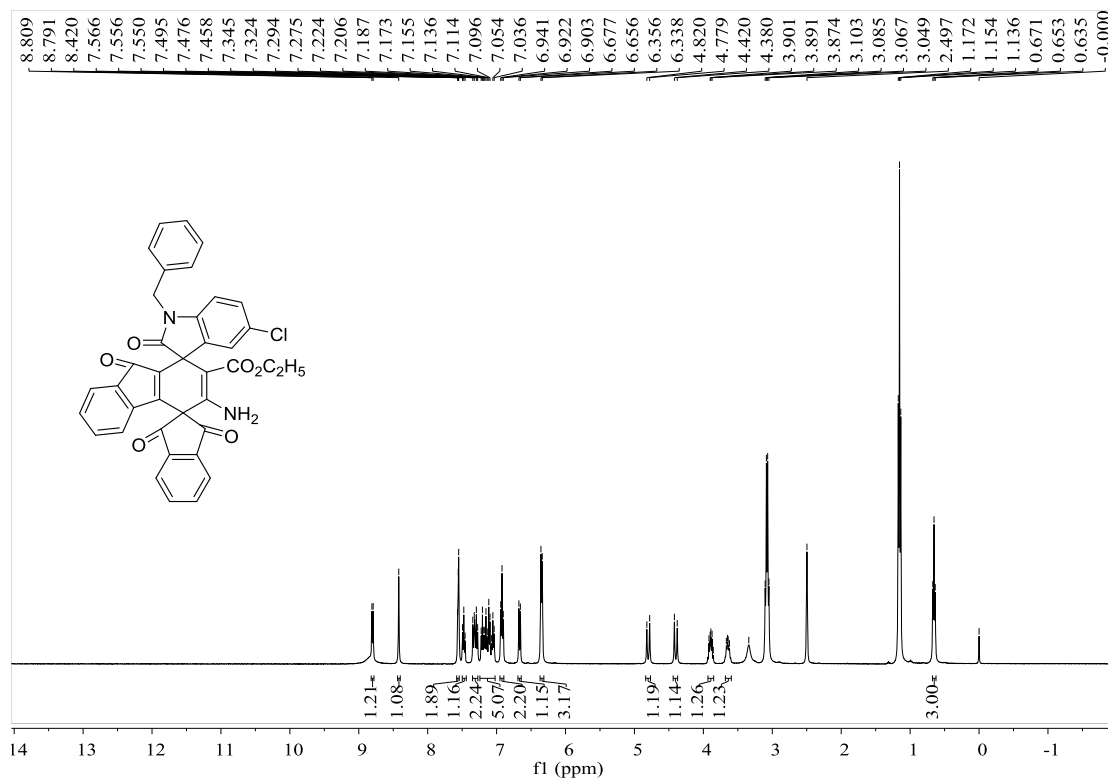


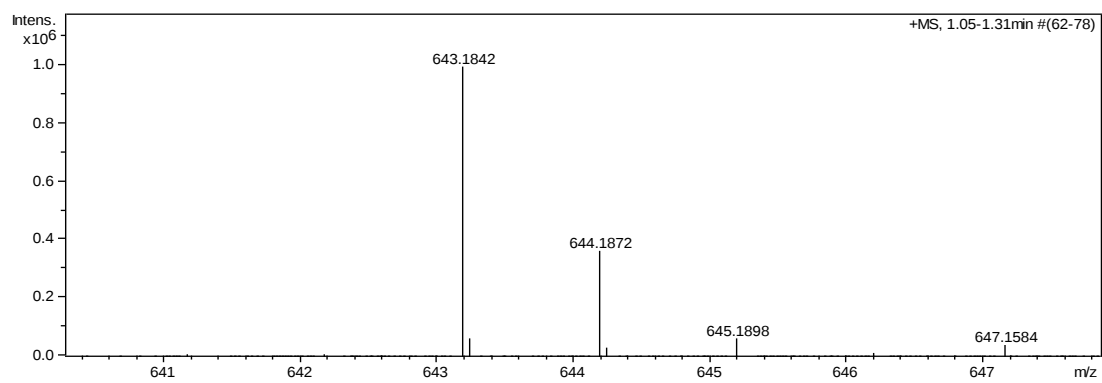
Ethyl 3'-amino-1''-benzyl-5''-methyl-1,2'',3,9'-tetraoxo-1,3-dihydro-9'*H*-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carboxylate (3h):



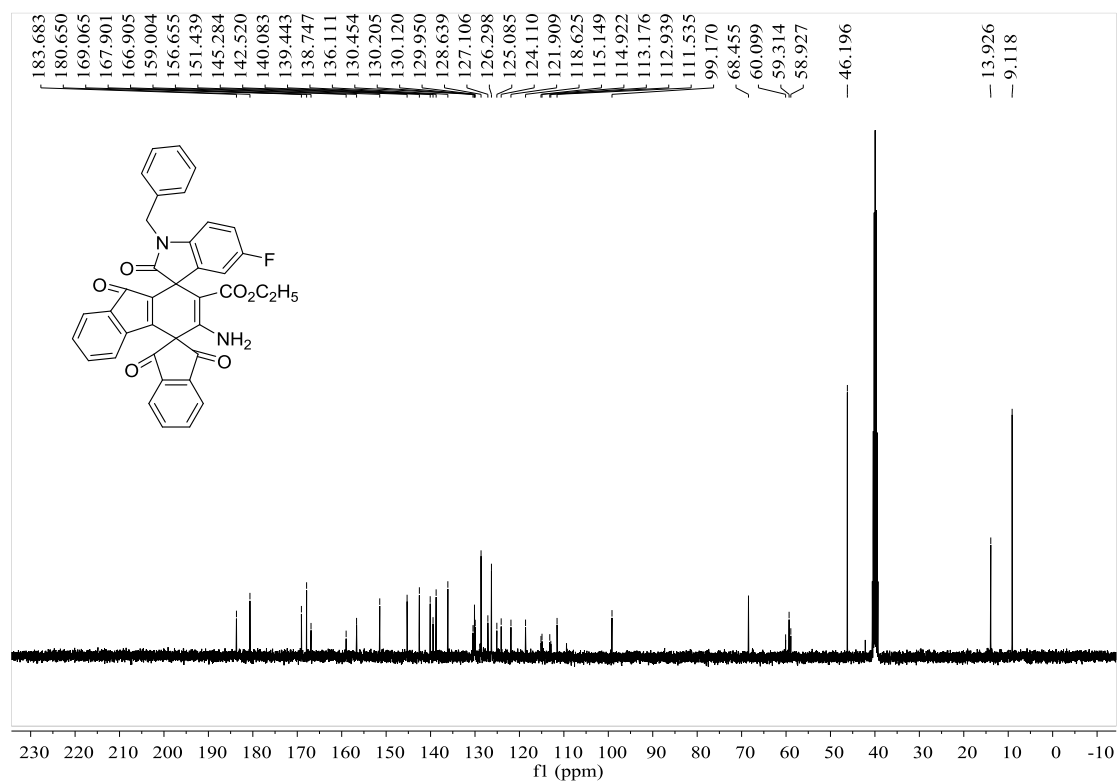
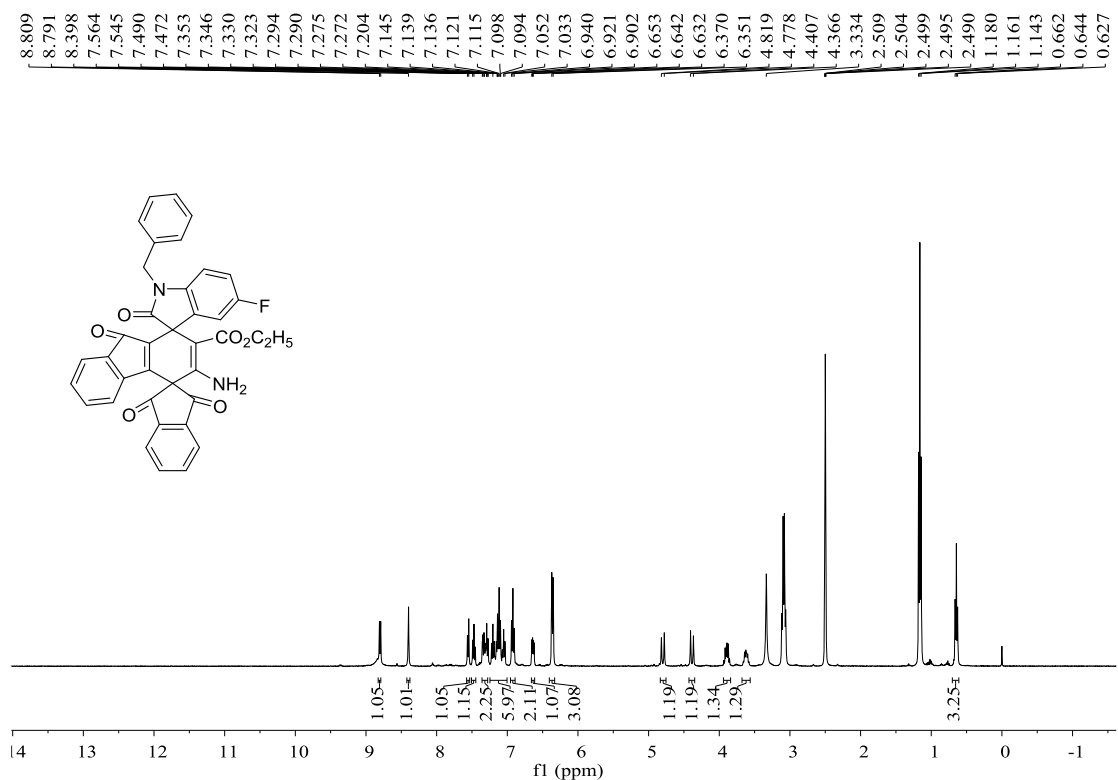


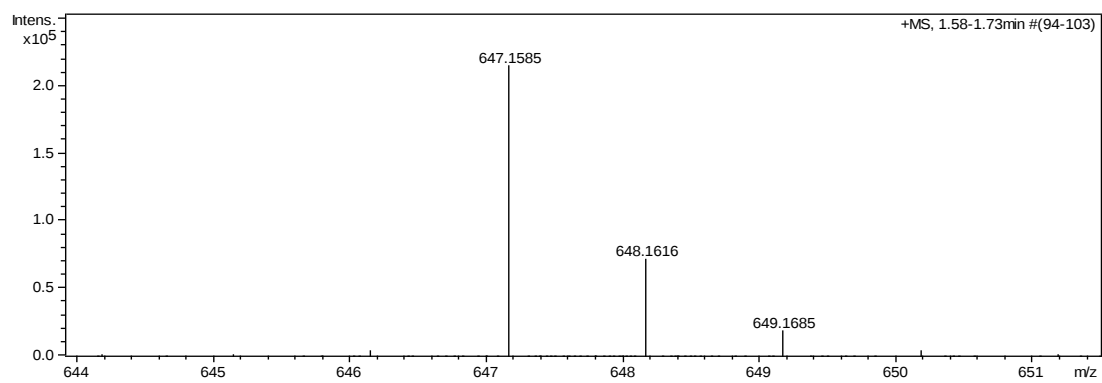
Ethyl 3'-amino-1''-benzyl-5''-chloro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'*H*-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carboxylate (3i):



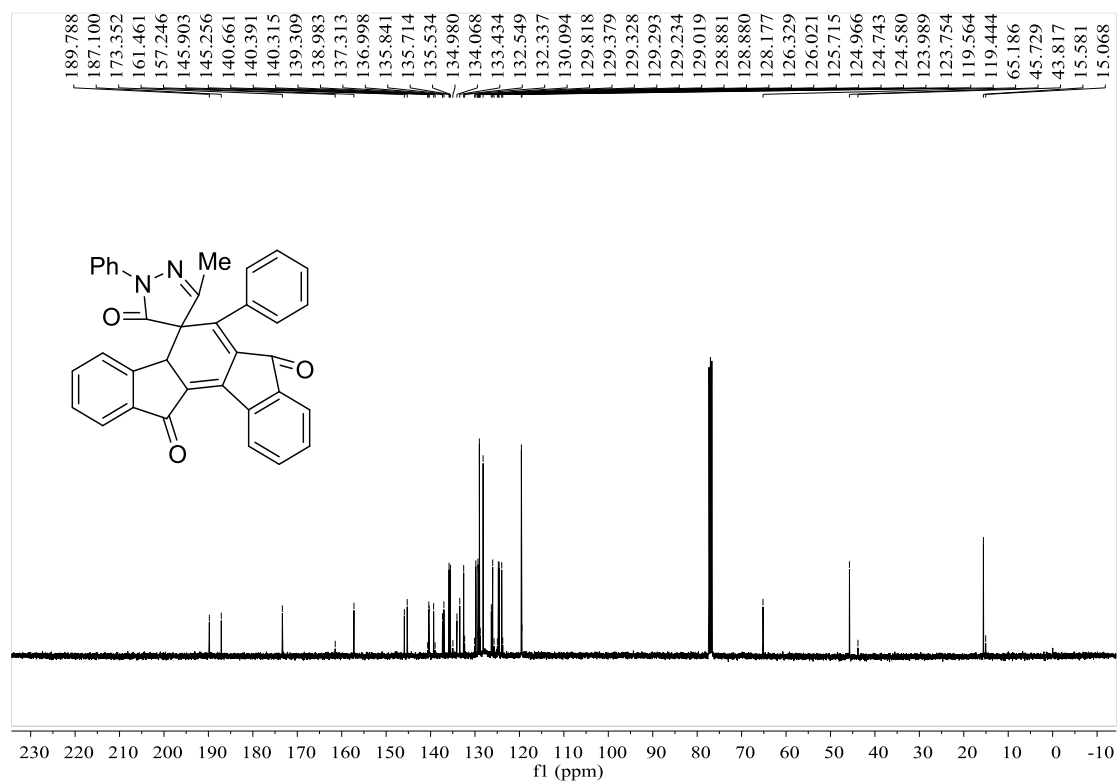
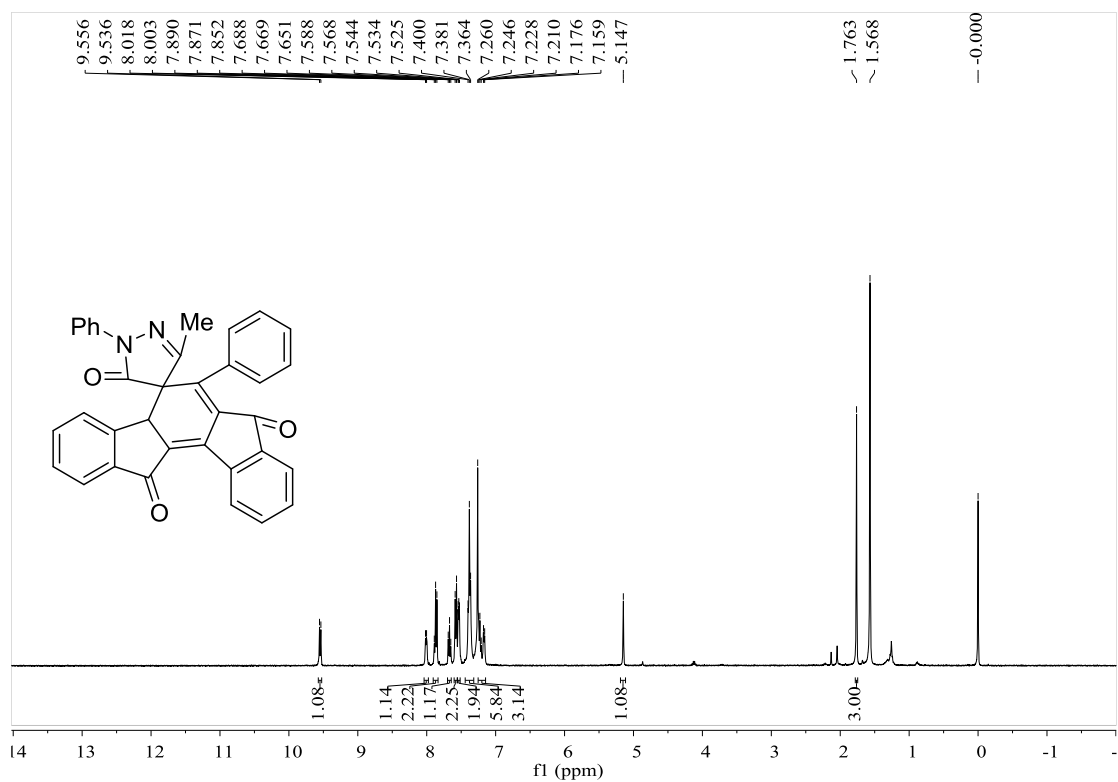


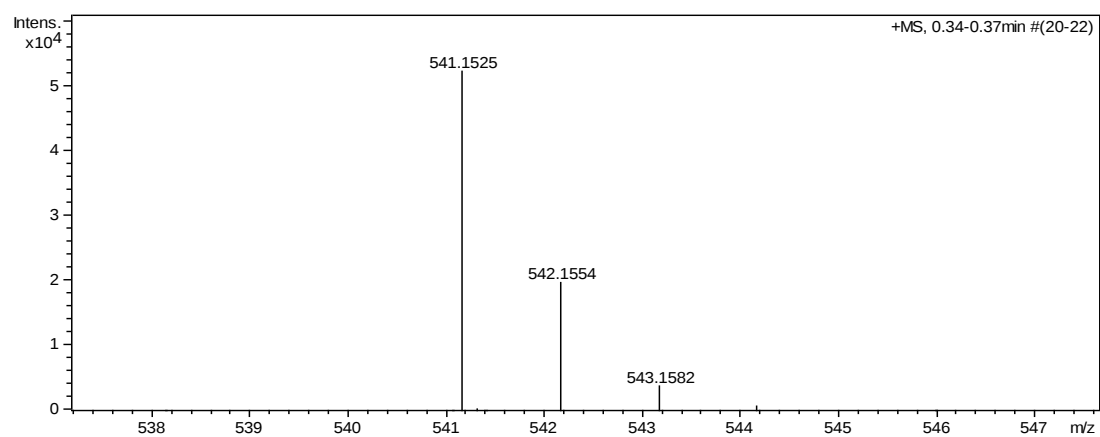
Ethyl 3'-amino-1''-benzyl-5''-fluoro-1,2'',3,9'-tetraoxo-1,3-dihydro-9'*H*-dispiro[indene-2,4'-fluorene-1',3''-indoline]-2'-carboxylate (3j):



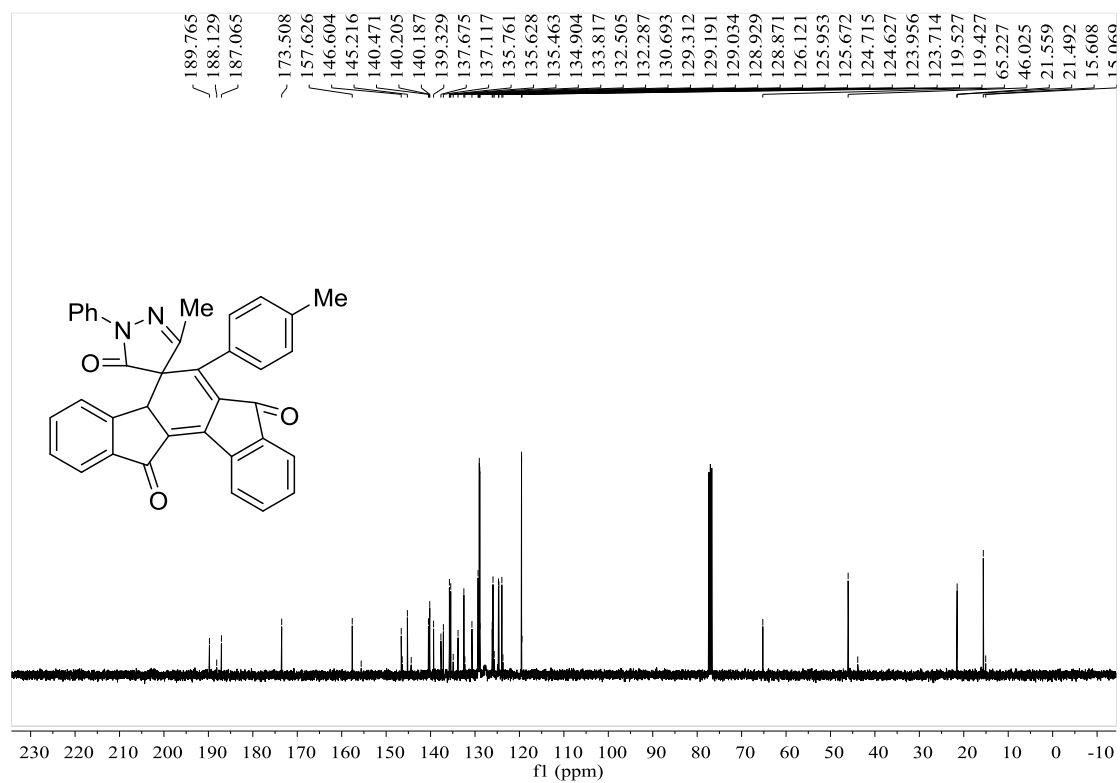
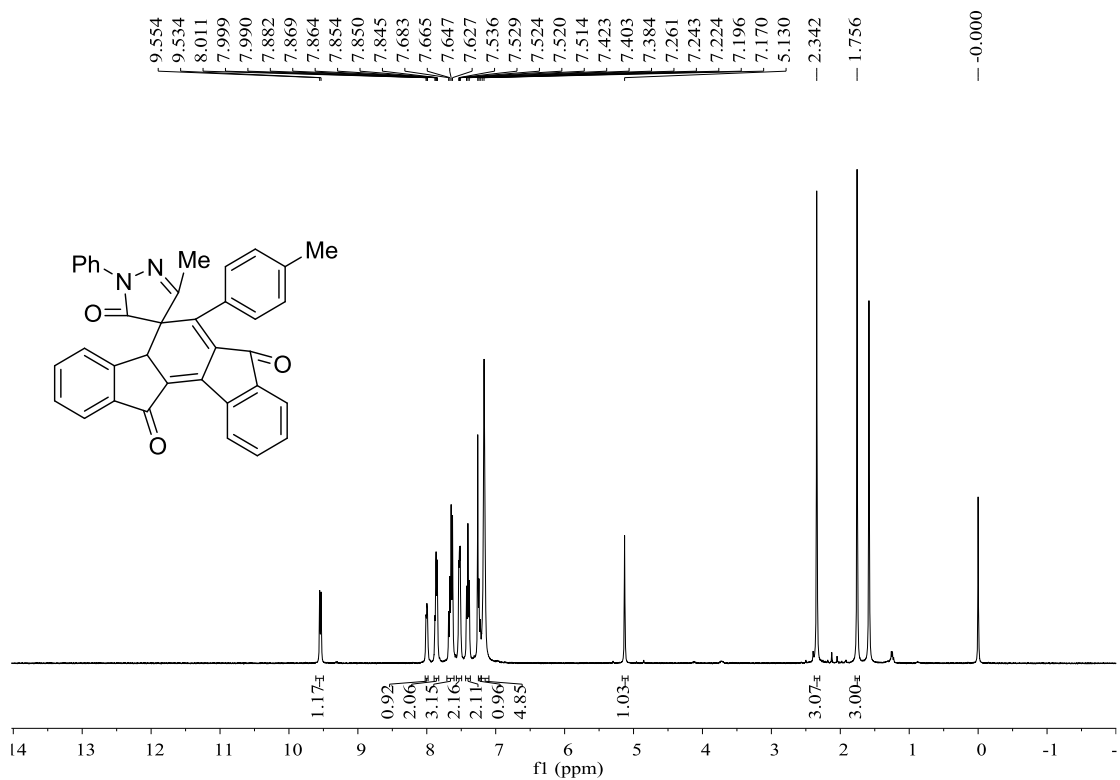


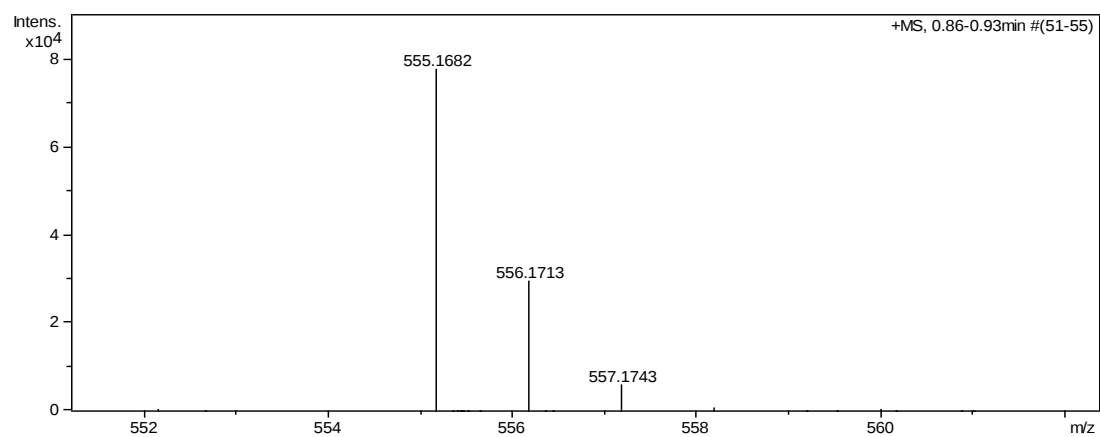
3'-methyl-1',6-diphenyl-7*H*-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5a):



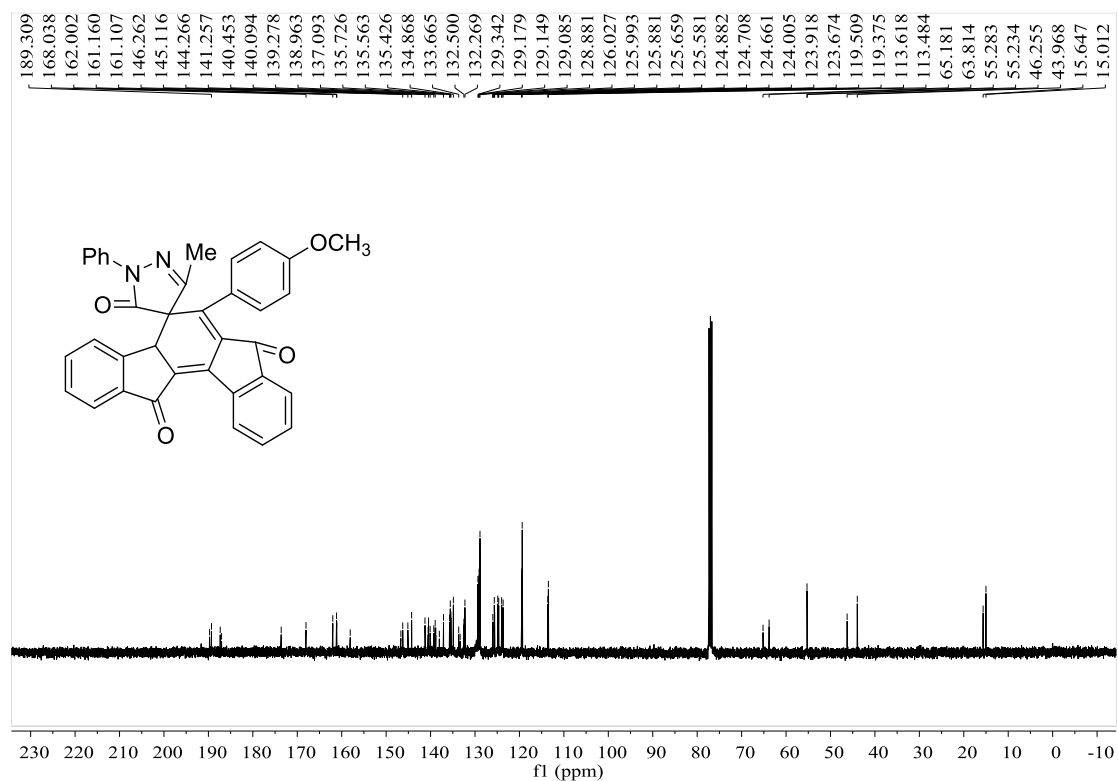
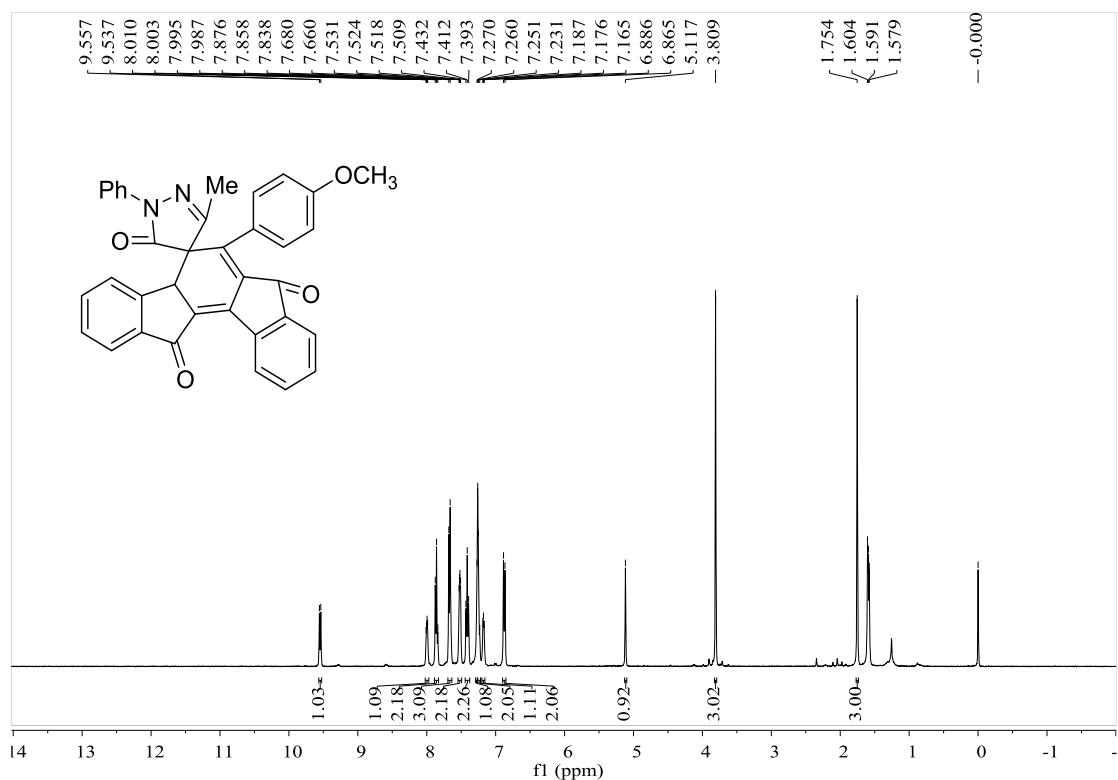


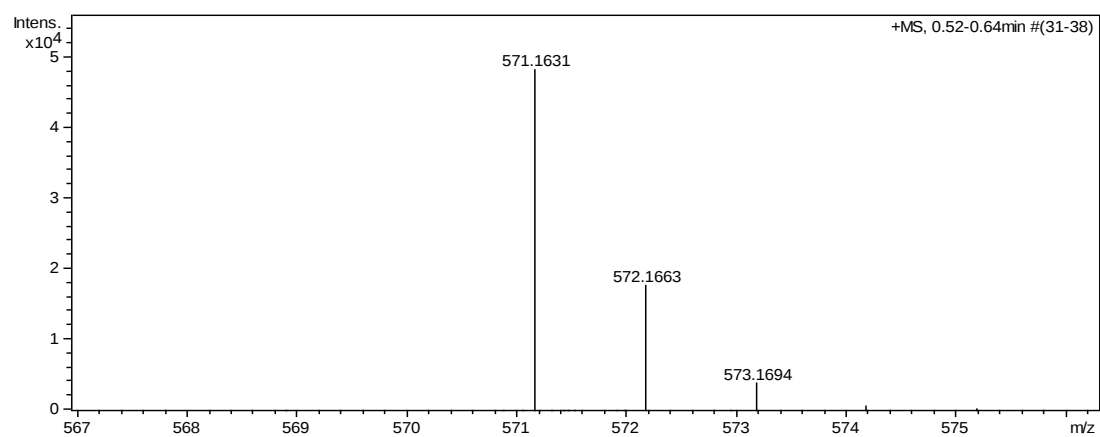
3'-methyl-1'-phenyl-6-(p-tolyl)-7*H*-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5b):



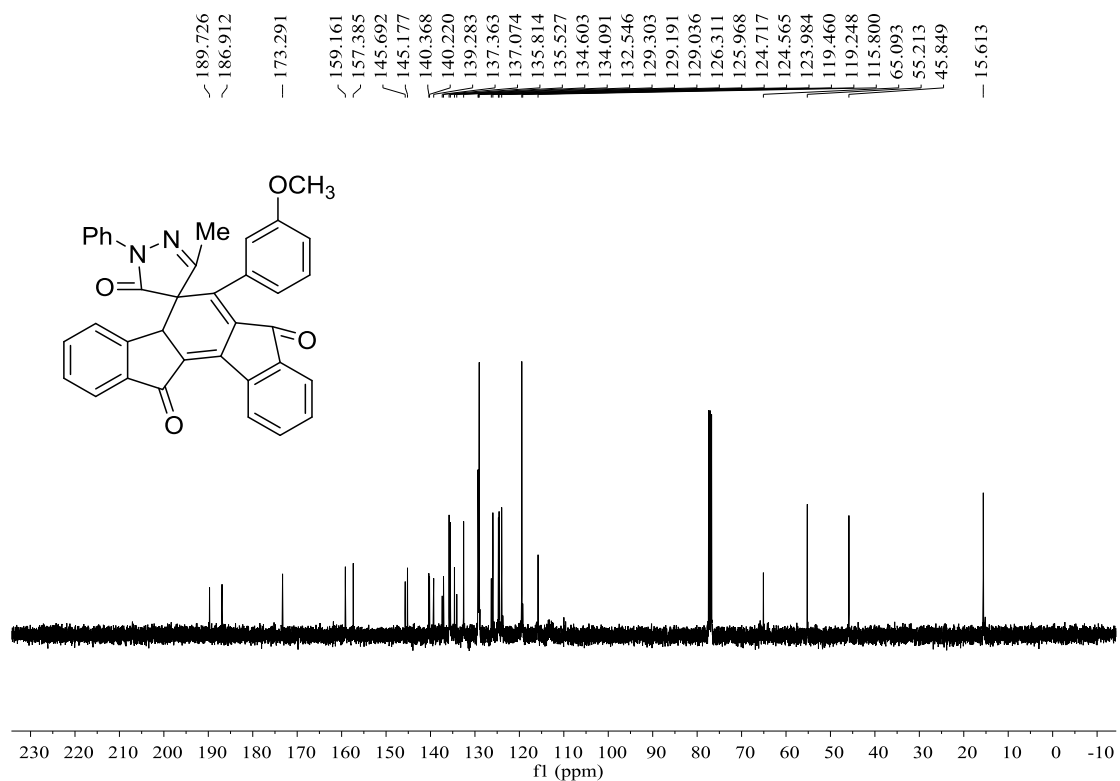
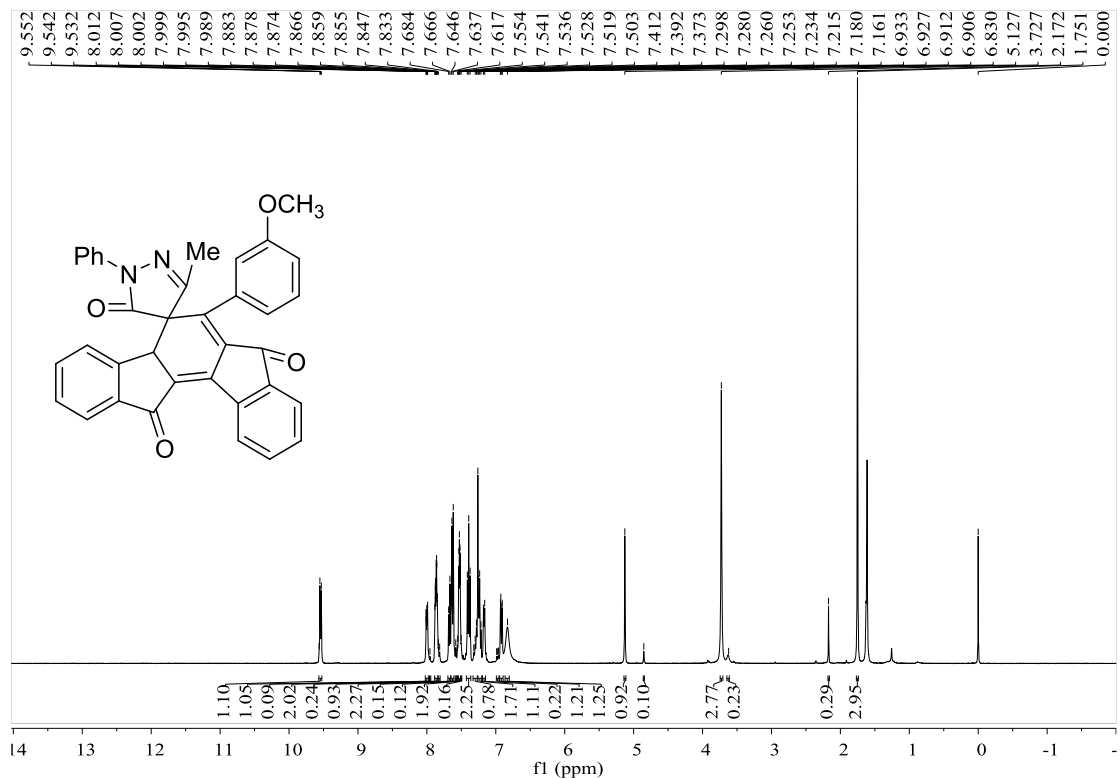


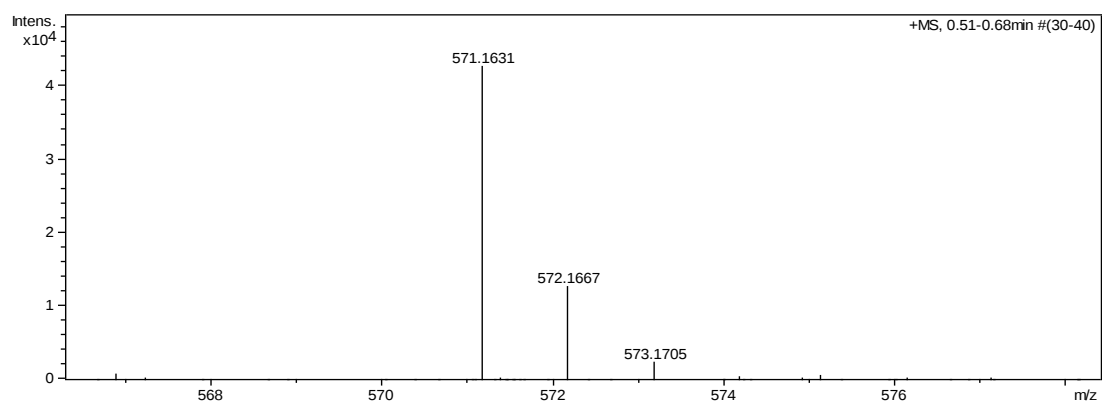
6-(4-methoxyphenyl)-3'-methyl-1'-phenyl-7*H*-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5c):



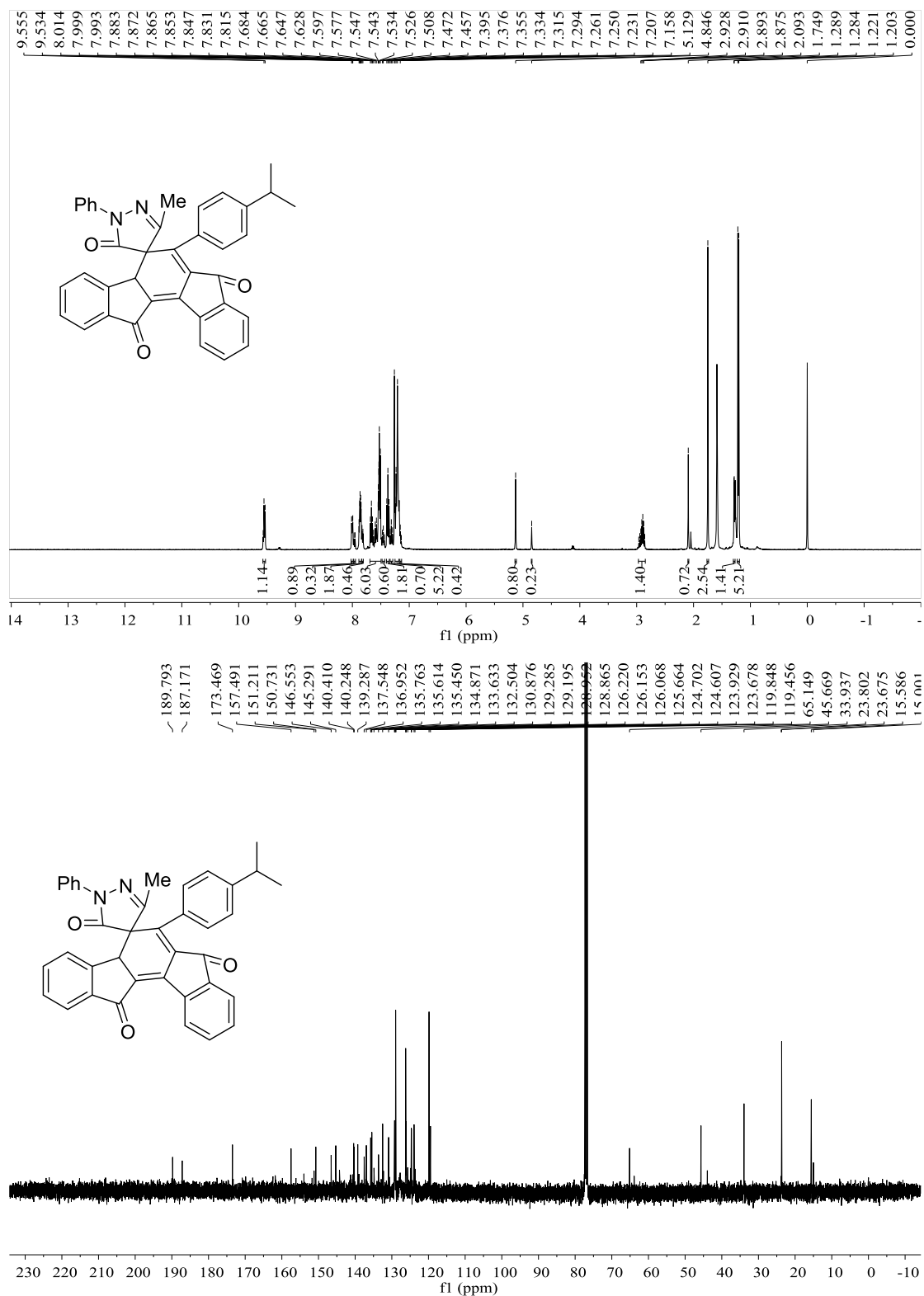


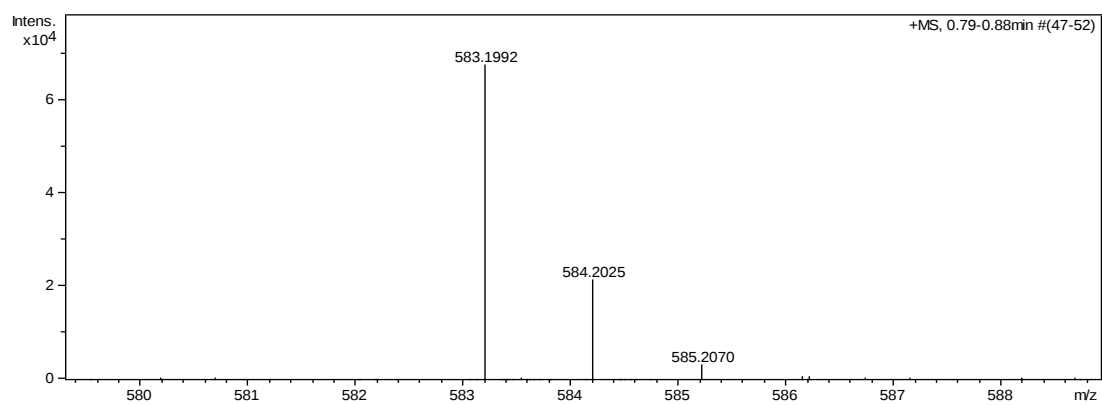
6-(3-methoxyphenyl)-3'-methyl-1'-phenyl-7*H*-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5d):



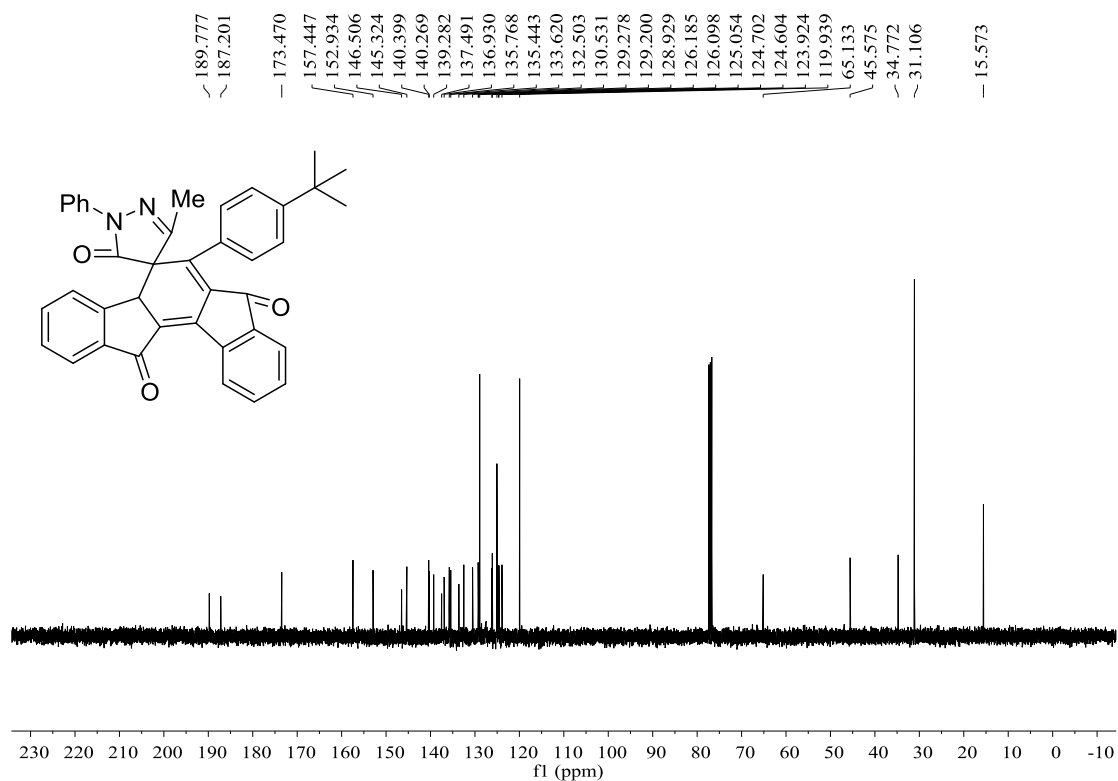
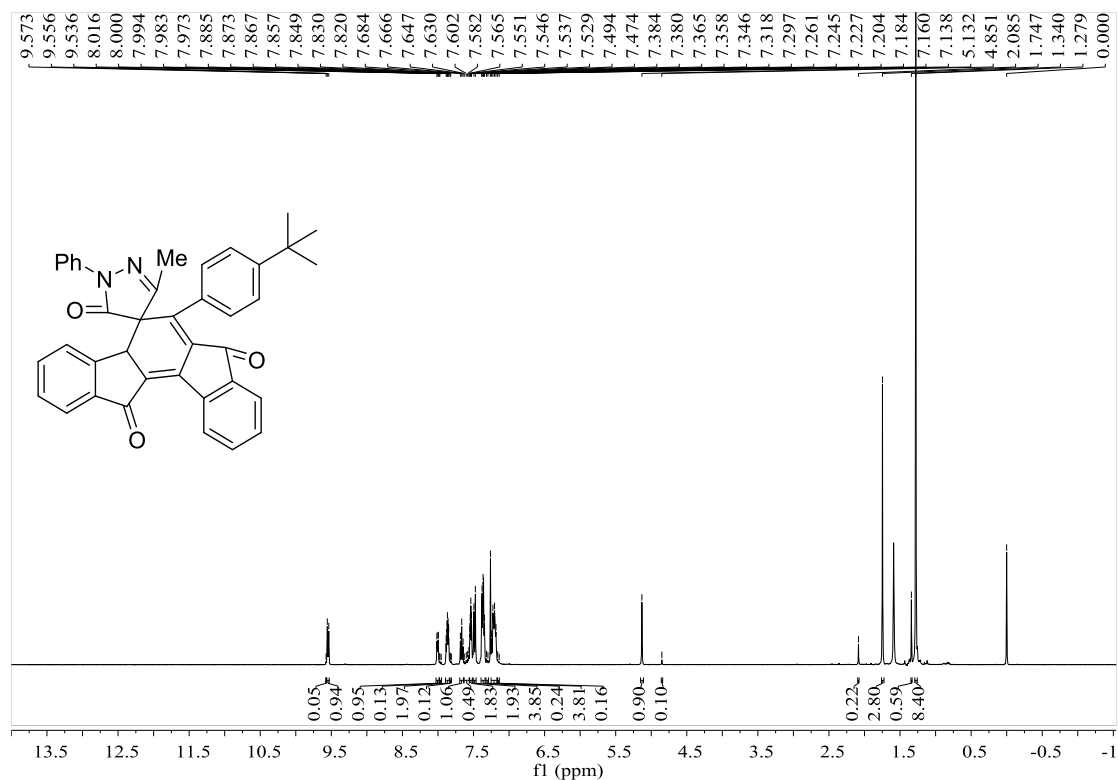


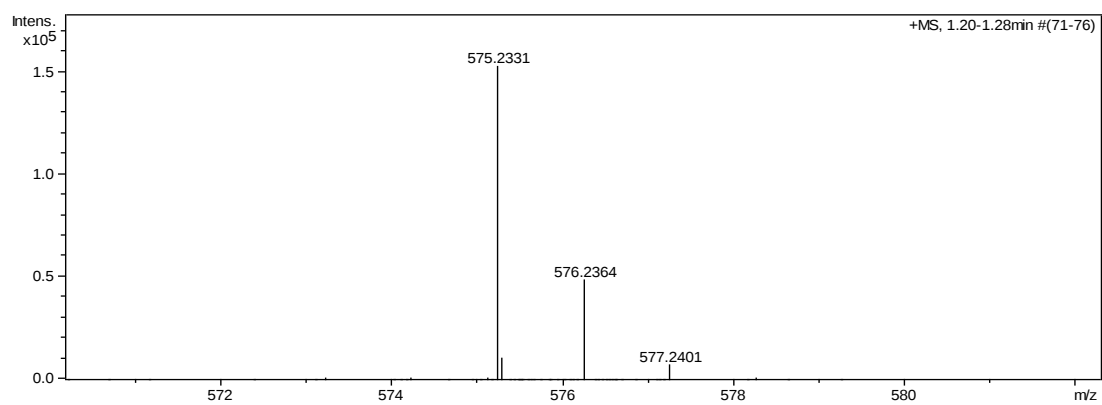
6-(4-isopropylphenyl)-3'-methyl-1'-phenyl-7*H*-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5e):



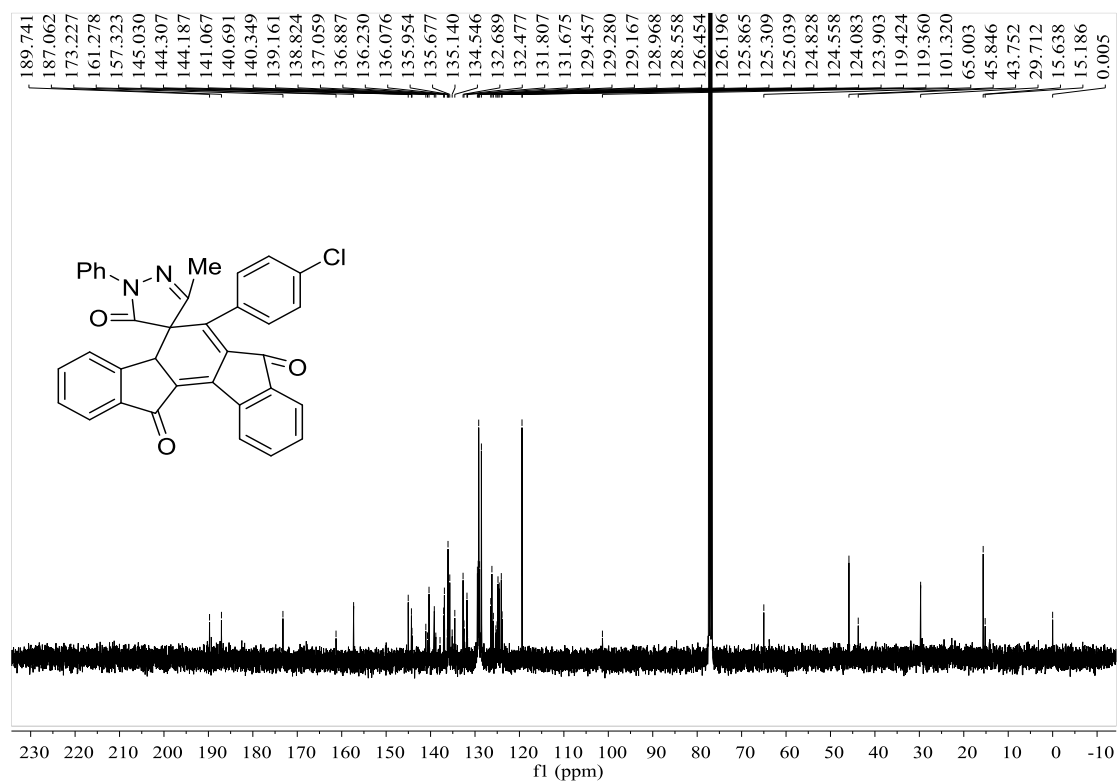
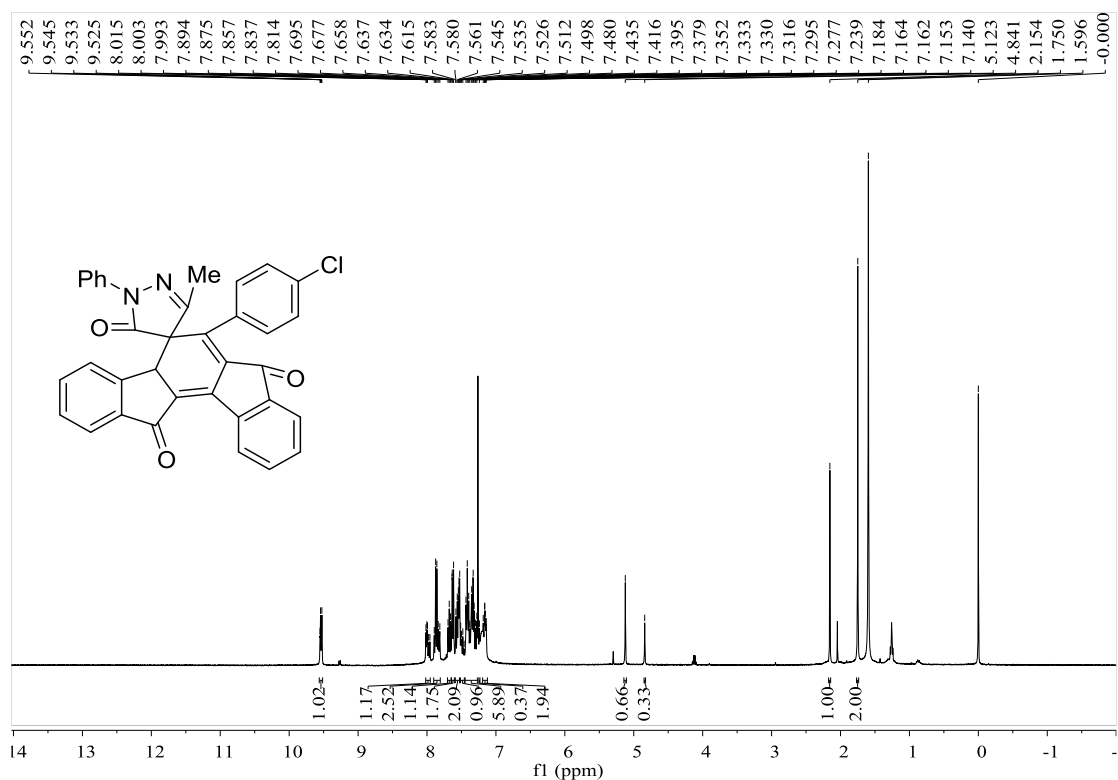


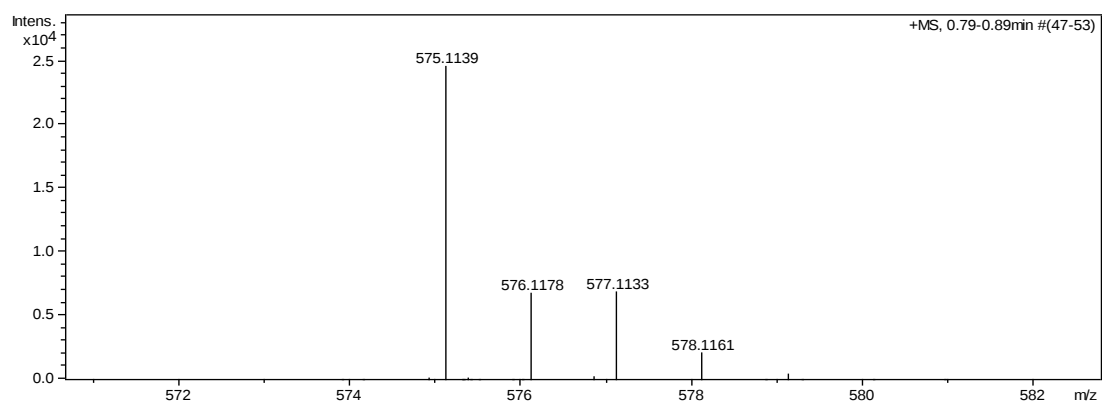
6-(4-(tert-butyl)phenyl)-3'-methyl-1'-phenyl-7H-spiro[indeno[1,2-a]fluorene-5,4'-pyrazole]-5',7,12(1*H*,4*bH*)-trione (5f):



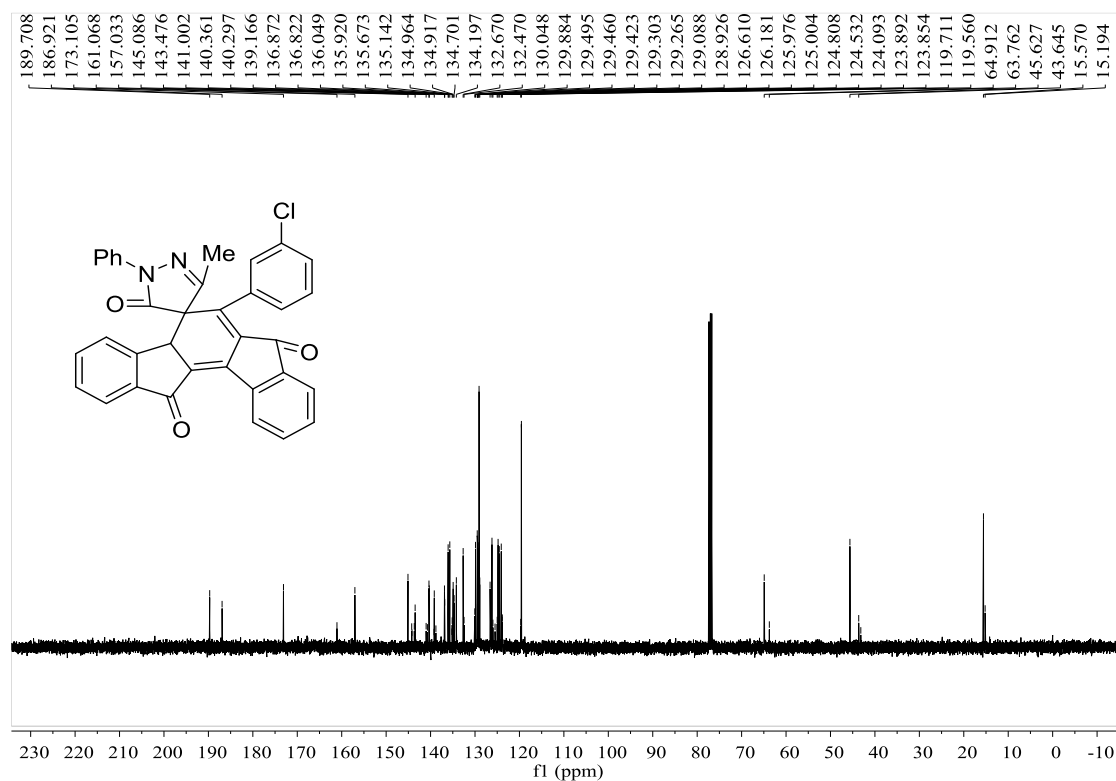
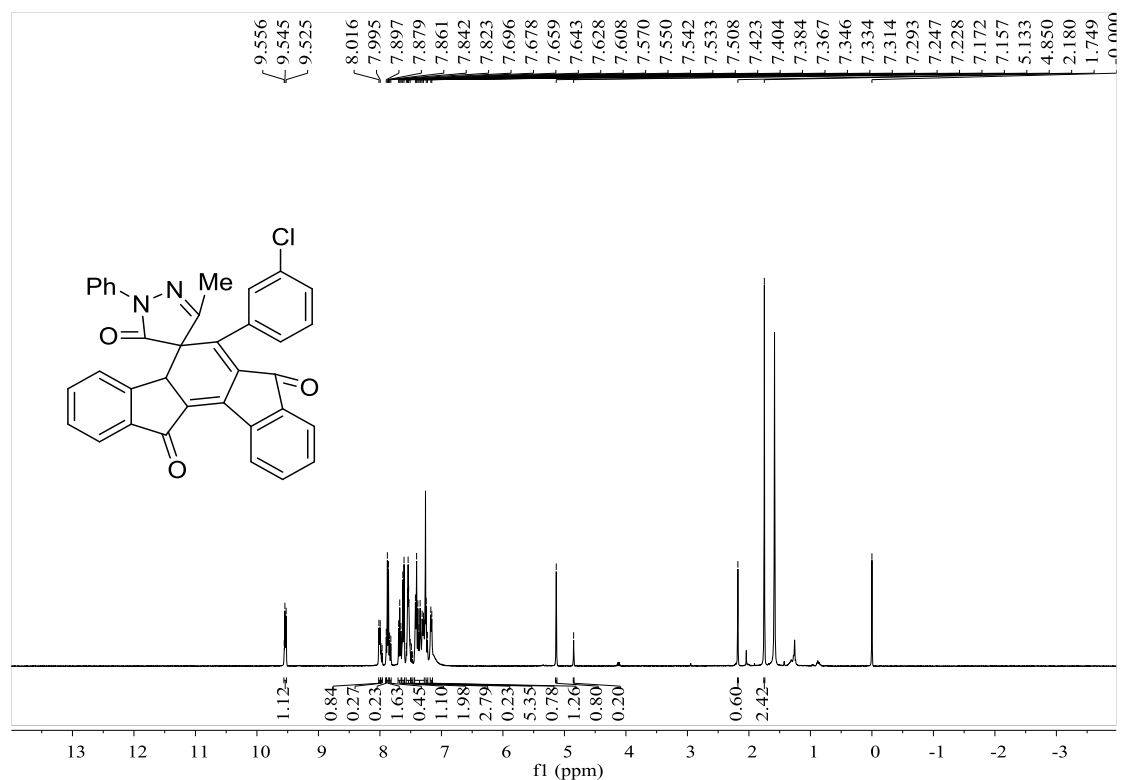


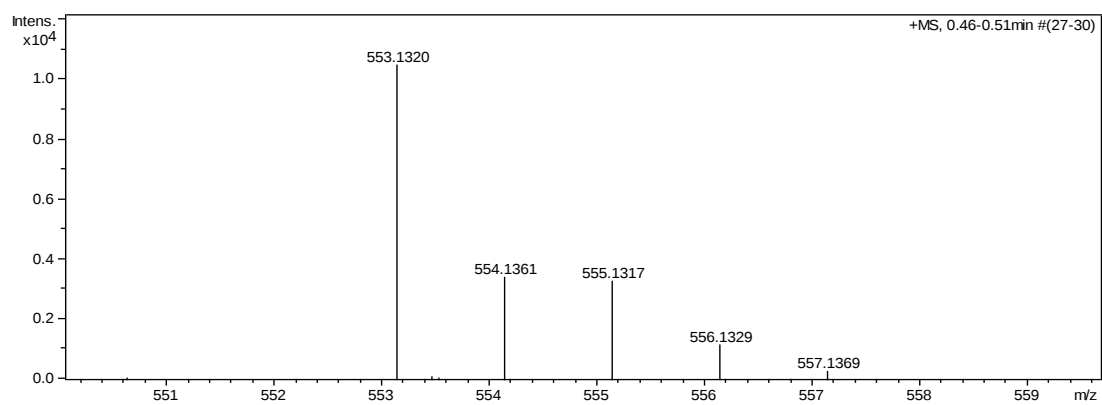
6-(4-chlorophenyl)-3'-methyl-1'-phenyl-7*H*-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5g):



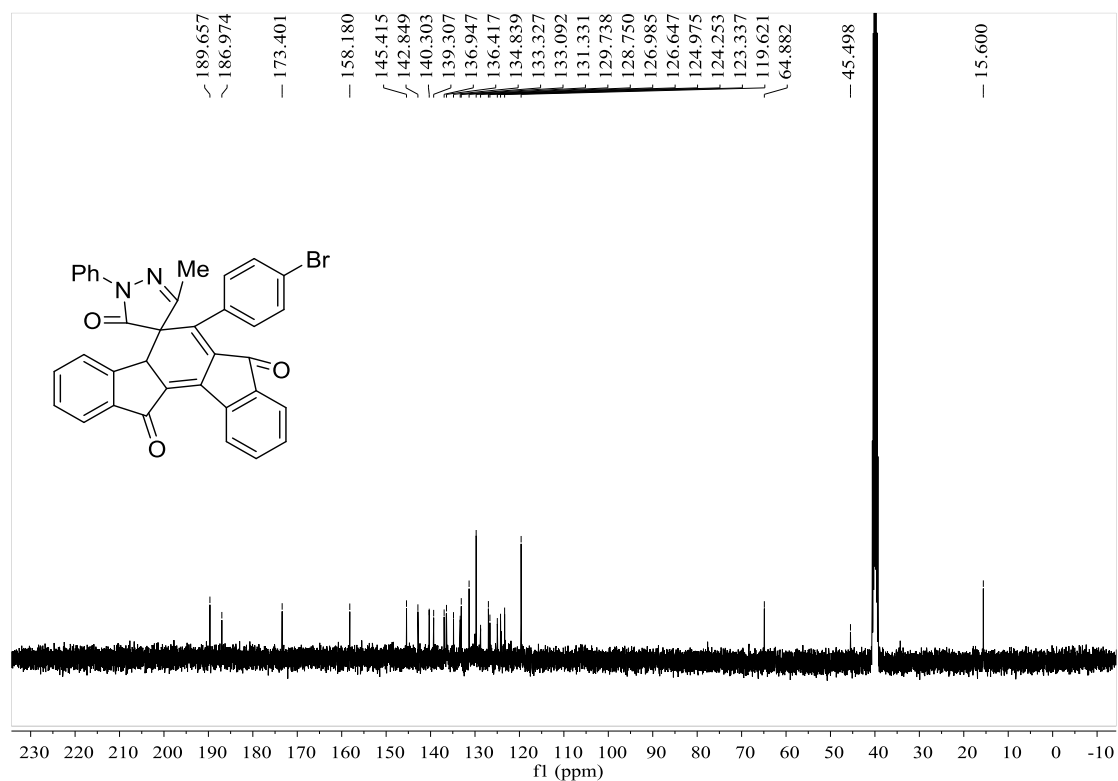
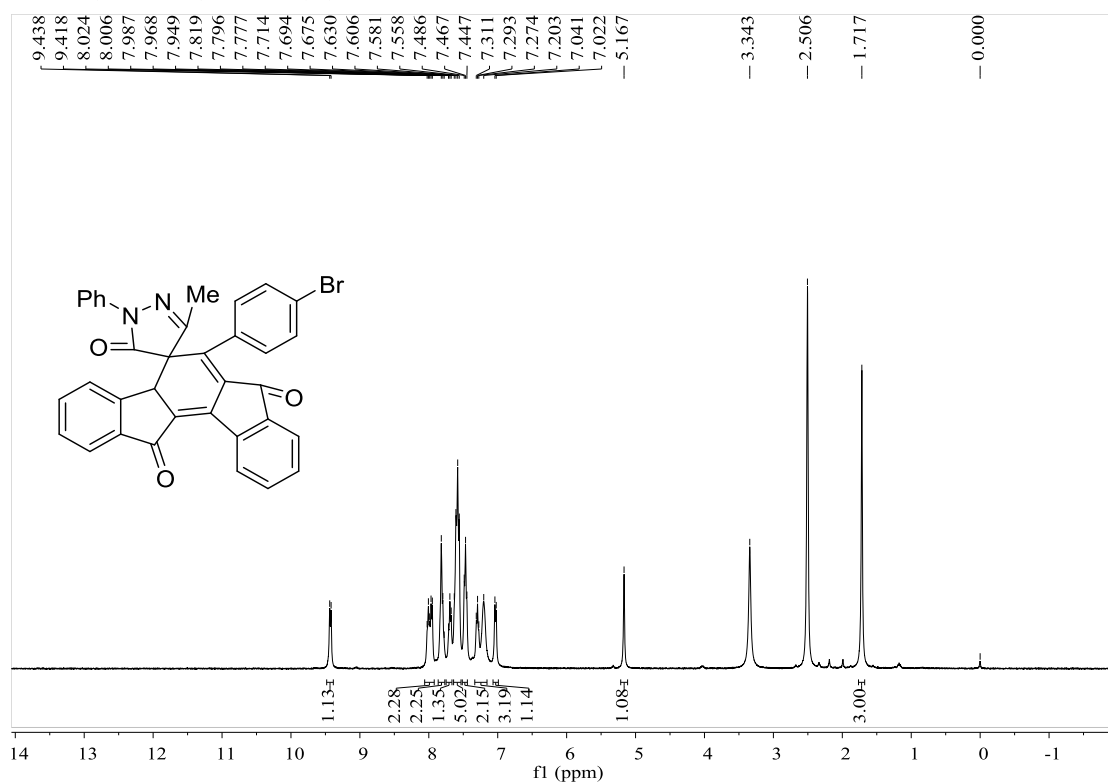


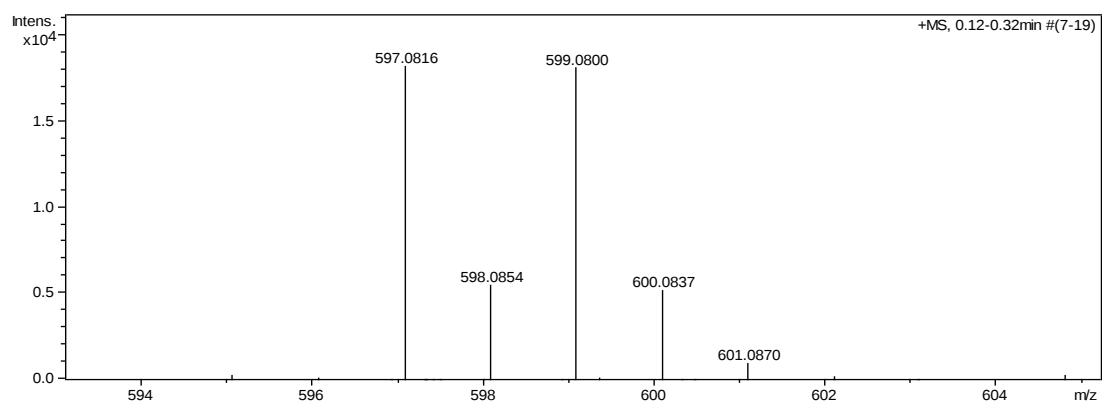
6-(3-chlorophenyl)-3'-methyl-1'-phenyl-7*H*-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5h):



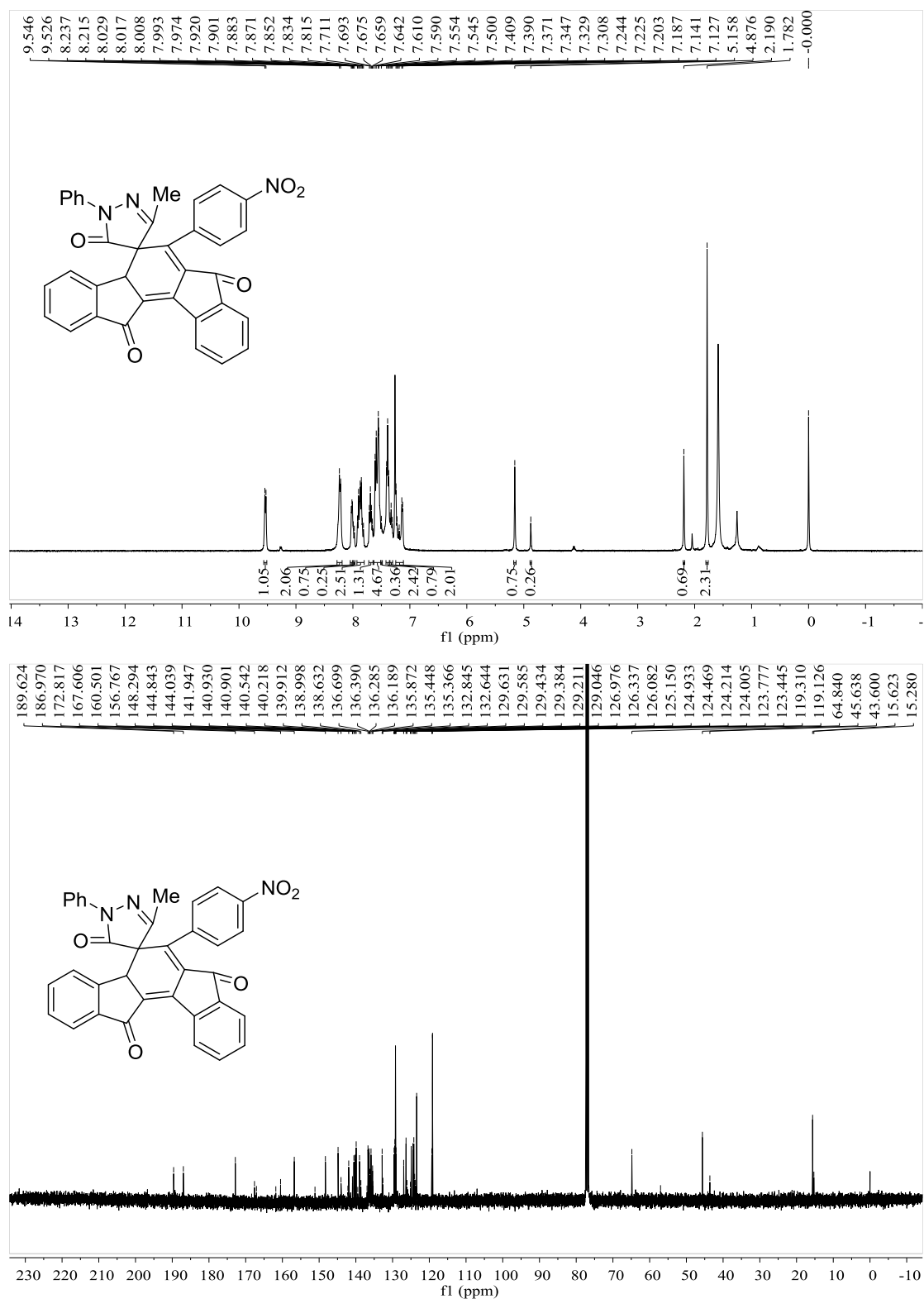


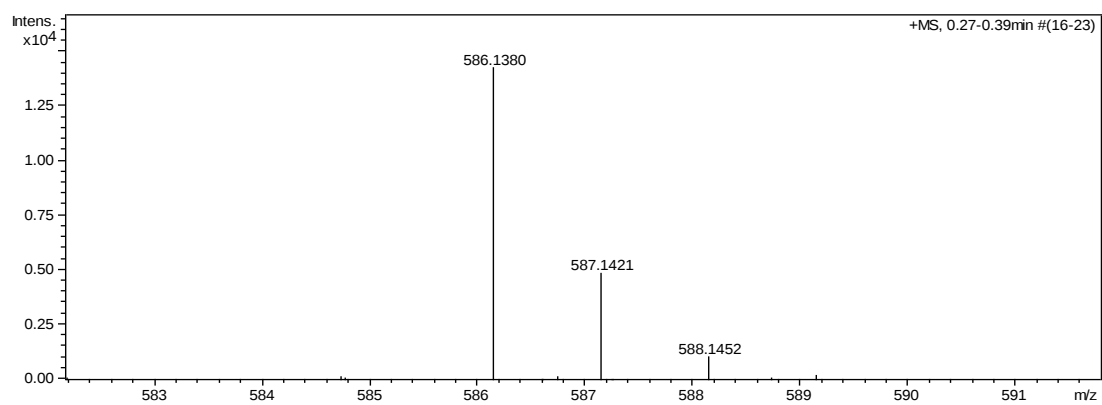
6-(4-bromophenyl)-3'-methyl-1'-phenyl-7*H*-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5i):



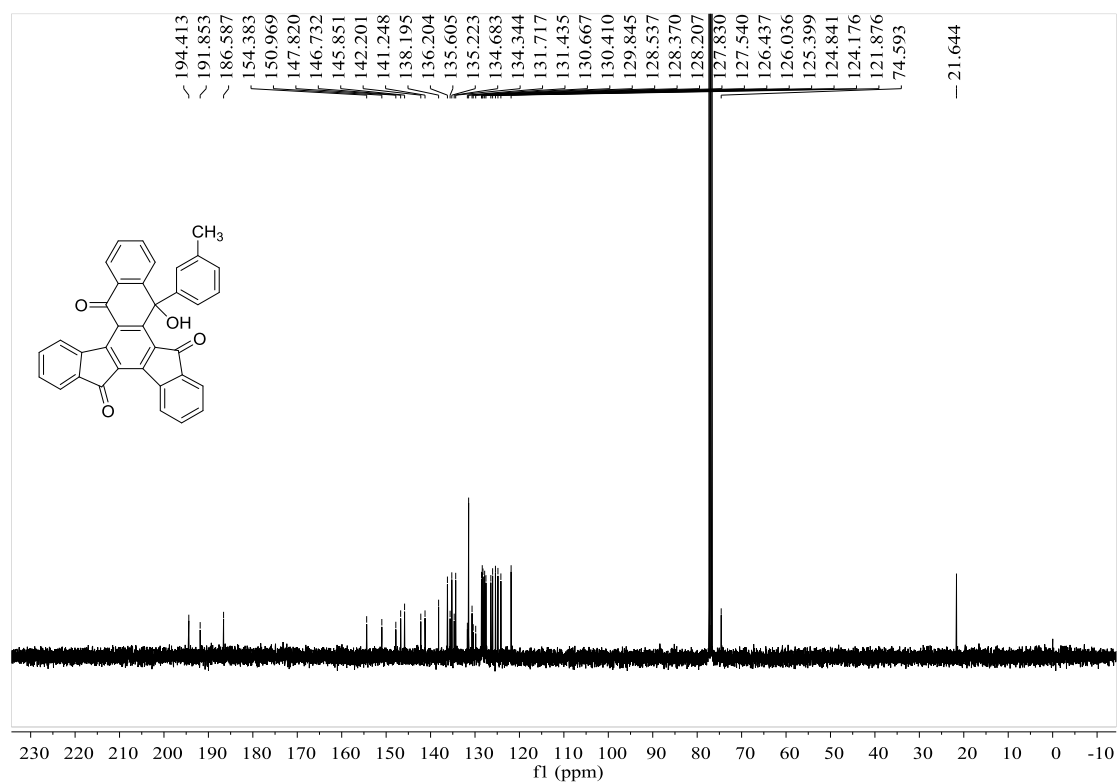
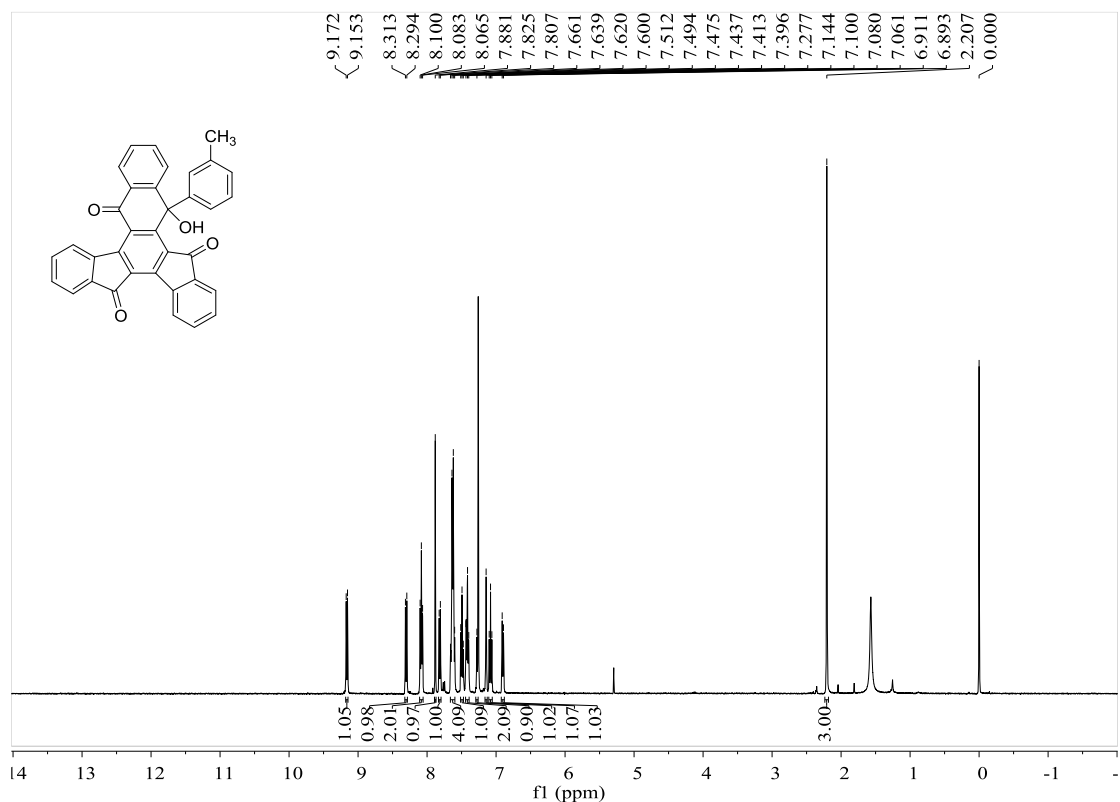


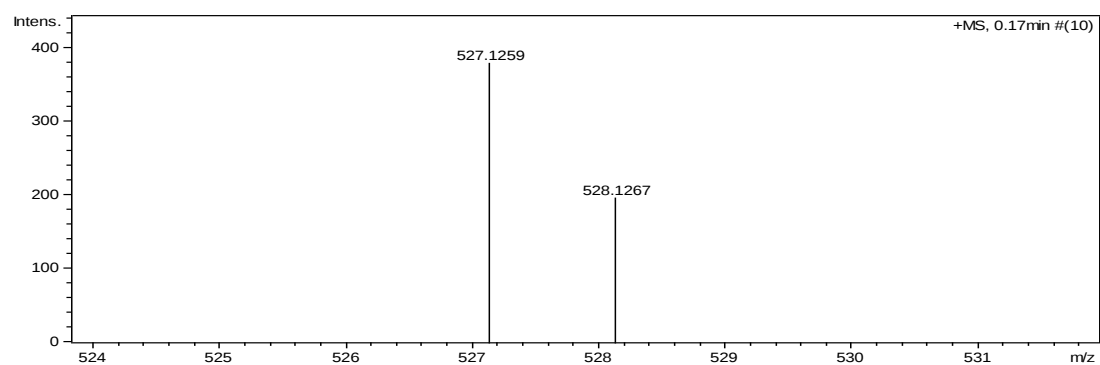
3'-methyl-6-(4-nitrophenyl)-1'-phenyl-7H-spiro[indeno[1,2-*a*]fluorene-5,4'-pyrazole]-5',7,12(1'*H*,4*bH*)-trione (5j):





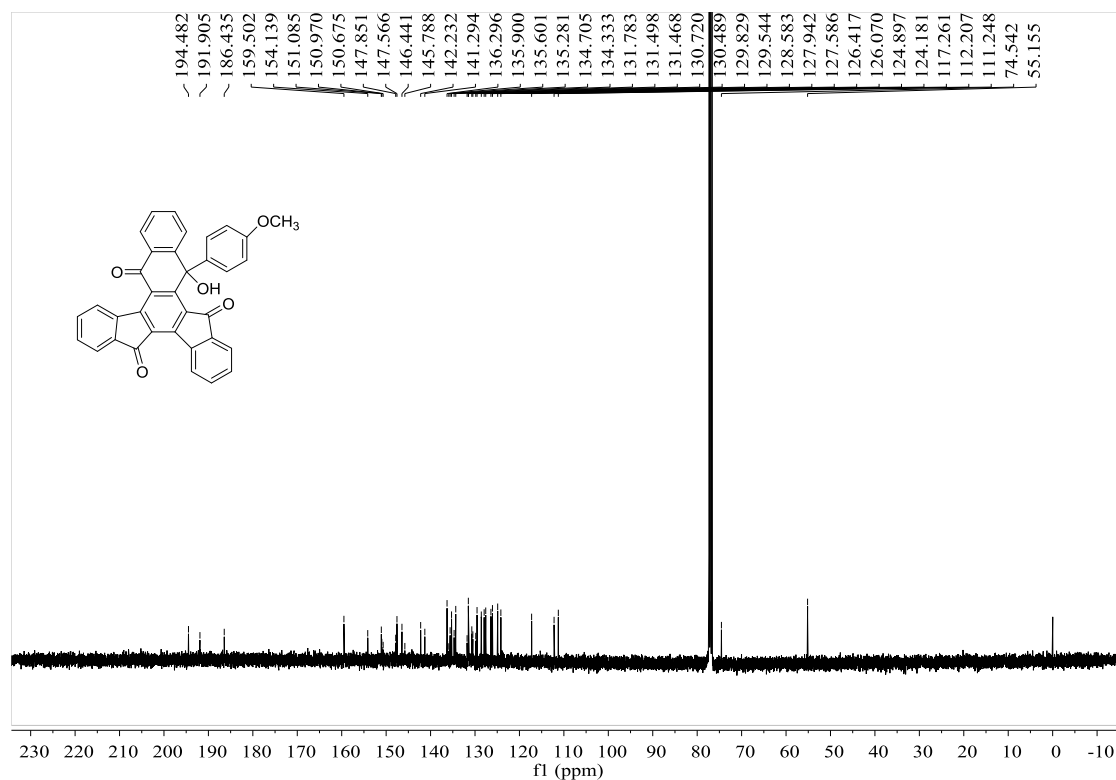
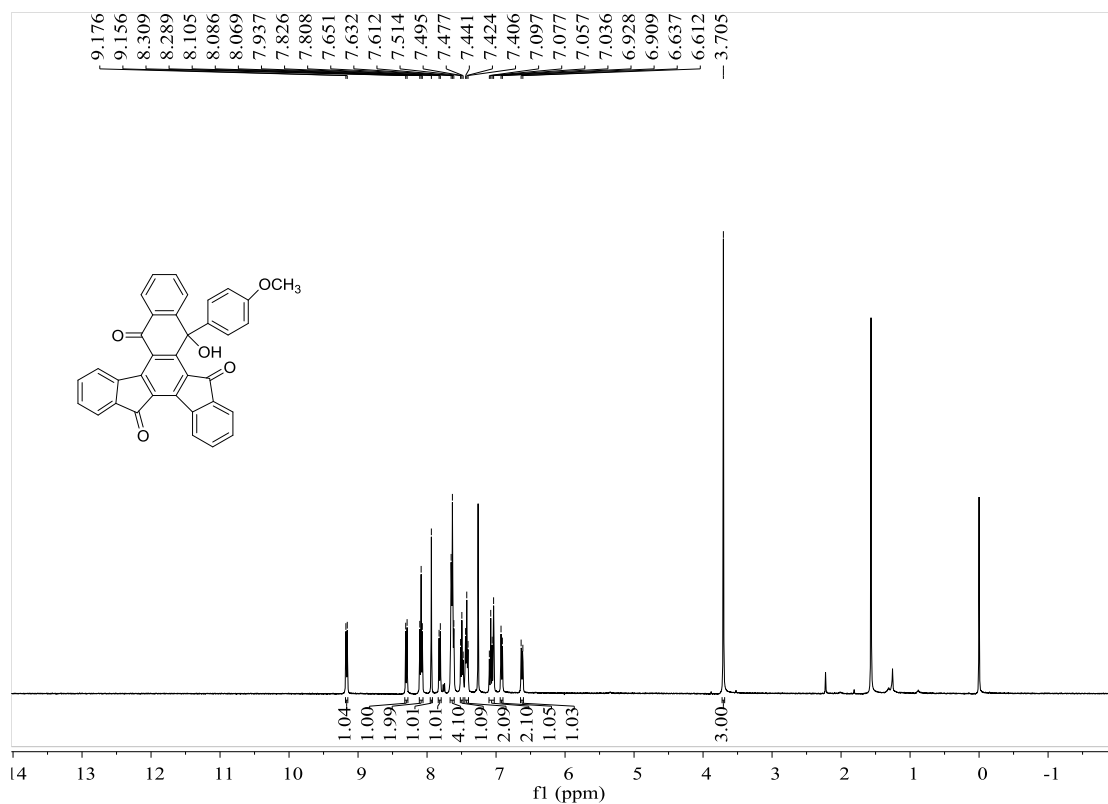
15-hydroxy-15-(*m*-tolyl)diindeno[1,2-*a*:1',2'-*c*]anthracene-5,10,16(15*H*)-trione[7a] :

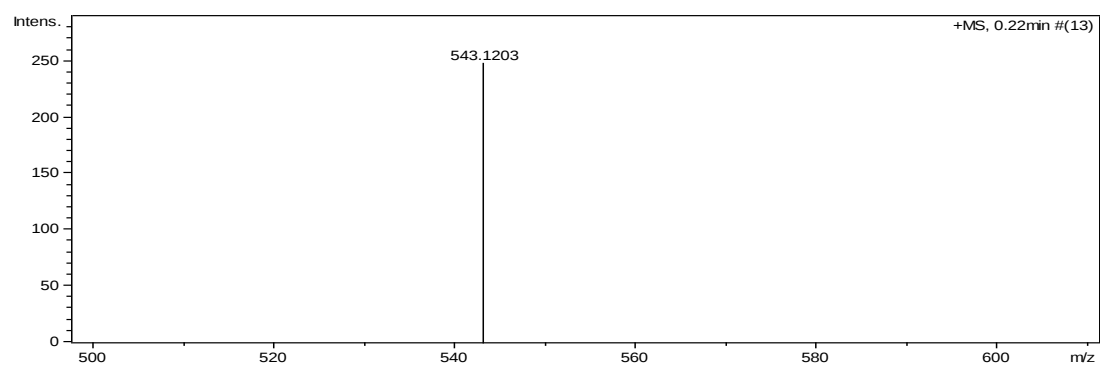




15-hydroxy-15-(4-methoxyphenyl)diindeno[1,2-*a*:1',2'-*c*]anthracene-5,10,16(15*H*)-trione

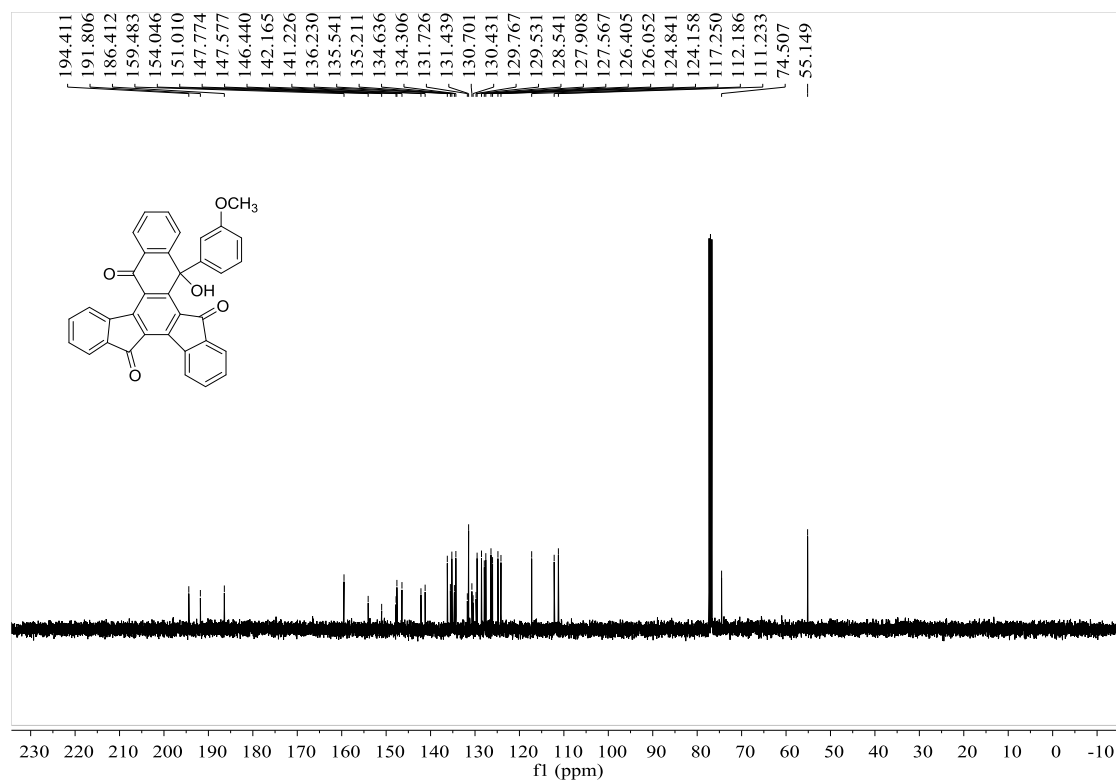
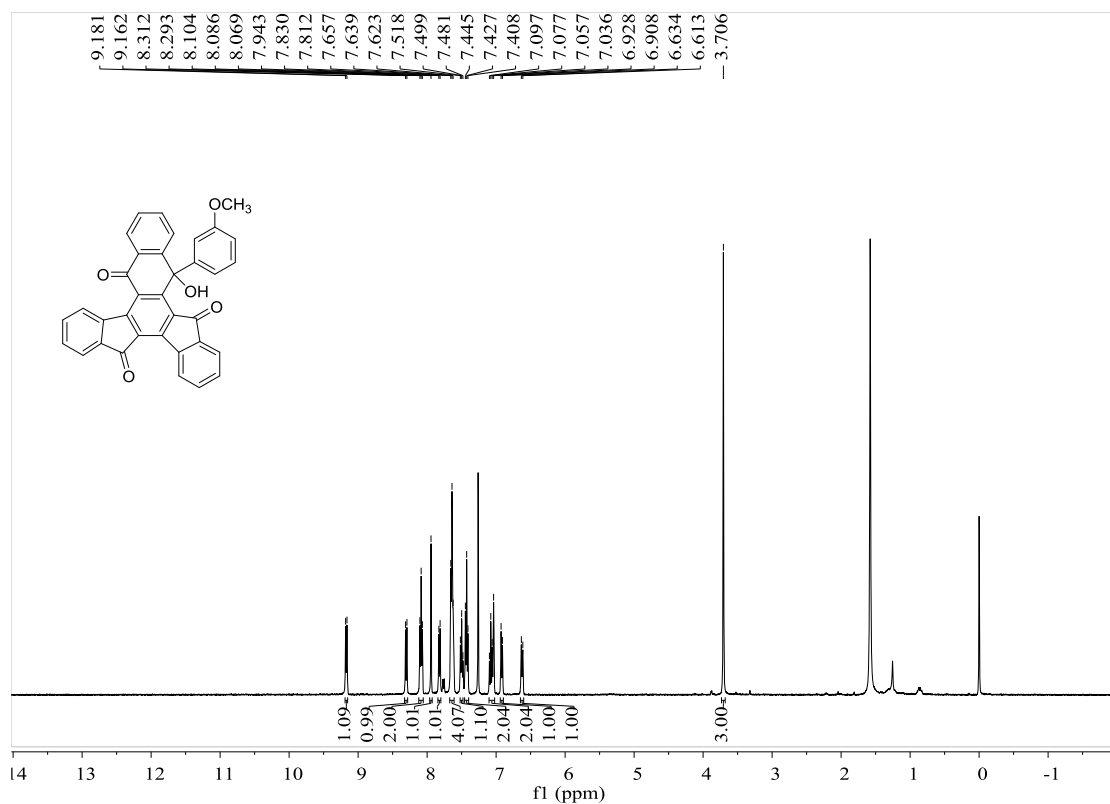
(7b) :

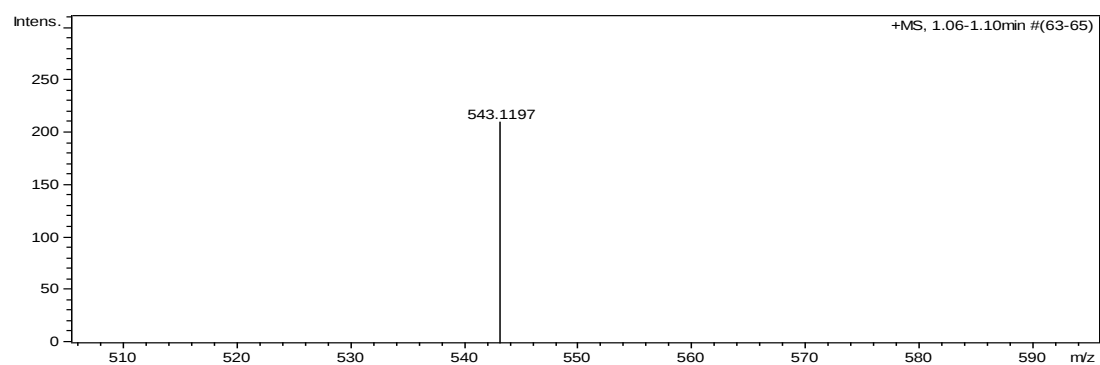




15-hydroxy-15-(3-methoxyphenyl)diindeno[1,2-*a*:1',2'-*c*]anthracene-5,10,16(15*H*)-trione

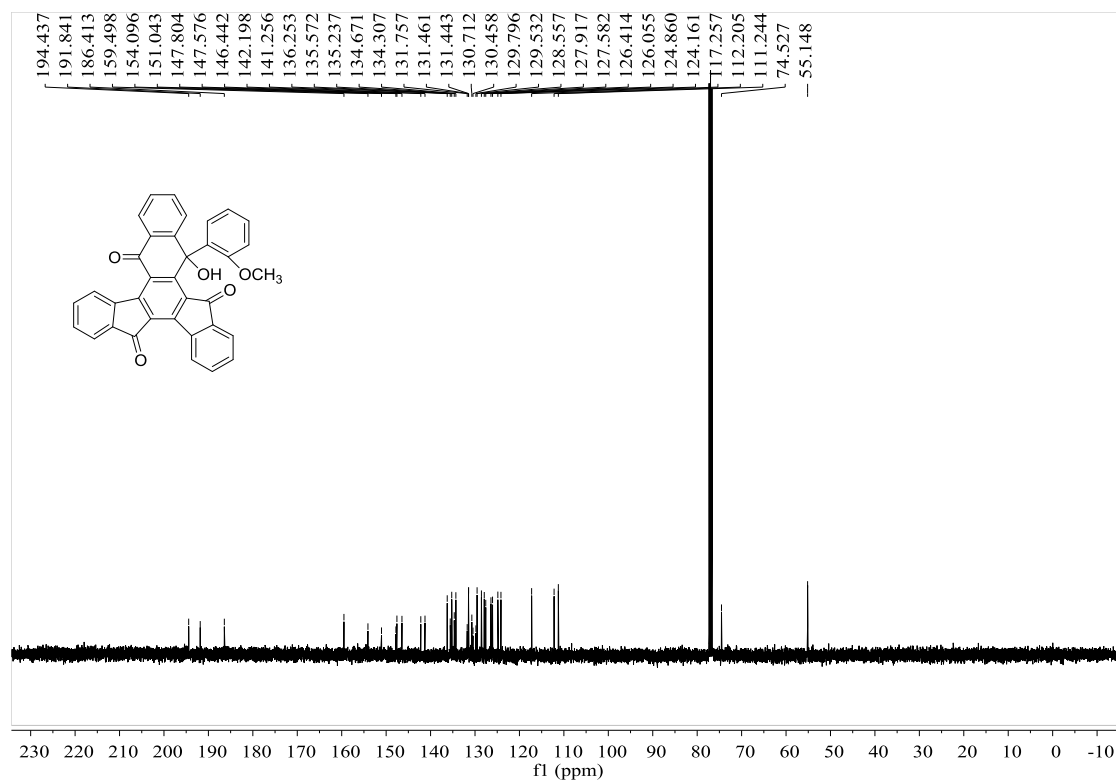
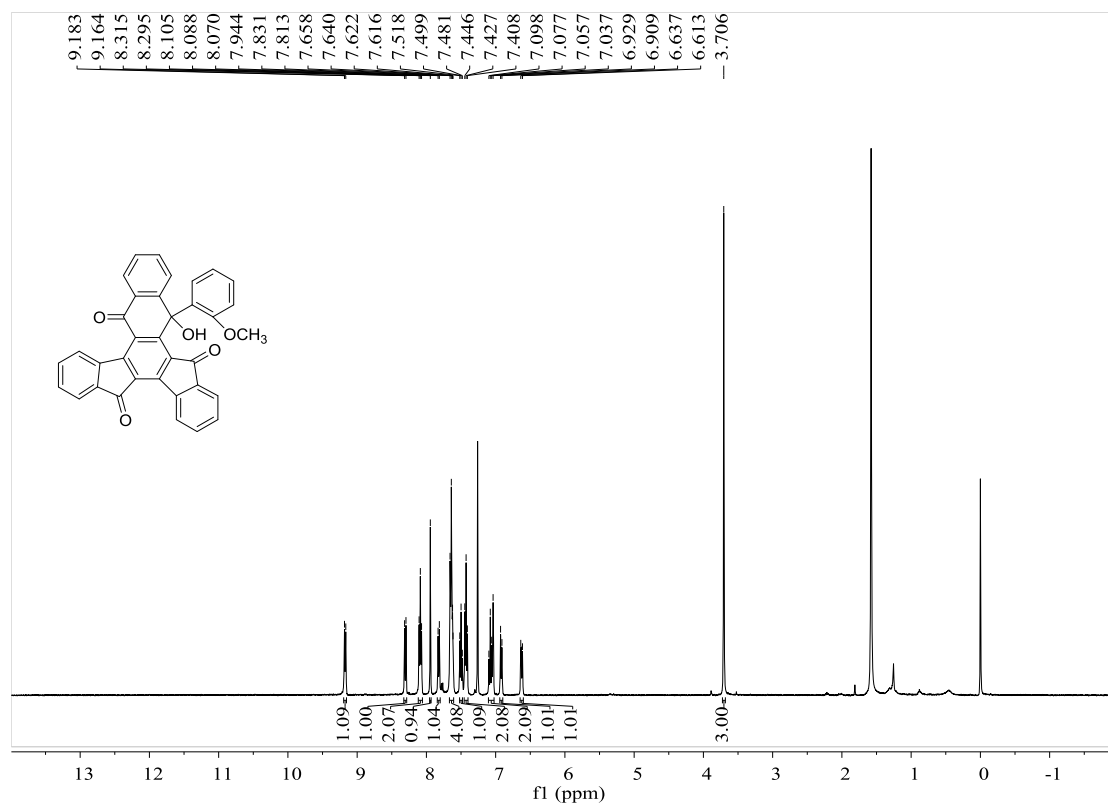
(7c):

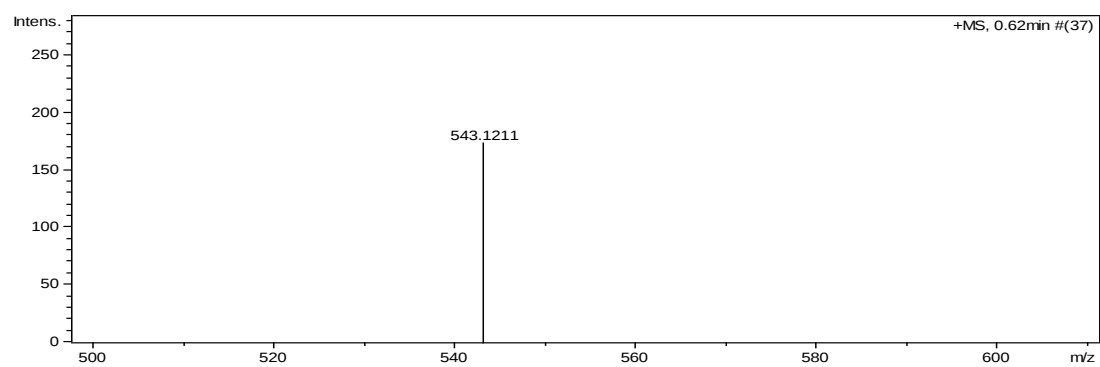




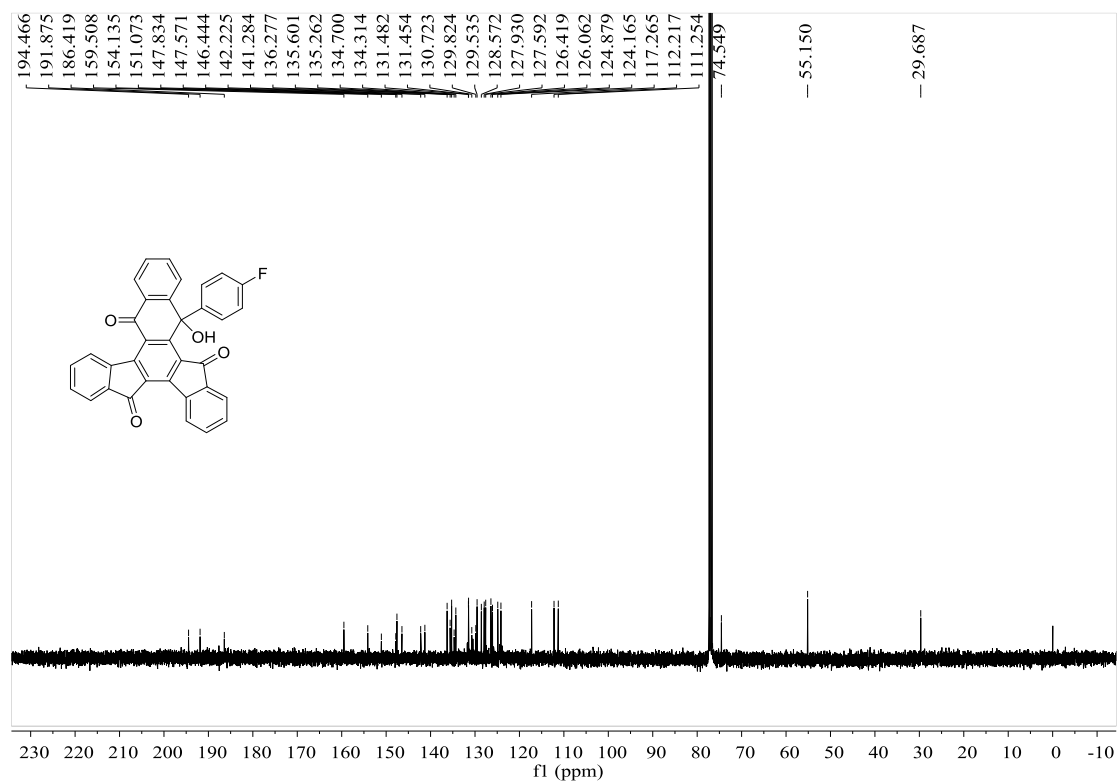
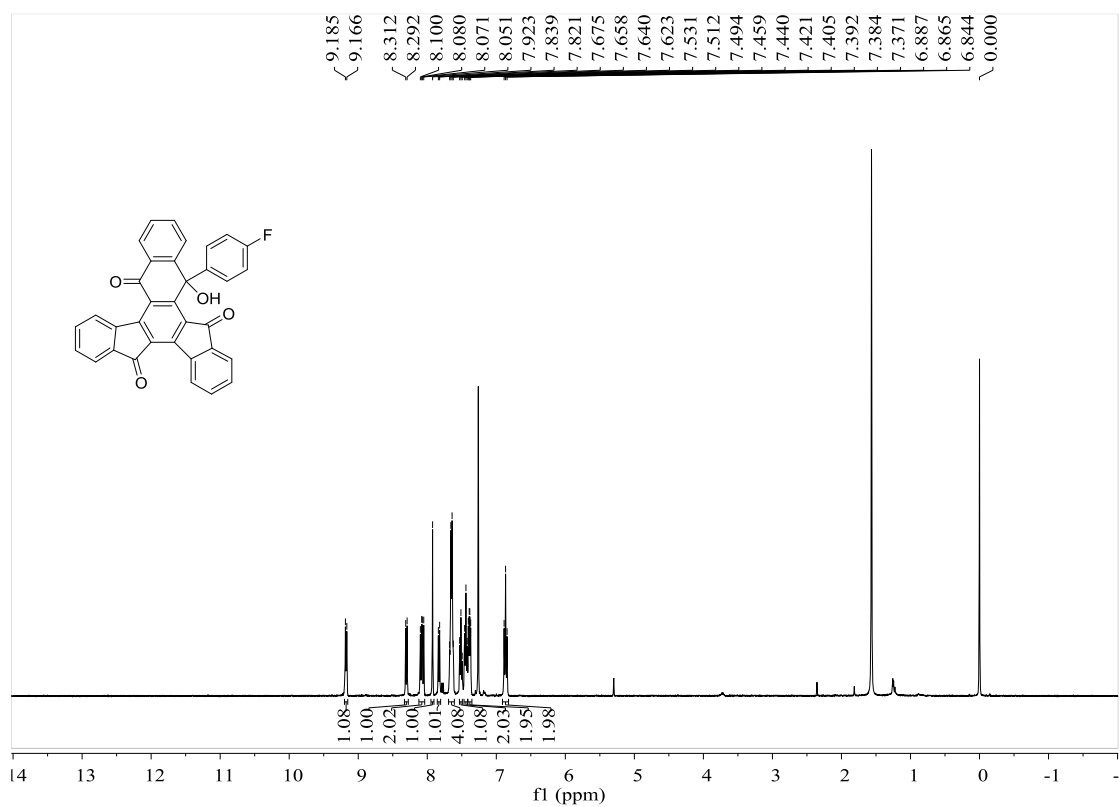
15-hydroxy-15-(2-methoxyphenyl)diindeno[1,2-*a*:1',2'-*c*]anthracene-5,10,16(15*H*)-trione

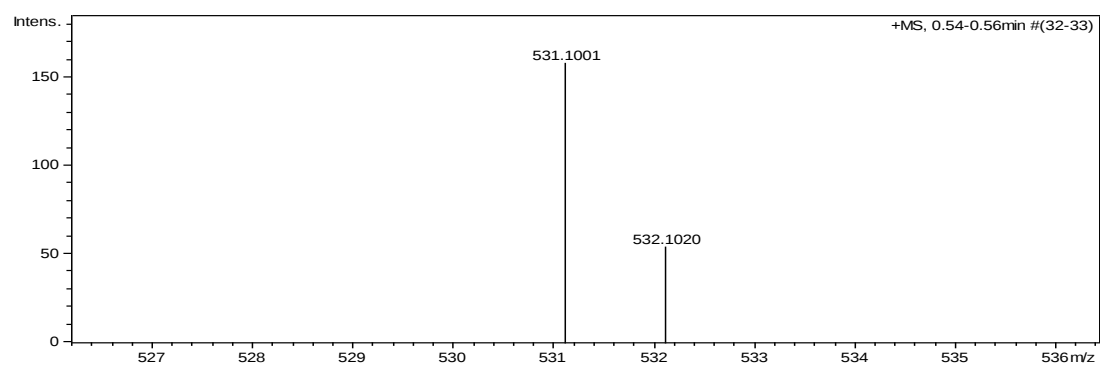
(7d):



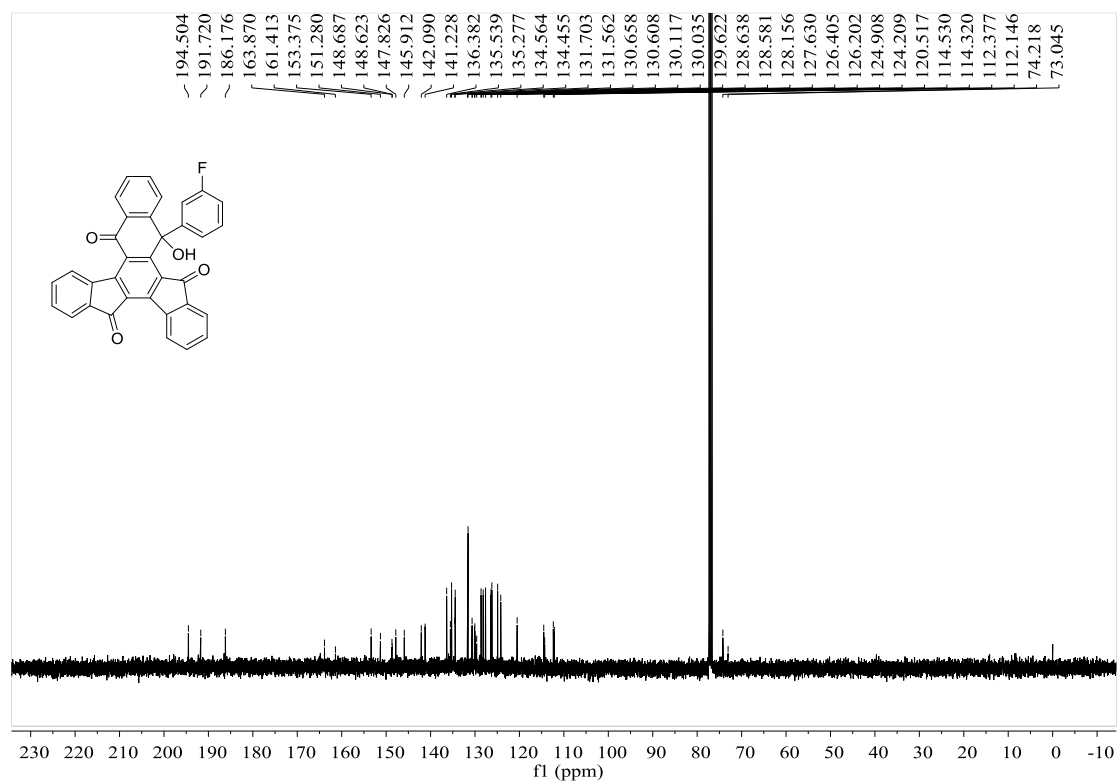
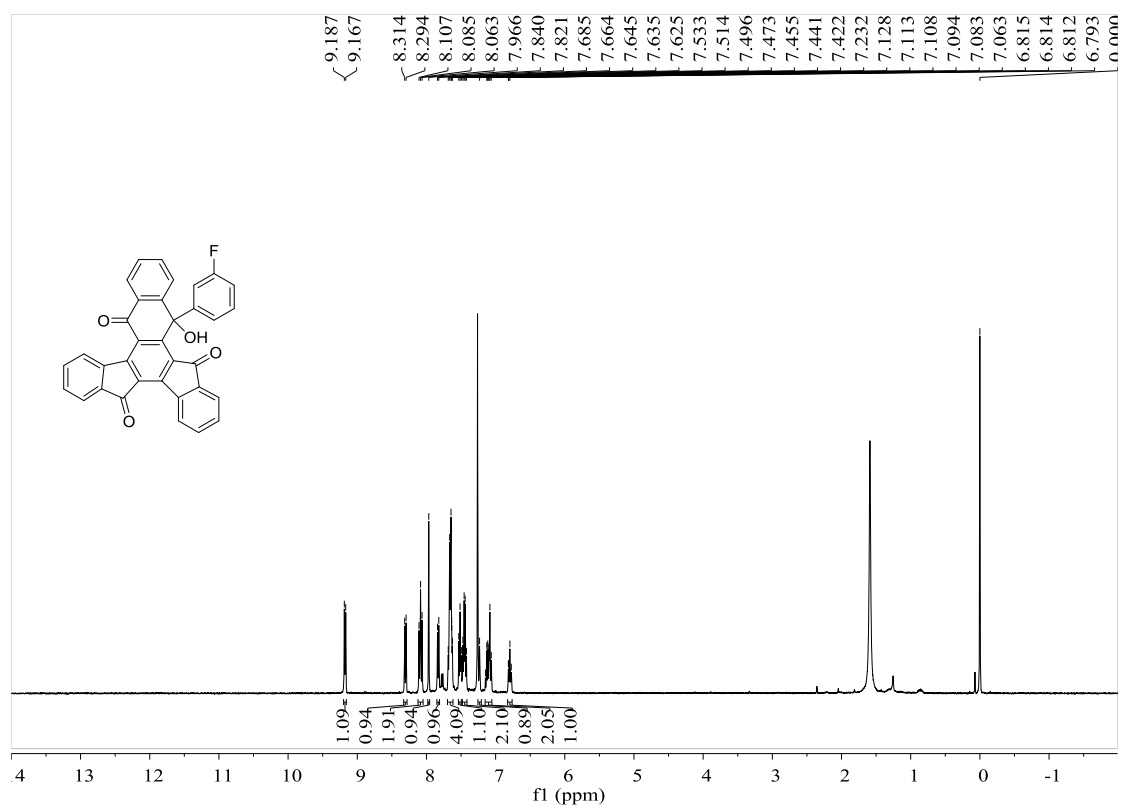


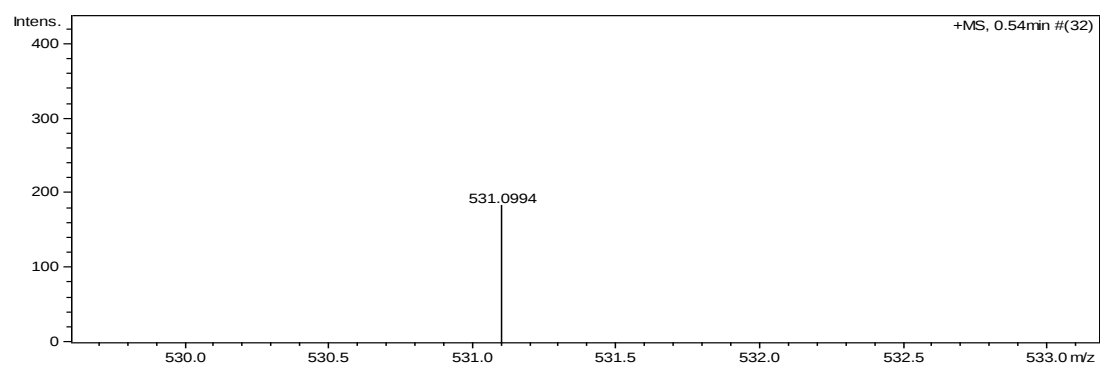
15-(4-fluorophenyl)-15-hydroxydiindeno[1,2-*a*:1',2'-*c*]anthracene-5,10,16(15*H*)-trione (7e):





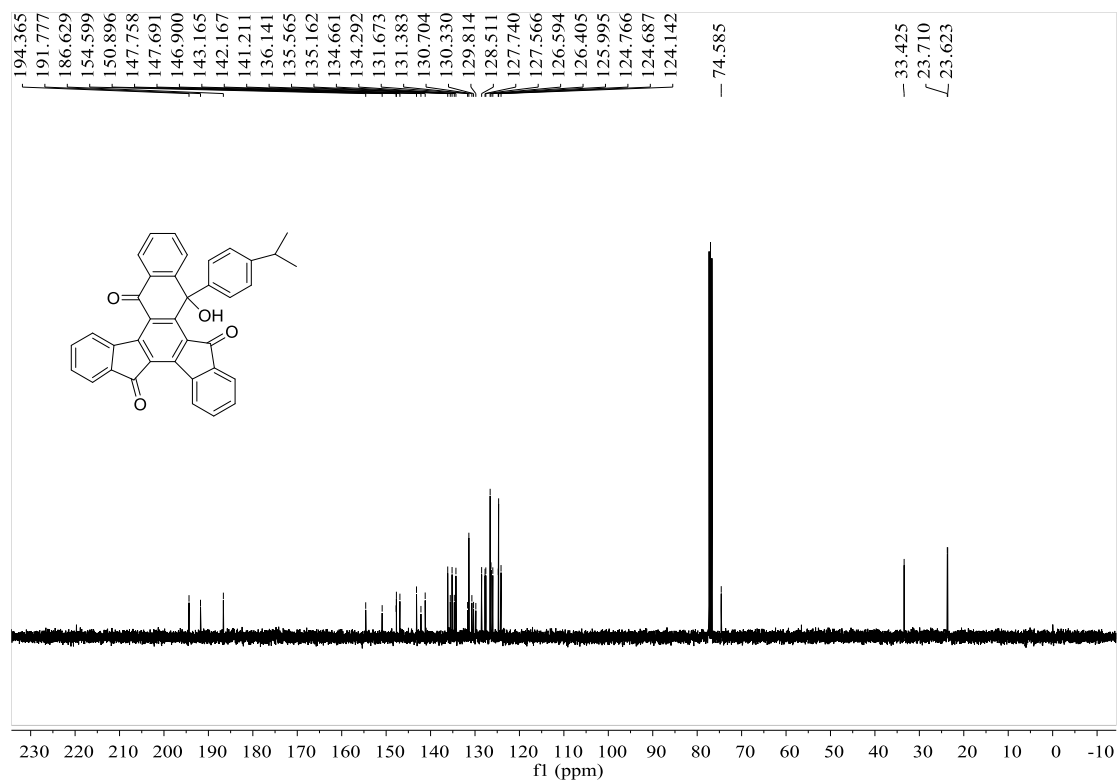
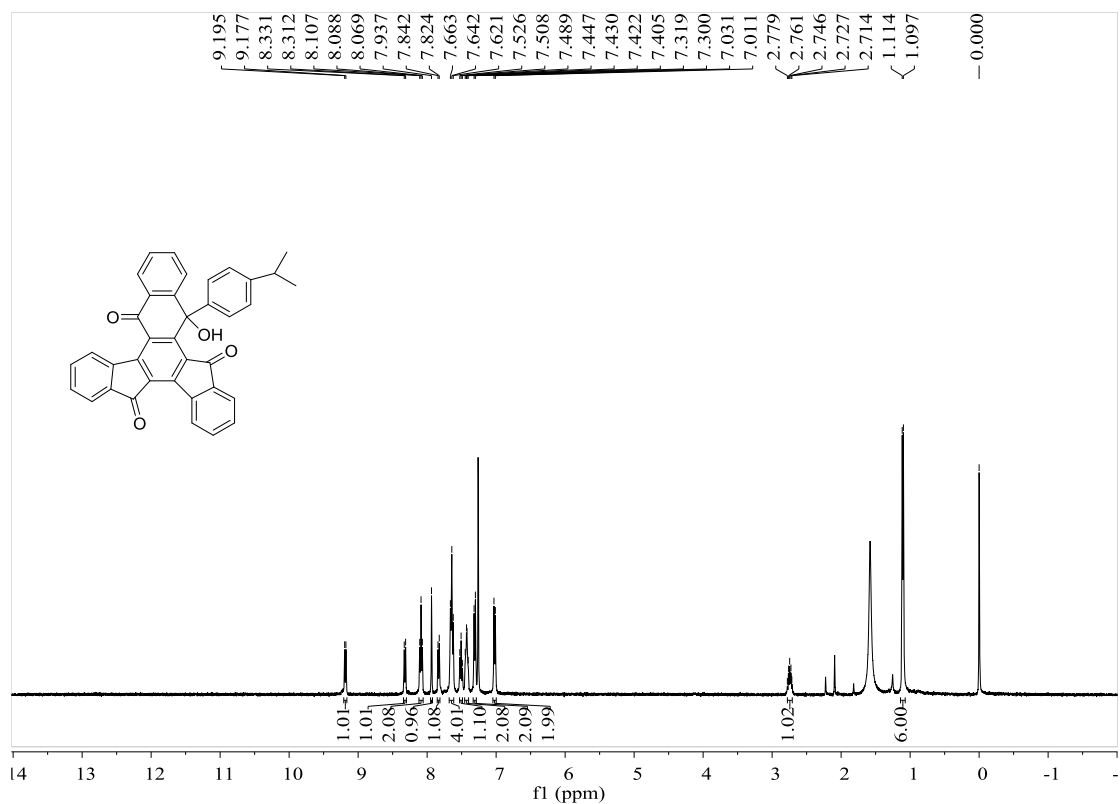
15-(3-fluorophenyl)-15-hydroxydiindeno[1,2-*a*:1',2'-*c*]anthracene-5,10,16(15*H*)-trione (7f):

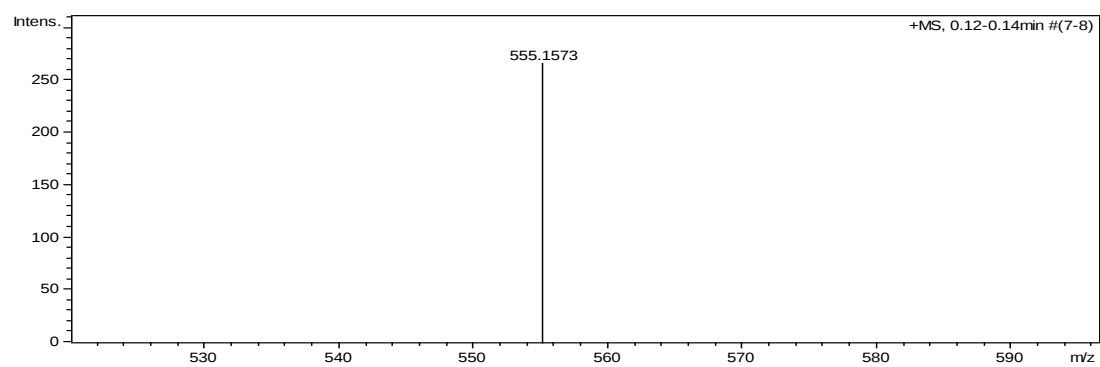




15-hydroxy-15-(4-isopropylphenyl)diindeno[1,2-*a*:1',2'-*c*]anthracene-5,10,16(15*H*)-trione

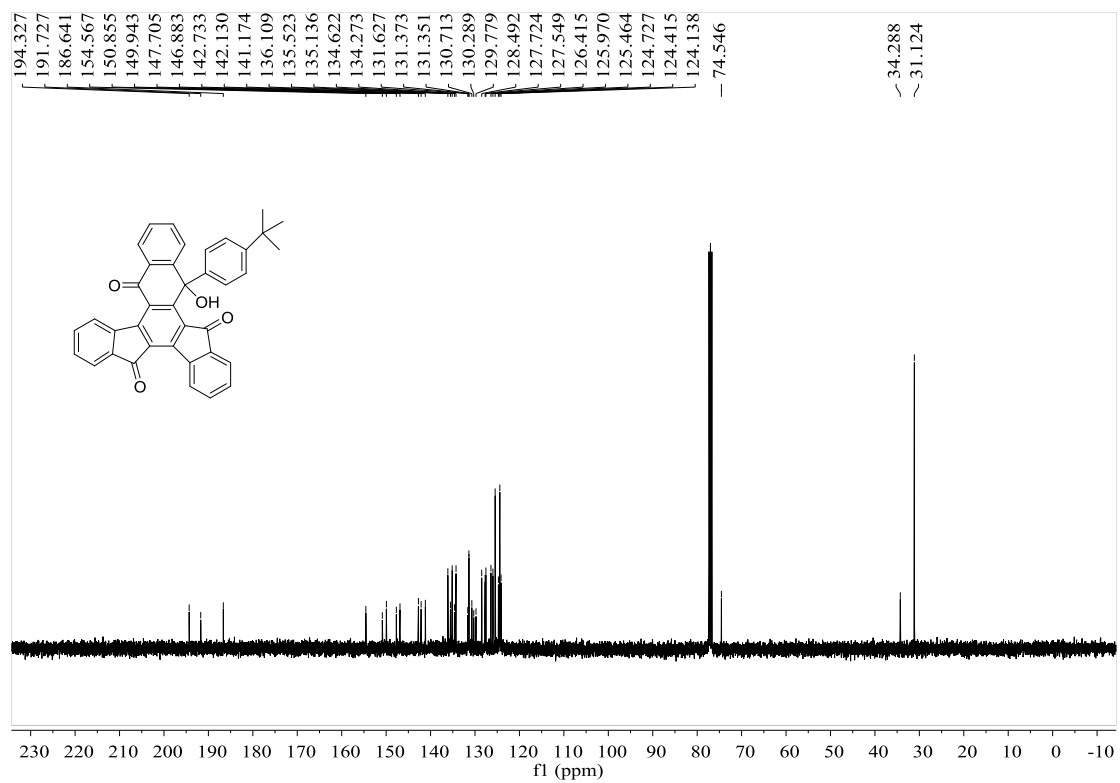
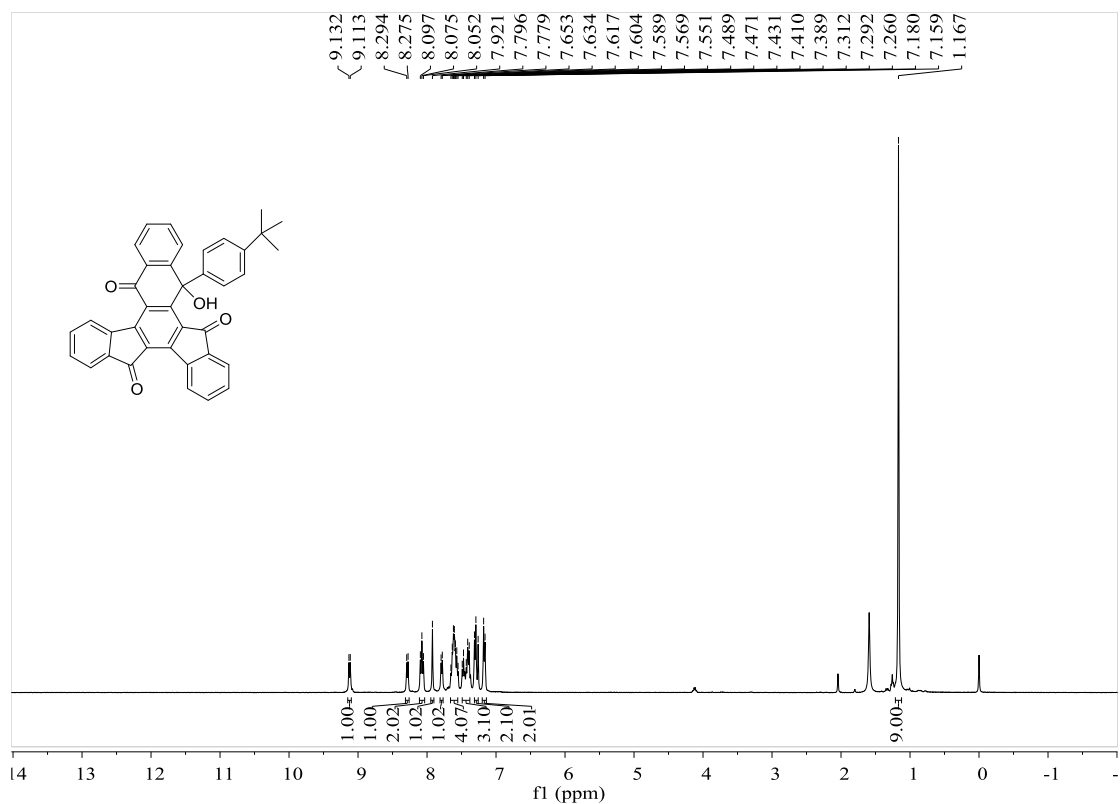
(7g):

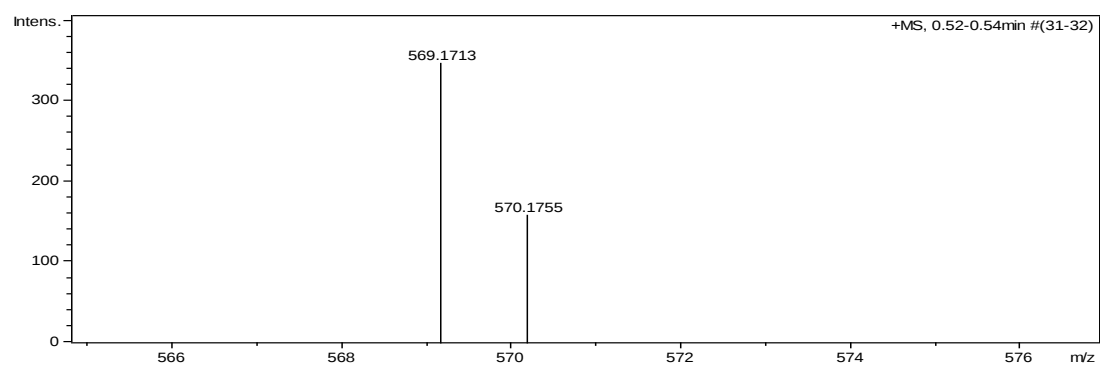




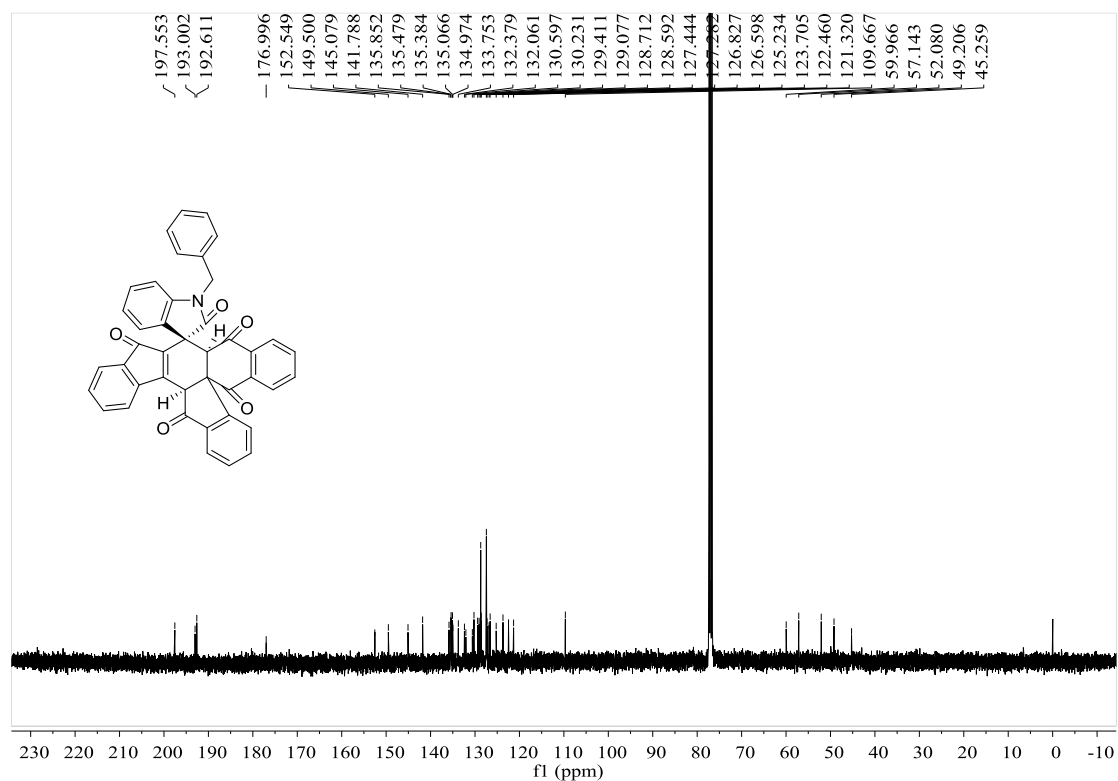
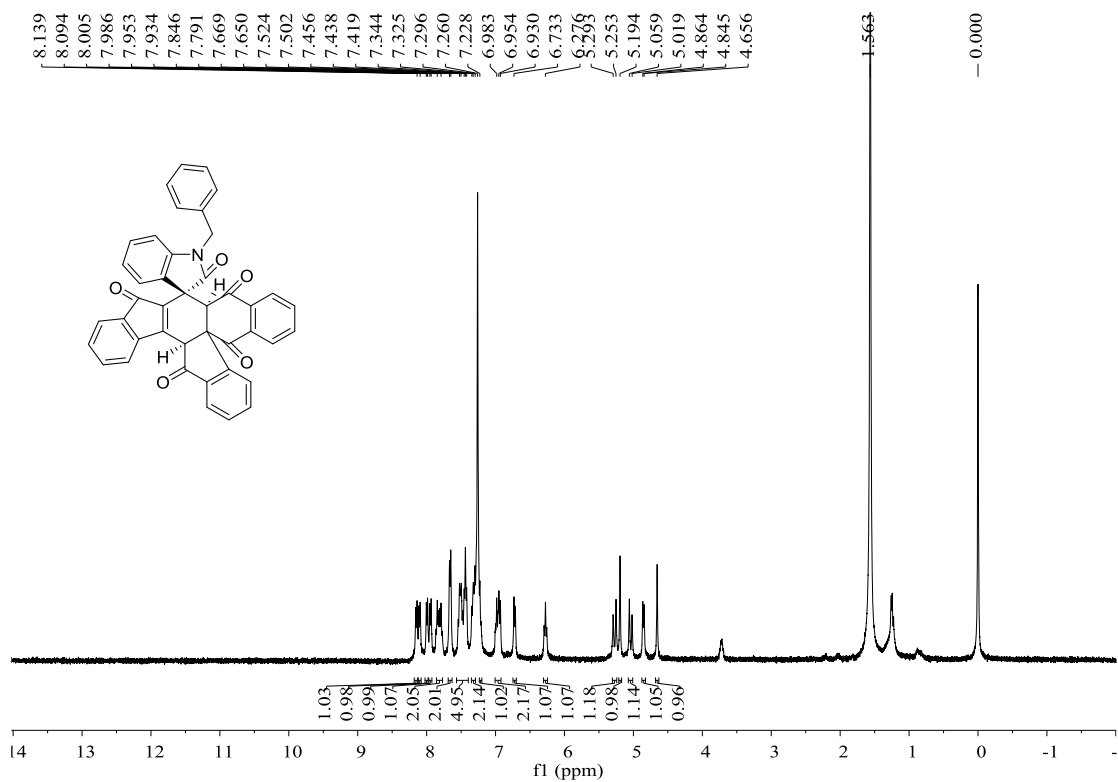
15-(4-(tert-butyl)phenyl)-15-hydroxydiindeno[1,2-*a*:1',2'-*c*]anthracene-5,10,16(15*H*)-trione

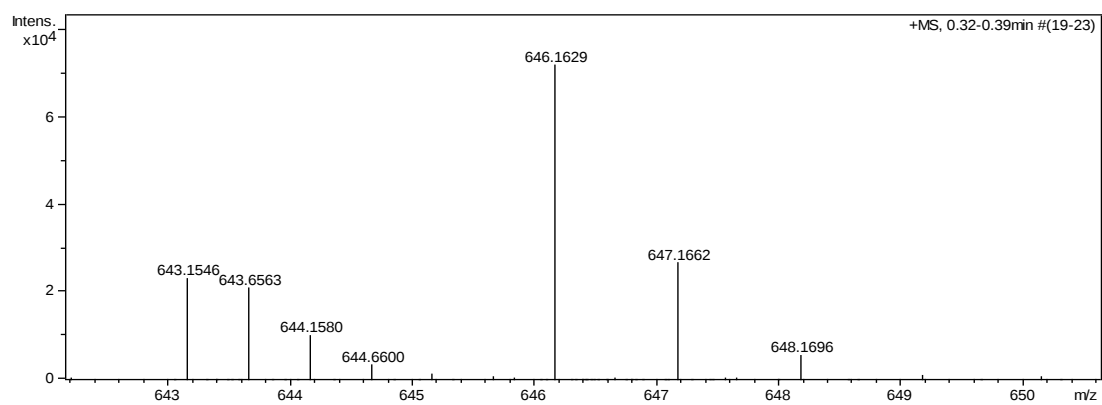
(7h):



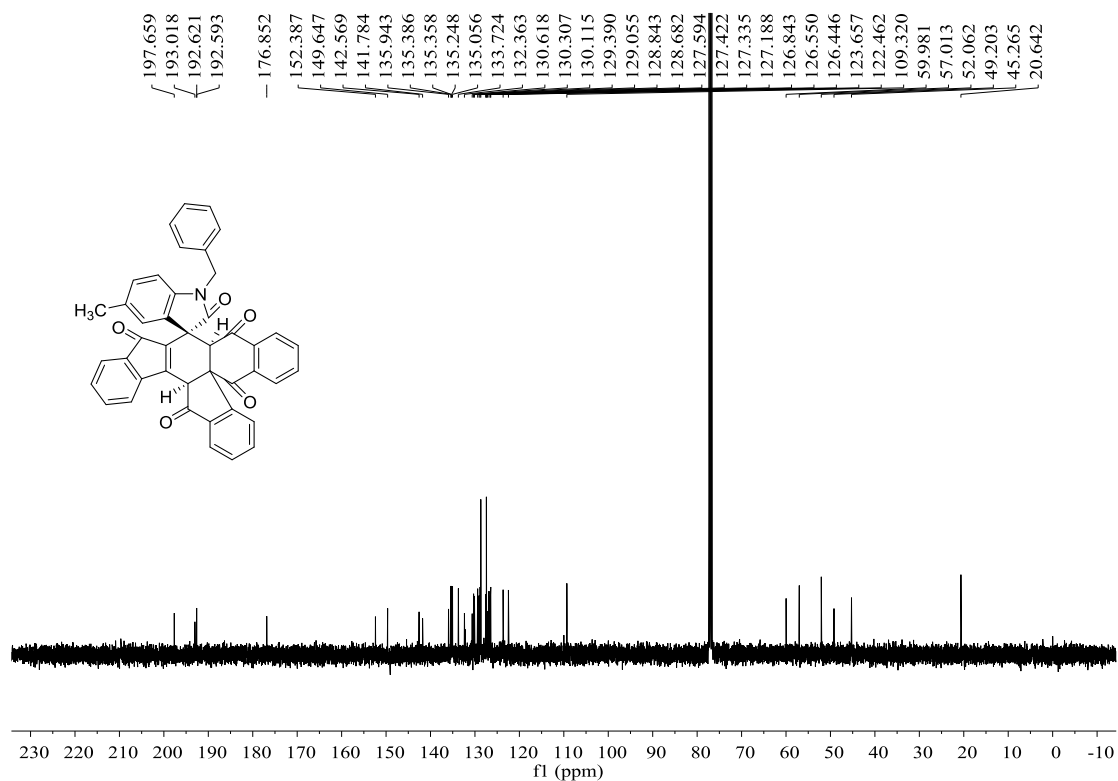
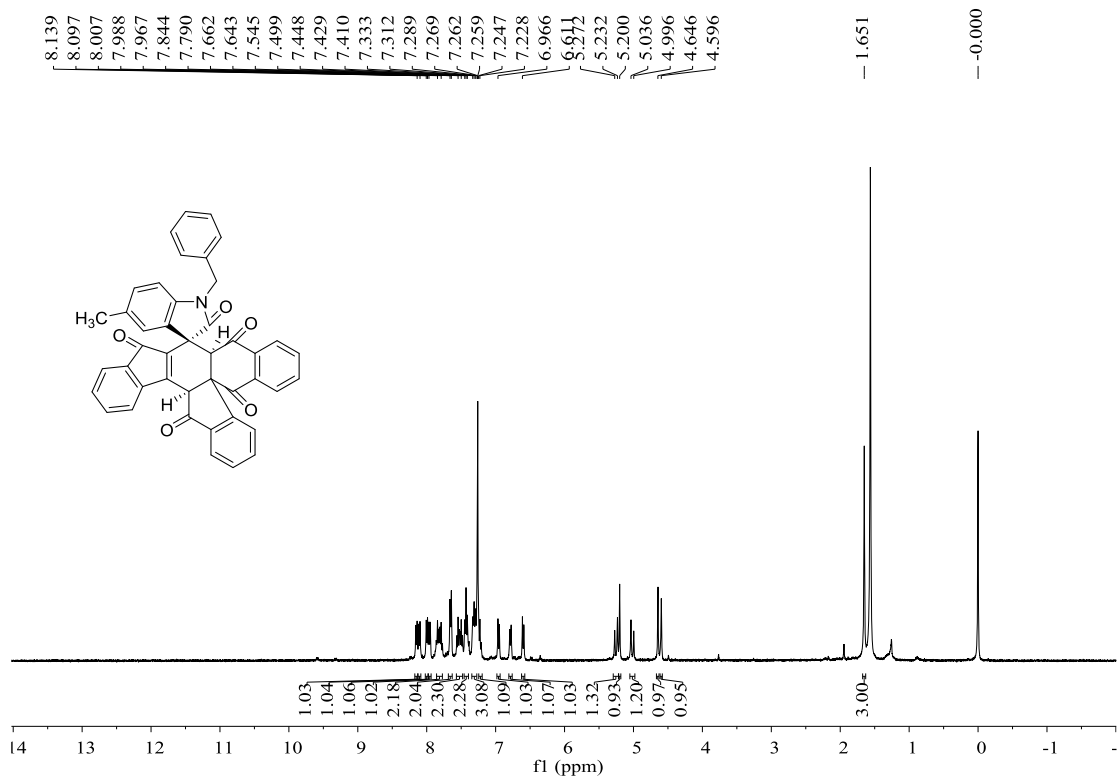


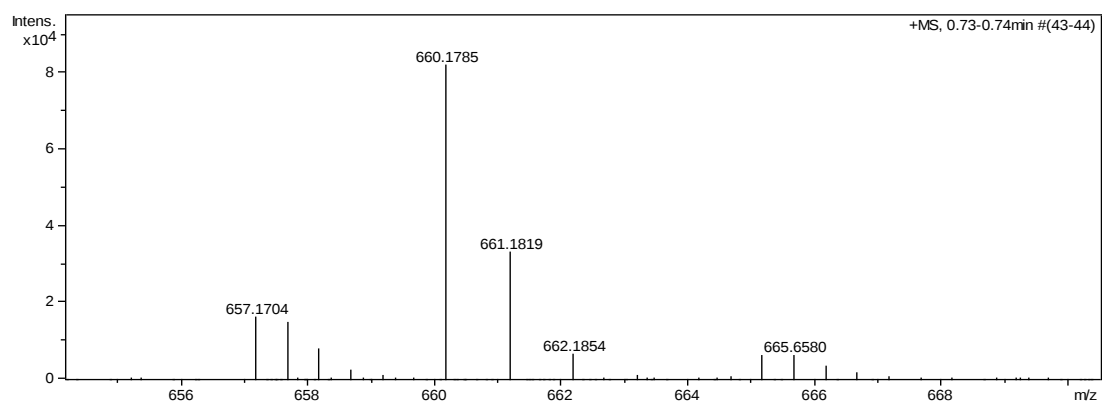
1'-benzyl-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8a):



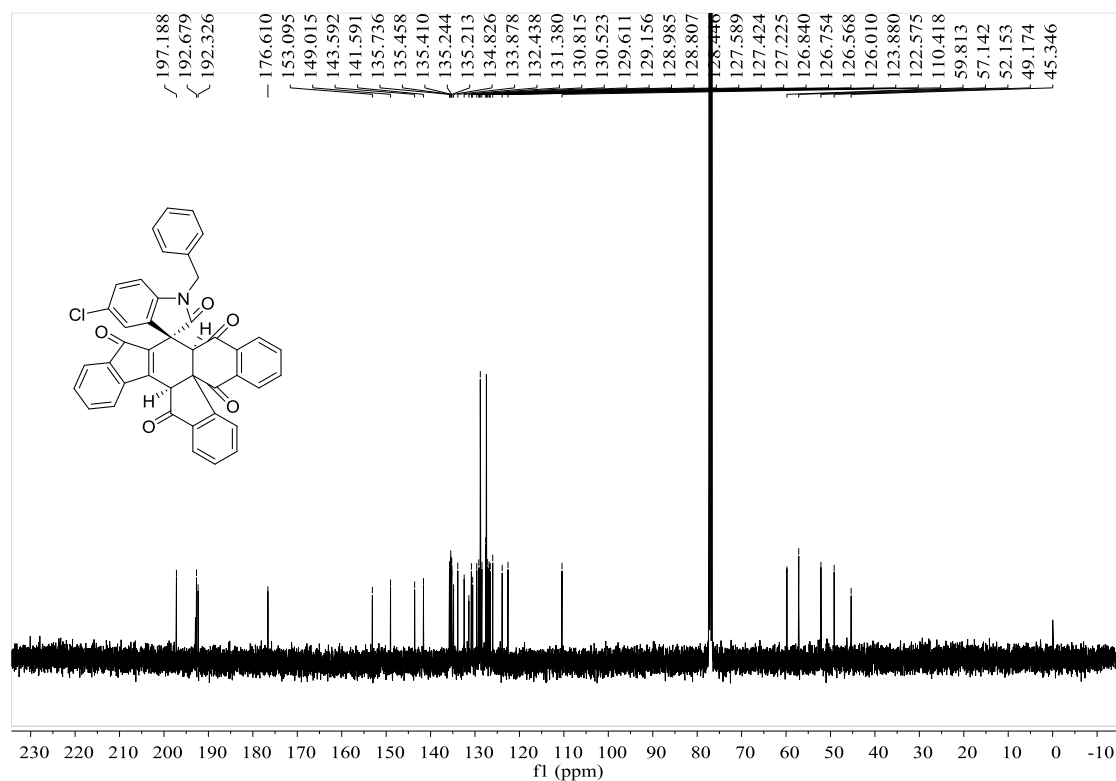
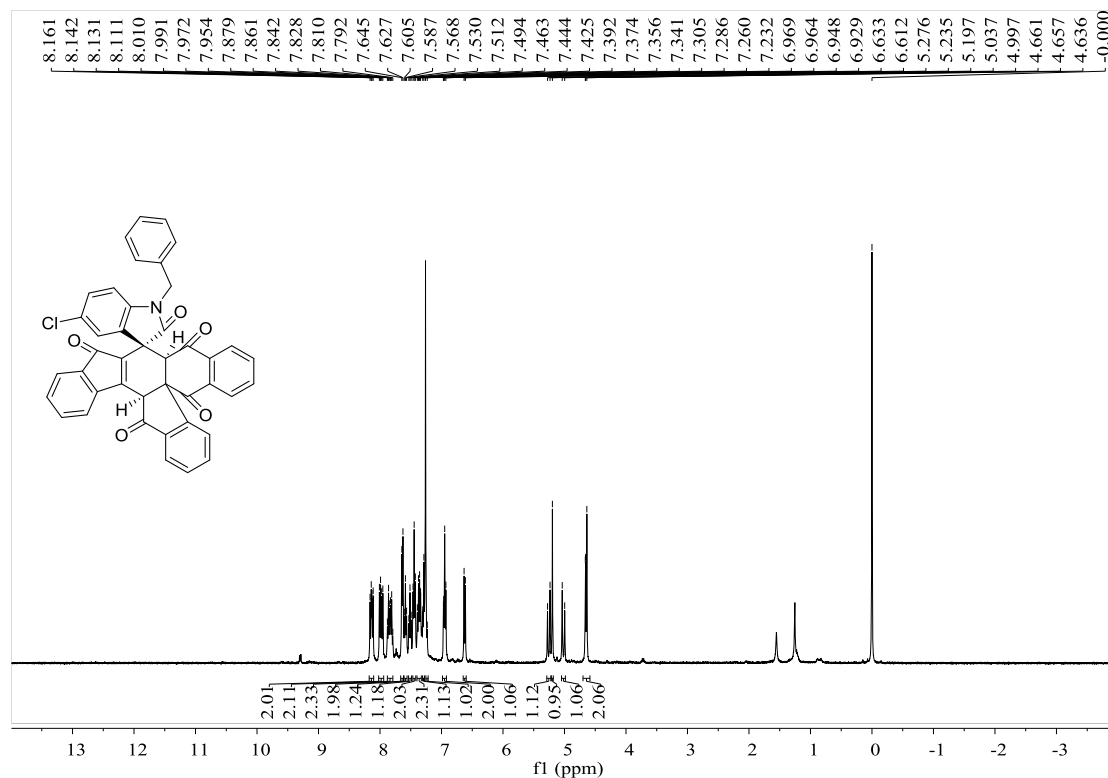


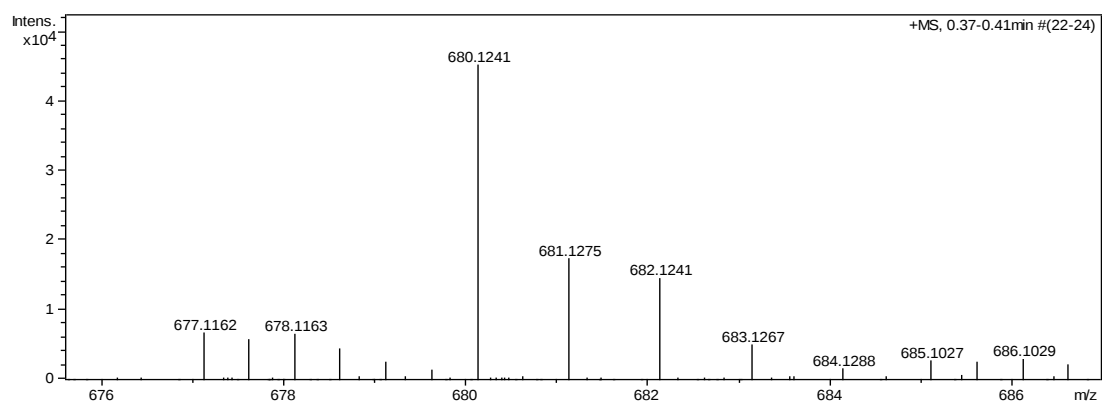
1'-benzyl-5'-methyl-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8b):



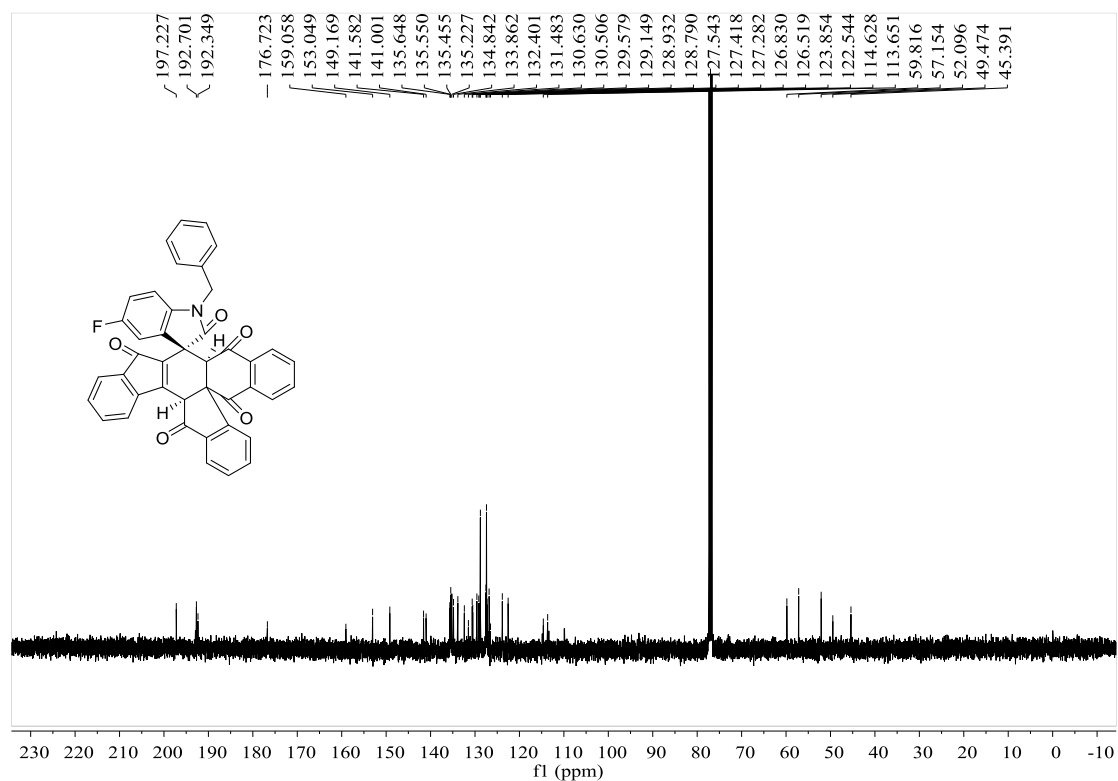
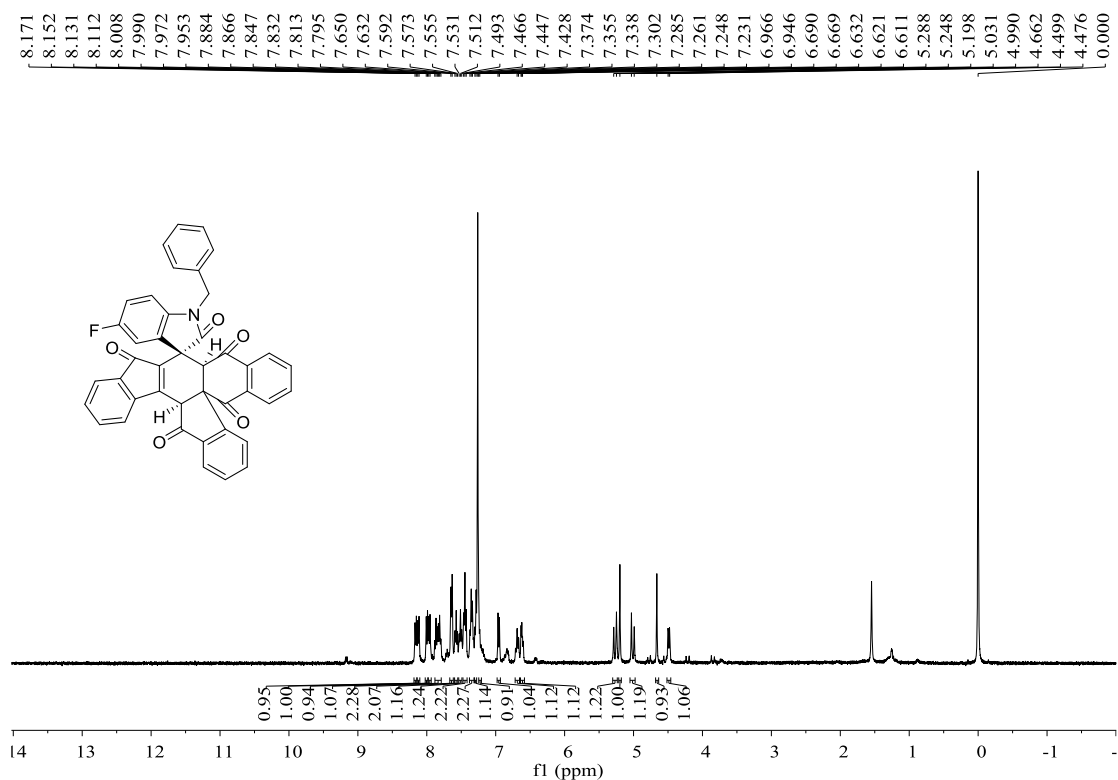


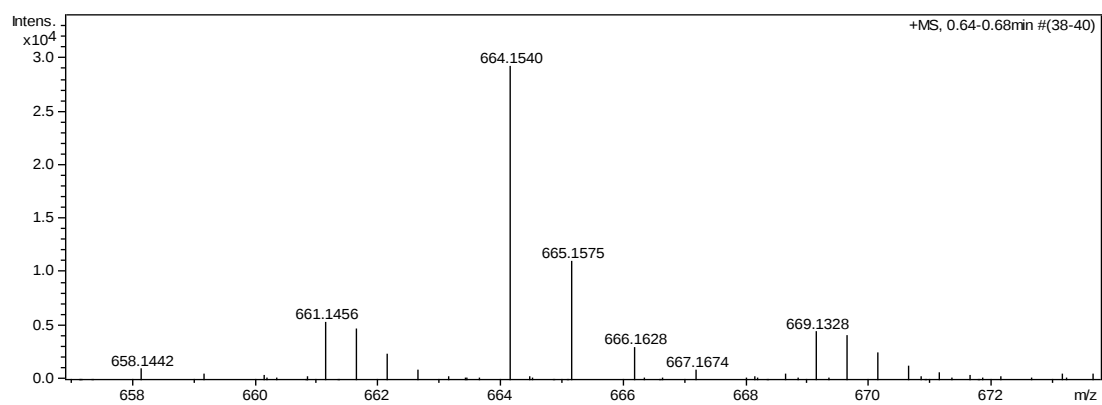
1'-benzyl-5'-chloro-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8c):



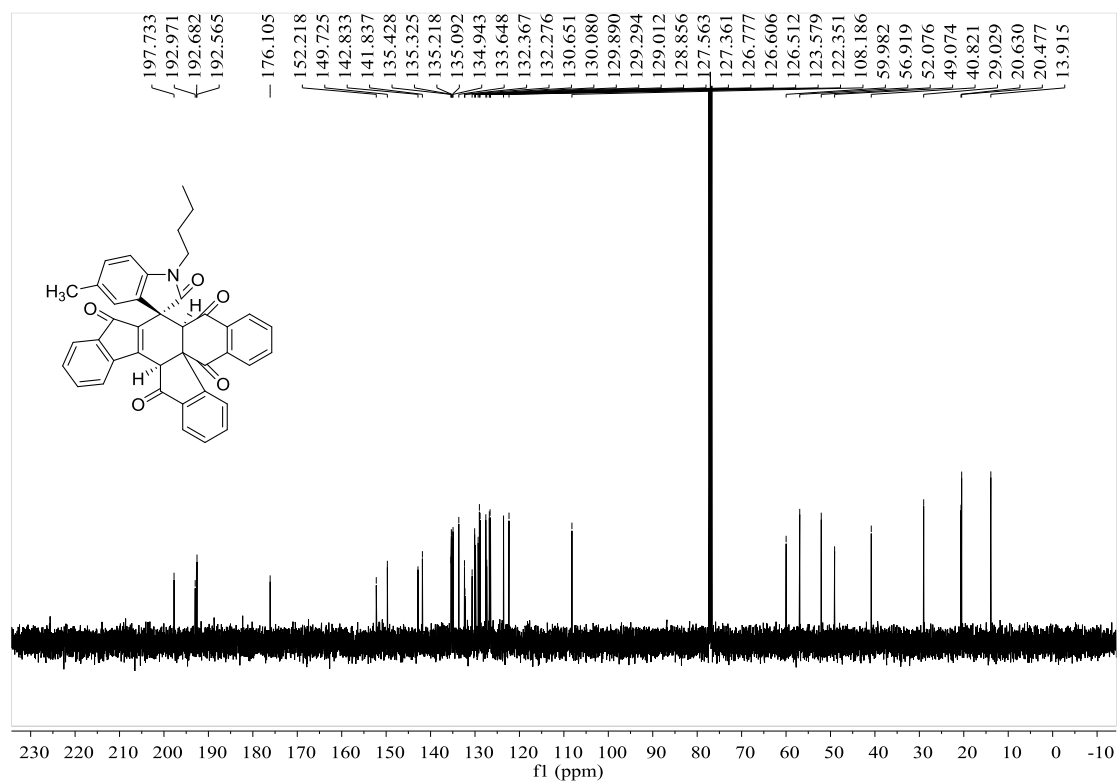
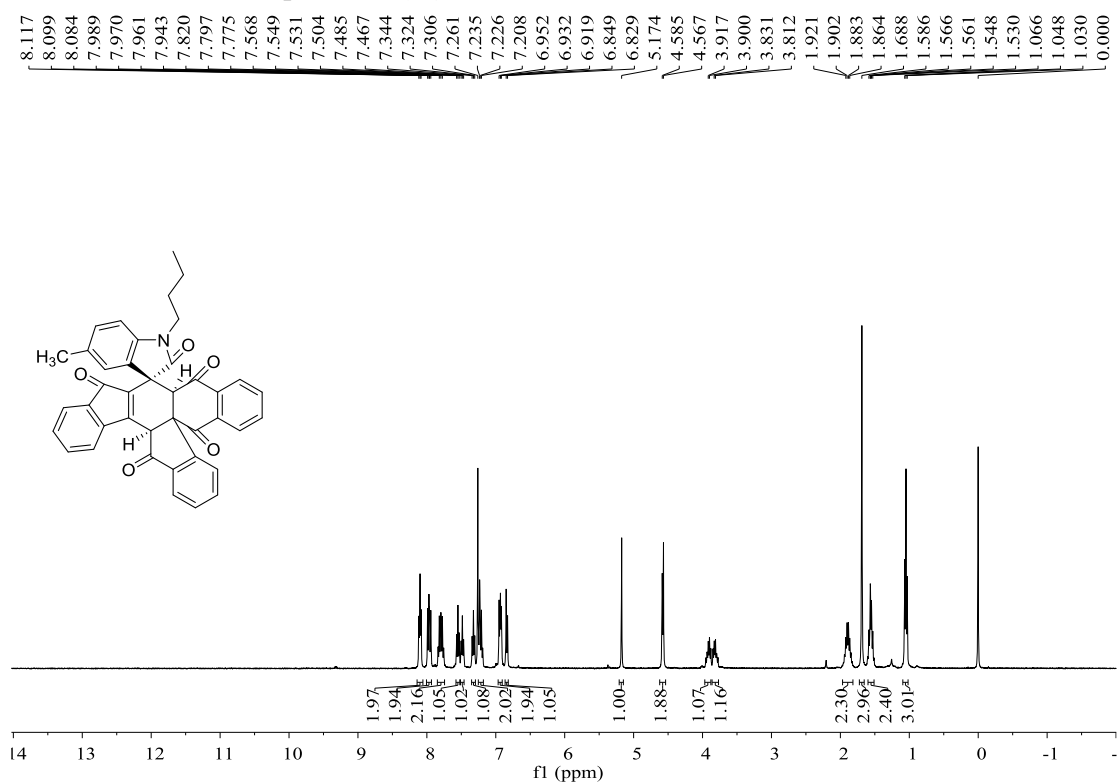


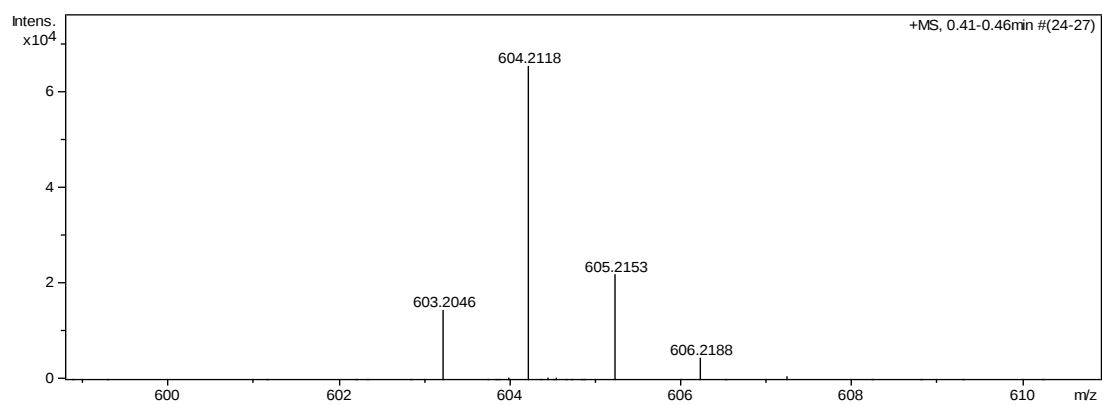
1'-benzyl-5'-fluoro-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8d):



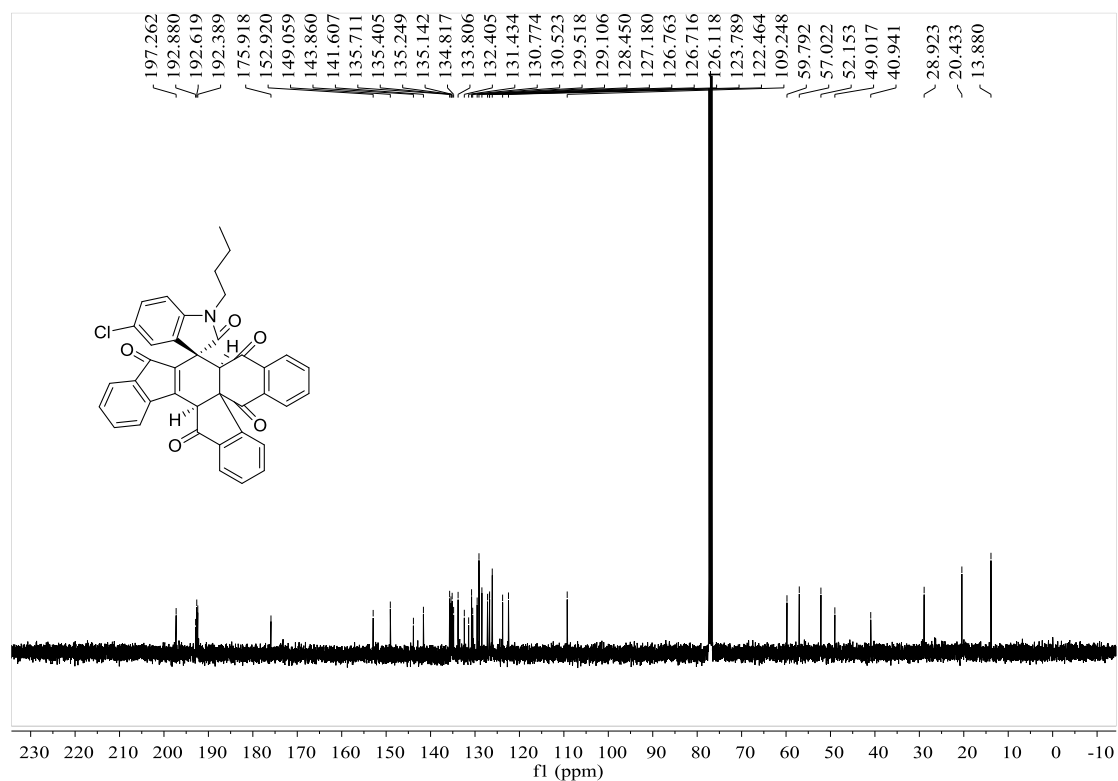
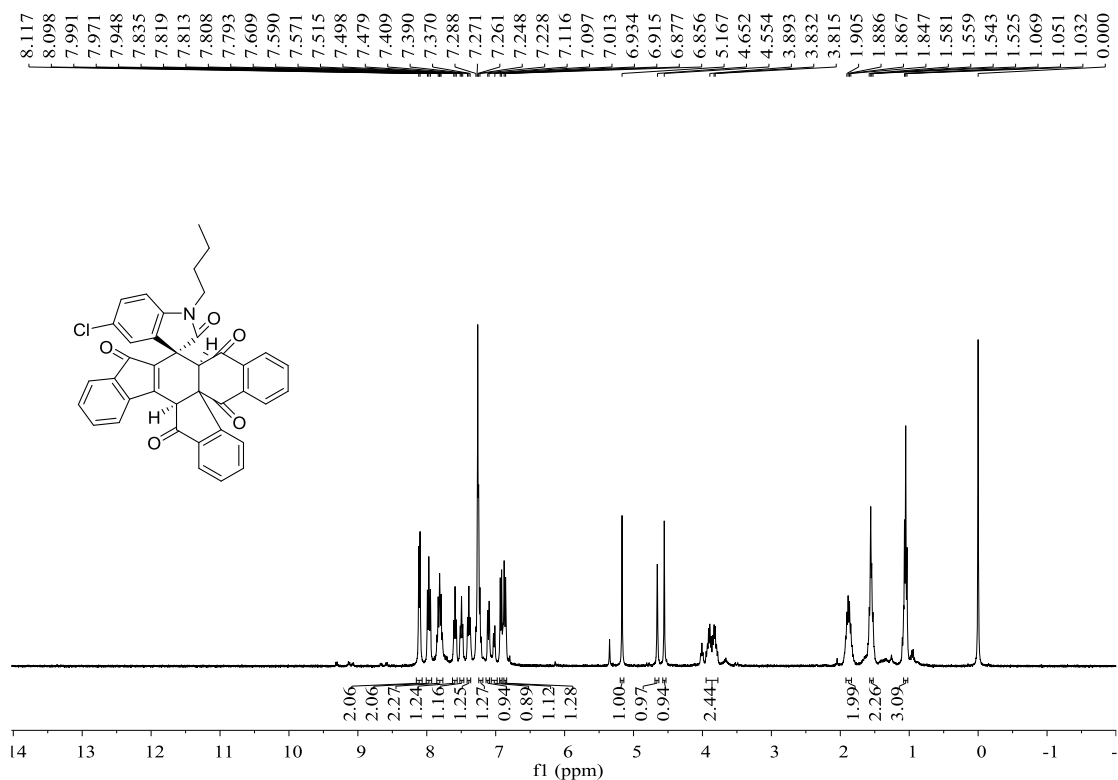


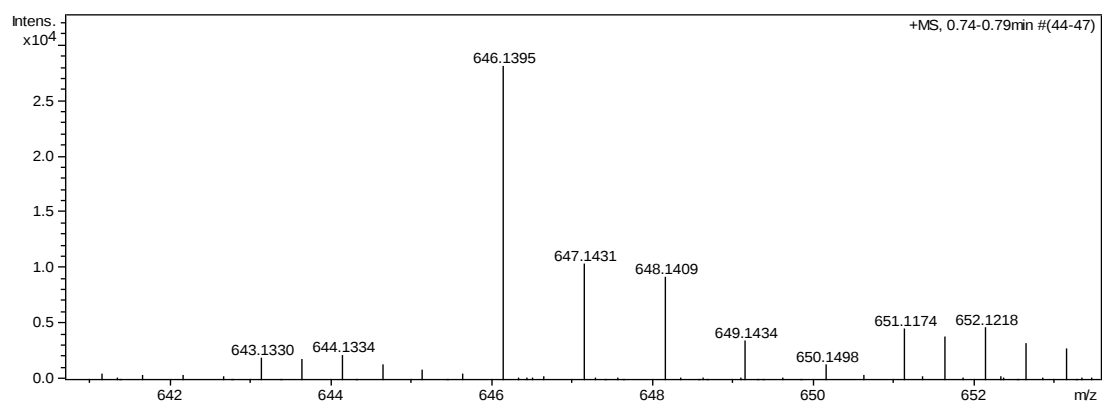
1'-butyl-5'-methyl-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8e):



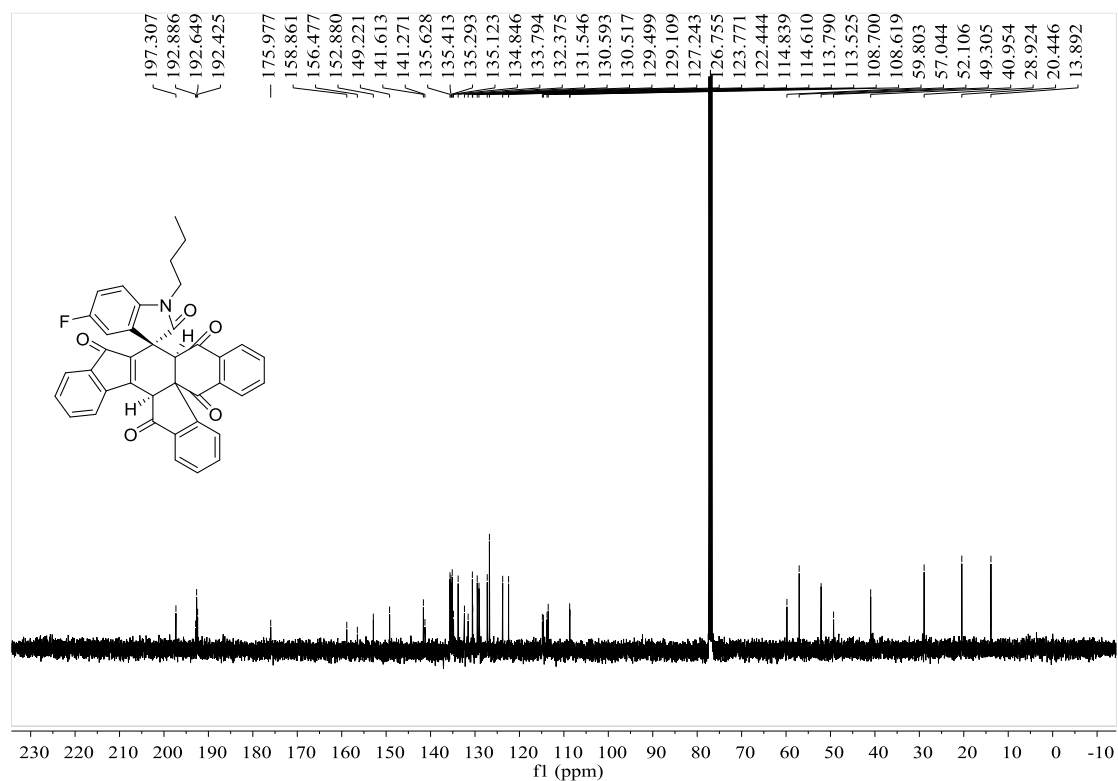
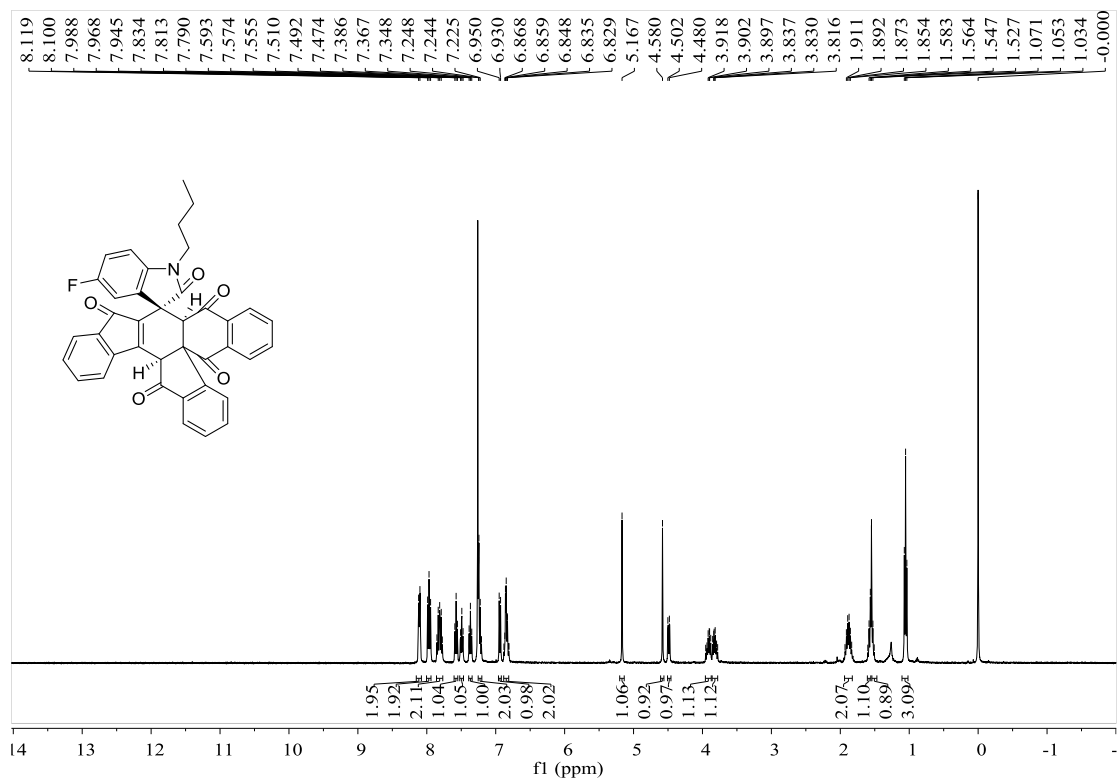


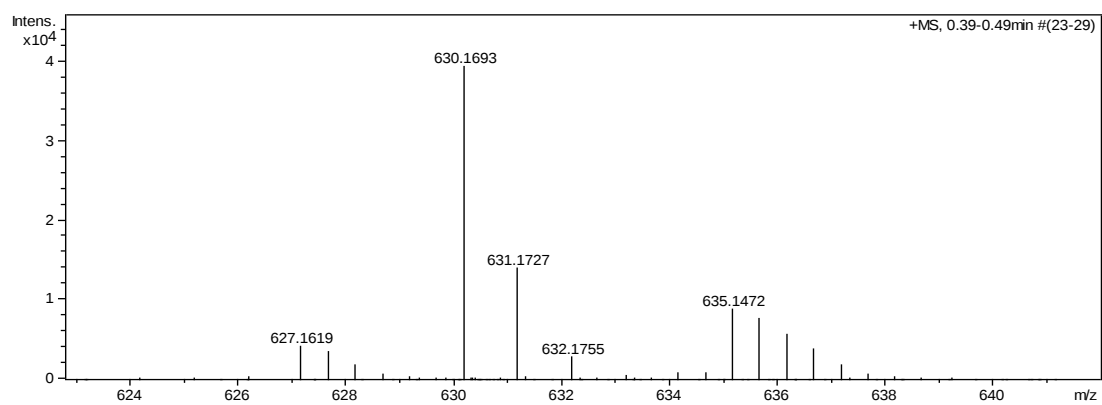
1'-butyl-5'-chloro-10a,16c-dihydro-5H,12H-spiro[diindeno[2,1-b:2',1'-d]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8f):





1'-butyl-5'-fluoro-10a,16c-dihydro-5*H*,12*H*-spiro[diindeno[2,1-*b*:2',1'-*d*]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8g):





5'-chloro-1'-methyl-10a,16c-dihydro-5*H*,12*H*-spiro[diindeno[2,1-*b*:2',1'-*d*]anthracene-11,3'-indoline]-2',5,10,12,17-pentaone (8h):

