

## Supporting Information

**Selective Construction of Spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine] and  
Spiro[indoline-3,3'-pyrrole] via [4+3]/[3+2] Cycloaddition Reaction of  $\alpha,\beta$ -Unsaturated  
Aldimines and MBH Adducts of Isatins**

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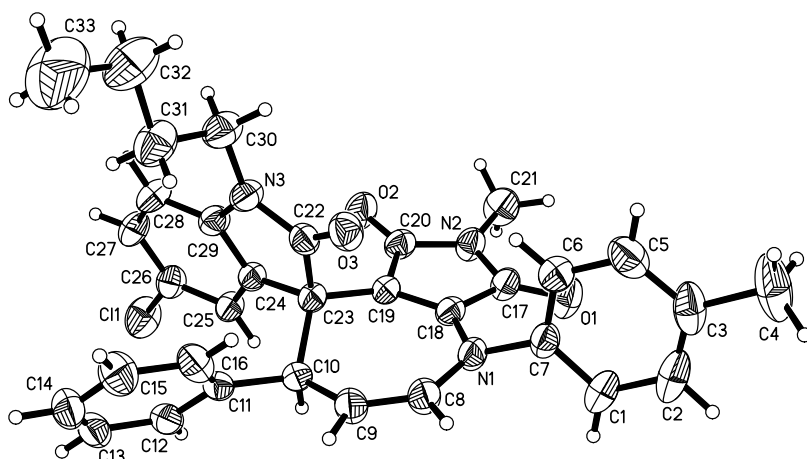


Fig. S1 ORTEP drawing (30%) of the crystal structure of **3e**

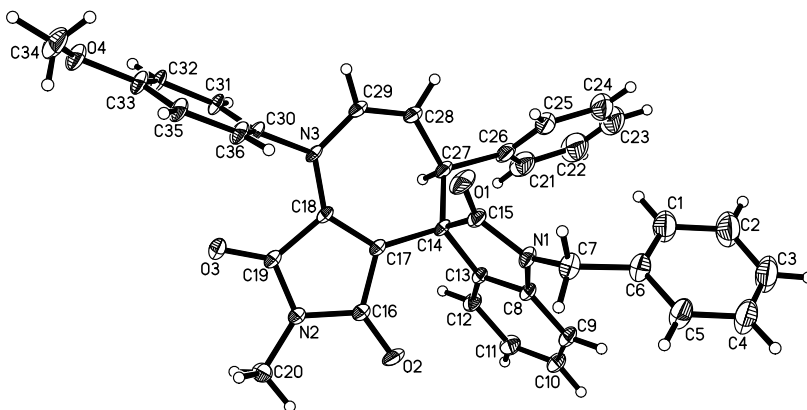


Fig. S2 ORTEP drawing (30%) of the crystal structure of **3g**

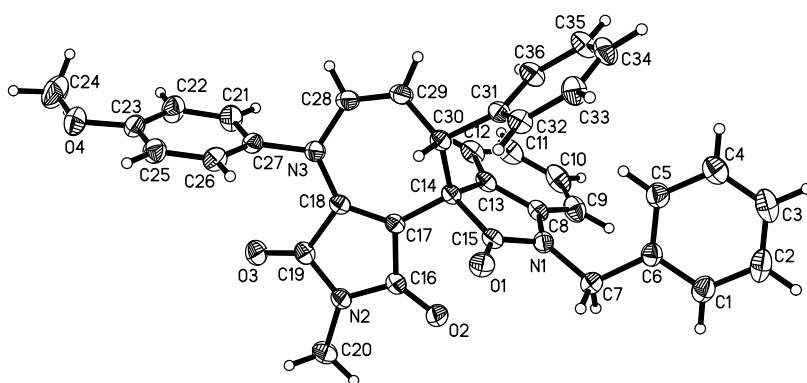


Fig. S3 ORTEP drawing (30%) of the crystal structure of **3g'**

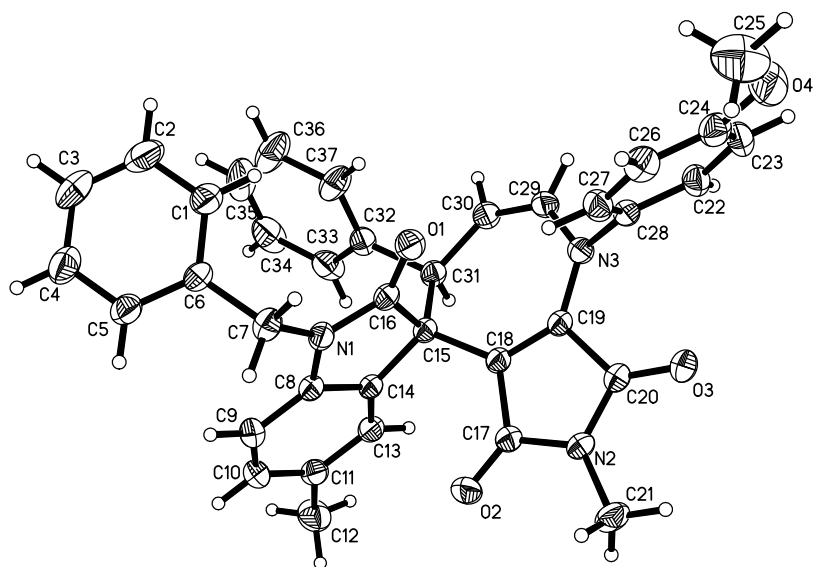


Fig. S4 ORTEP drawing (30%) of the crystal structure of **3h**

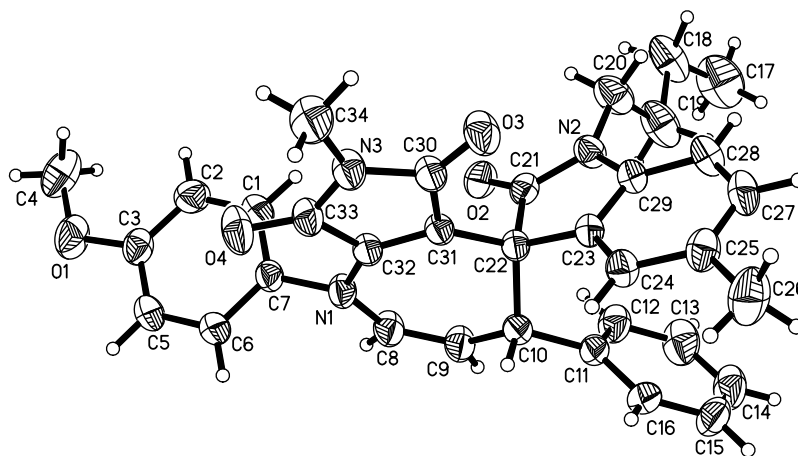


Fig. S5 ORTEP drawing (30%) of the crystal structure of **3j**

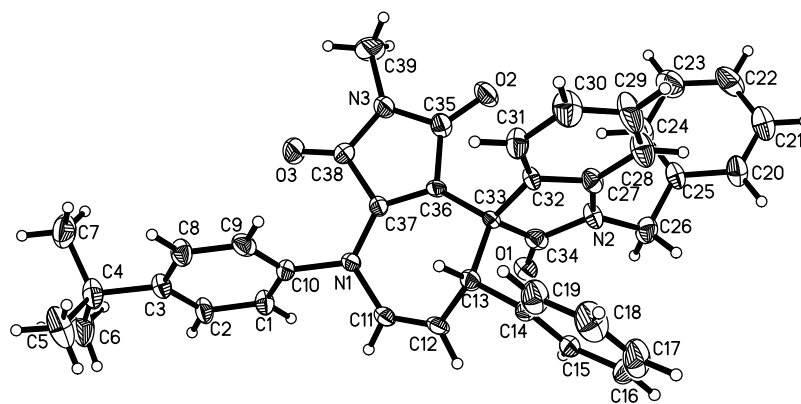


Fig. S6 ORTEP drawing (30%) of the crystal structure of **3k**

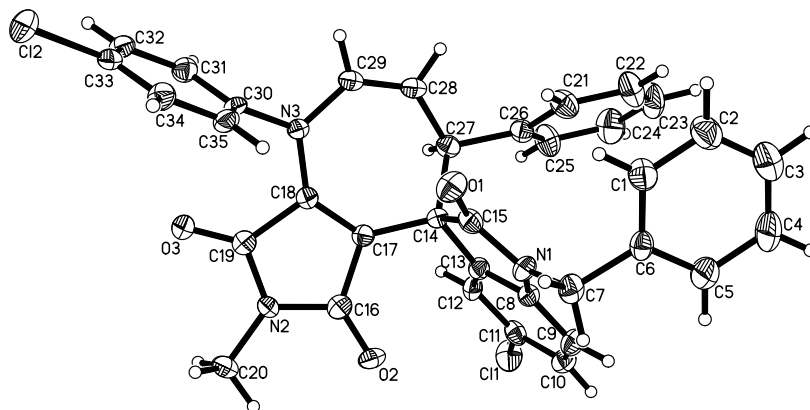


Fig. s7 ORTEP drawing (30%) of the crystal structure of **3m**

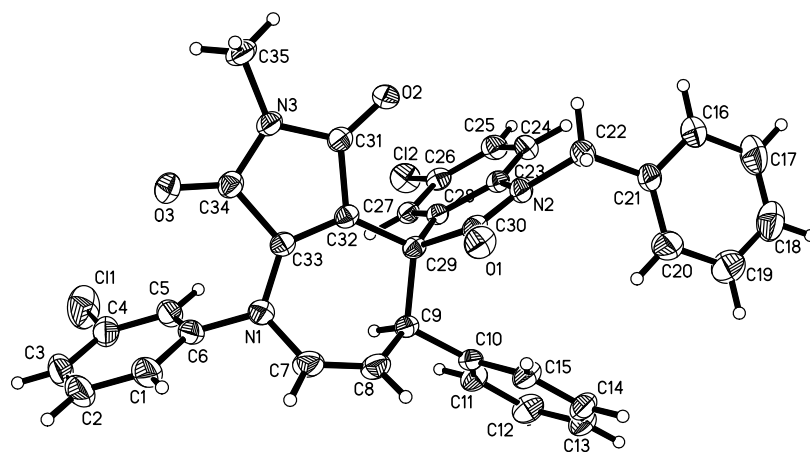


Fig. s8 ORTEP drawing (30%) of the crystal structure of **3n**

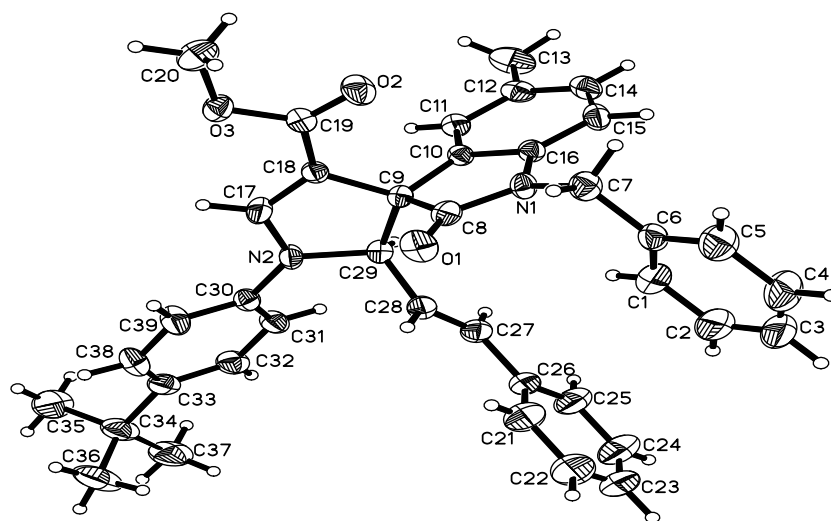


Fig. s9 ORTEP drawing (30%) of the crystal structure of **6d**

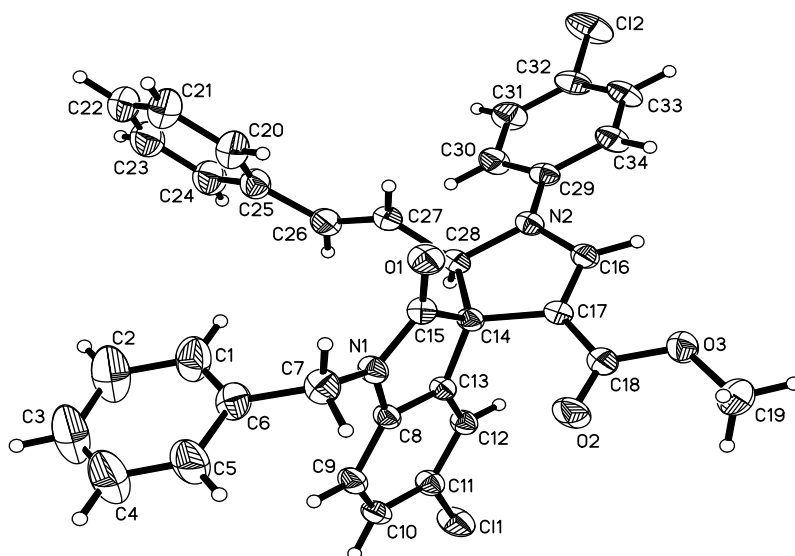


Fig. s10 ORTEP drawing (30%) of the crystal structure of **6g**

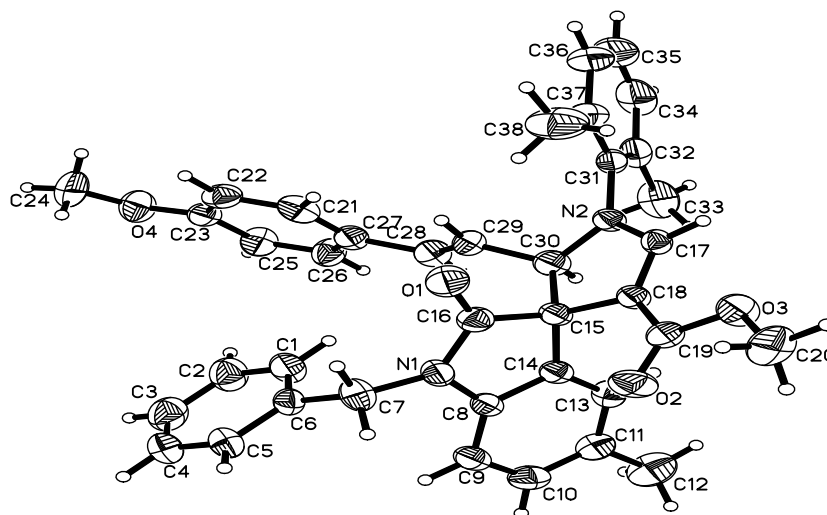


Fig. s11 ORTEP drawing (30%) of the crystal structure of **6k**

**Table S1** The single crystal data of compounds **3e**, **3g**, **3g'**

Phase	<b>3e</b>	<b>3g</b>	<b>3g'</b>
Empirical formula	C <sub>33</sub> H <sub>30</sub> ClN <sub>3</sub> O <sub>3</sub>	C <sub>36</sub> H <sub>29</sub> N <sub>3</sub> O <sub>4</sub>	C <sub>36</sub> H <sub>29</sub> N <sub>3</sub> O <sub>4</sub>
Formula weight	552.05	567.62	567.62
Temperature/K	296(2) K	296(2) K	296(2) K
Wavelength/ Å	0.71073	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic	Triclinic
Space group	P2(1)/n	P2(1)/c	P-1
<i>a</i> / Å	13.3537(11)	17.418(5)	11.262(3)
<i>b</i> / Å	9.0059(7)	9.262(3)	12.483(4)
<i>c</i> / Å	24.4471(19)	18.010(5)	13.478(4)
$\alpha$ (°)	90	90	66.218(9)
$\beta$ (°)	105.317(2)	98.824(7)	84.506(9)
$\gamma$ (°)	90	90	67.262(8)
<i>V</i> (Å <sup>3</sup> )	2835.6(4)	2871.1(13)	1595.3(9)
<i>Z</i>	4	4	2
Calculated density (g·cm <sup>-3</sup> )	1.293	1.313	1.182
Absorption coefficient (mm <sup>-1</sup> )	0.174	0.087	0.078
<i>F</i> (000)	1160	1192	596
$\theta$ range / (°)	2.421 to 26.000	2.289 to 25.000	3.002 to 25.999
Limiting indices	-16<= <i>h</i> <=16, -10<= <i>k</i> <=11, -30<= <i>l</i> <=30	-20<= <i>h</i> <=20, -11<= <i>k</i> <=10, -21<= <i>l</i> <=20	-13<= <i>h</i> <=13, -15<= <i>k</i> <=15, -16<= <i>l</i> <=16
Reflections collected/unique	37162 / 5568 [R(int) = 0.0762]	18110 / 4982 [R(int) = 0.1131]	21555 / 6208 [R(int) = 0.0496]
Completeness to theta	99.9 %	98.8 %	98.8 %
Max. and min. transmission	0.7456 and 0.6871	0.7456 and 0.3679	0.7456 and 0.6396
Refinement method	Full-matrix least-squares on F <sup>2</sup>	Full-matrix least-squares on F <sup>2</sup>	Full-matrix least-squares on F <sup>2</sup>
Data/restraints/parameters	5568 / 28 / 364	4982 / 609 / 455	6208 / 0 / 390
Goodness-of-fit on <i>F</i> <sup>2</sup>	1.013	1.045	1.031
Final <i>R</i> indices[I>2sigma(I)]	R1 = 0.0635, wR2 = 0.1472	R1 = 0.1145, wR2 = 0.2933	R1 = 0.0515, wR2 = 0.1163
<i>R</i> indices (all data)	R1 = 0.1538, wR2 = 0.1855	R1 = 0.1793, wR2 = 0.3221	R1 = 0.0990, wR2 = 0.1348
Largest diff. peak and hole /(e · Å <sup>-3</sup> )	0.346 and -0.338	0.273 and -0.321	0.197 and -0.193

**Table S2** The single crystal data of compounds **3h**, **3j**, **3k**

Phase	<b>3h</b>	<b>3j</b>	<b>3k</b>
Empirical formula	C <sub>37</sub> H <sub>31</sub> N <sub>3</sub> O <sub>4</sub>	C <sub>34</sub> H <sub>33</sub> N <sub>3</sub> O <sub>4</sub>	C <sub>39</sub> H <sub>35</sub> N <sub>3</sub> O <sub>3</sub>
Formula weight	581.65	547.63	593.70
Temperature/K	296(2) K	296(2) K	296(2) K
Wavelength/ Å	0.71073	0.71073	0.71073
Crystal system	Triclinic	Monoclinic	Monoclinic
Space group	P-1	P2(1)/n	P2(1)/c
<i>a</i> / Å	8.9086(4)	13.6892(12)	12.170(3)
<i>b</i> / Å	13.0844(5)	9.2534(9)	20.468(4)
<i>c</i> / Å	13.9872(5)	24.022(2)	26.223(6)
$\alpha$ (°)	68.3519(11)	90	90
$\beta$ (°)	79.9344(12)	104.263(3)	93.494(5)
$\gamma$ (°)	88.3197(13)	90	90
<i>V</i> (Å <sup>3</sup> )	1490.98(10)	2949.1(5)	6520(3)
<i>Z</i>	2	4	8
Calculated density (g·cm <sup>-3</sup> )	1.296	1.233	1.210
Absorption coefficient (mm <sup>-1</sup> )	0.085	0.081	0.077
<i>F</i> (000)	612	1160	2512
$\theta$ range / (°)	2.574 to 25.999	2.684 to 25.999	1.676 to 25.000
Limiting indices	-10<= <i>h</i> <=10, -16<= <i>k</i> <=16, -16<= <i>l</i> <=17	-16<= <i>h</i> <=15, -11<= <i>k</i> <=11, -29<= <i>l</i> <=29	-14<= <i>h</i> <=14, -24<= <i>k</i> <=24, -31<= <i>l</i> <=29
Reflections collected/unique	21201 / 5797 [R(int) = 0.0325]	40435 / 5778 [R(int) = 0.0678]	53873 / 11478 [R(int) = 0.3024]
Completeness to theta	99.1 %	99.3 %	99.9 %
Max. and min. transmission	0.7456 and 0.7047	0.7456 and 0.6580	0.7456 and 0.6297
Refinement method	Full-matrix least-squares on F <sup>2</sup>	Full-matrix least-squares on F <sup>2</sup>	Full-matrix least-squares on F <sup>2</sup>
Data/restraints/parameters	5797 / 0 / 400	5778 / 72 / 394	11478 / 0 / 819
Goodness-of-fit on <i>F</i> <sup>2</sup>	1.029	1.030	0.872
Final <i>R</i> indices[I>2sigma(I)]	R1 = 0.0476, wR2 = 0.1118	R1 = 0.0603, wR2 = 0.1238	R1 = 0.0935, wR2 = 0.1344
<i>R</i> indices (all data)	R1 = 0.0741, wR2 = 0.1252	R1 = 0.1303, wR2 = 0.1527	R1 = 0.3359, wR2 = 0.1756
Largest diff. peak and hole /(e · Å <sup>-3</sup> )	0.180 and -0.198	0.222 and -0.257	0.225 and -0.222

**Table S3** The single crystal data of compounds **3m**, **3n**, **6d**

Phase	<b>3m</b>	<b>3n</b>	<b>6d</b>
Empirical formula	C <sub>35</sub> H <sub>25</sub> Cl <sub>2</sub> N <sub>3</sub> O <sub>3</sub>	C <sub>35</sub> H <sub>25</sub> Cl <sub>2</sub> N <sub>3</sub> O <sub>3</sub>	C <sub>39</sub> H <sub>38</sub> N <sub>2</sub> O <sub>3</sub>
Formula weight	606.48	606.48	582.7
Temperature/K	296(2) K	296(2) K	296(2) K
Wavelength/ Å	0.71073	0.71073	0.71073
Crystal system	Monoclinic	Triclinic	Monoclinic
Space group	P2(1)/n	P-1	P2/n
<i>a</i> / Å	18.3180(15)	9.1967(9)	20.948(2)
<i>b</i> / Å	9.2296(7)	10.8849(11)	6.4429(6)
<i>c</i> / Å	34.735(3)	16.9182(16)	24.542(2)
$\alpha$ (°)	90	72.099(3)	90
$\beta$ (°)	97.205(2)	88.525(3)	92.486(3)
$\gamma$ (°)	90	70.068(3)	90
<i>V</i> (Å <sup>3</sup> )	5826.2(8)	1509.2(3)	3309.1(5)
<i>Z</i>	8	2	4
Calculated density (g·cm <sup>-3</sup> )	1.383	1.335	1.170
Absorption coefficient (mm <sup>-1</sup> )	0.265	0.256	0.074
<i>F</i> (000)	2512	628	1240
$\theta$ range / (°)	1.199 to 25.000	1.270 to 26.000	2.504 to 25.000
Limiting indices	-21 ≤ <i>h</i> ≤ 21, -10 ≤ <i>k</i> ≤ 10, -41 ≤ <i>l</i> ≤ 40	-11 ≤ <i>h</i> ≤ 11, -13 ≤ <i>k</i> ≤ 13, -20 ≤ <i>l</i> ≤ 20	-24 ≤ <i>h</i> ≤ 21, -7 ≤ <i>k</i> ≤ 7, -29 ≤ <i>l</i> ≤ 29
Reflections collected/unique	58255 / 10238 [R(int) = 0.1603]	21424 / 5915 [R(int) = 0.0305]	28096 / 5829 [R(int) = 0.1464]
Completeness to theta	99.9 %	99.5 %	99.9 %
Max. and min. transmission	0.7456 and 0.6518	0.7455 and 0.6880	0.7456 and 0.6924
Refinement method	Full-matrix least-squares on <i>F</i> <sup>2</sup>	Full-matrix least-squares on <i>F</i> <sup>2</sup>	Full-matrix least-squares on <i>F</i> <sup>2</sup>
Data/restraints/parameters	10238 / 0 / 777	5915 / 0 / 389	5829 / 85 / 433
Goodness-of-fit on <i>F</i> <sup>2</sup>	0.896	1.011	0.932
Final <i>R</i> indices [ <i>I</i> > 2σ( <i>I</i> )]	R1 = 0.0660, wR2 = 0.1115	R1 = 0.0455, wR2 = 0.1079	R1 = 0.0699, wR2 = 0.1185
<i>R</i> indices (all data)	R1 = 0.2153, wR2 = 0.1380	R1 = 0.0692, wR2 = 0.1211	R1 = 0.2201, wR2 = 0.1447
Largest diff. peak and hole /(e · Å <sup>-3</sup> )	0.235 and -0.265	0.208 and -0.322	0.168 and -0.193



**Table S4** The single crystal data of compounds **6g**, **6k**

Phase	<b>6g</b>	<b>6k</b>
Empirical formula	C <sub>34</sub> H <sub>26</sub> Cl <sub>2</sub> N <sub>2</sub> O <sub>3</sub>	C <sub>38</sub> H <sub>36</sub> N <sub>2</sub> O <sub>4</sub>
Formula weight	581.47	584.69
Temperature/K	296(2) K	296(2) K
Wavelength/ Å	0.71073	0.71073
Crystal system	Triclinic	Monoclinic
Space group	P-1	P2(1)/n
<i>a</i> / Å	9.7184(14)	13.0507(7)
<i>b</i> / Å	12.5333(18)	11.5992(6)
<i>c</i> / Å	14.973(2)	21.3575(10)
α (°)	99.526(5)	90
β (°)	106.553(4)	91.006(2)
γ (°)	108.581(4)	90
<i>V</i> (Å <sup>3</sup> )	1589.8(4)	3232.6(3)
<i>Z</i>	2	4
Calculated density (g·cm <sup>-3</sup> )	1.215	1.201
Absorption coefficient (mm <sup>-1</sup> )	0.239	0.078
<i>F</i> (000)	604	1240
θ range / (°)	2.297 to 24.999	2.349 to 24.999
Limiting indices	-11<= <i>h</i> <=11, -14<= <i>k</i> <=14, -17<= <i>l</i> <=17	-15<= <i>h</i> <=15, -13<= <i>k</i> <=13, -25<= <i>l</i> <=25
Reflections collected/unique	20731 / 5540 [R(int) = 0.0689]	28015 / 5663 [R(int) = 0.0461]
Completeness to theta	99.0 %	99.5 %
Max. and min. transmission	0.7456 and 0.6434	0.7456 and 0.6889
Refinement method	Full-matrix least-squares on F <sup>2</sup>	Full-matrix least-squares on F <sup>2</sup>
Data/restraints/parameters	5540 / 103 / 371	5663 / 0 / 402
Goodness-of-fit on <i>F</i> <sup>2</sup>	1.069	1.012
Final <i>R</i> indices[I>2σ(I)]	R1 = 0.0662, wR2 = 0.1790	R1 = 0.0756, wR2 = 0.1887
<i>R</i> indices (all data)	R1 = 0.1137, wR2 = 0.1966	R1 = 0.1233, wR2 = 0.2066
Largest diff. peak and hole /(e · Å <sup>-3</sup> )	0.287 and -0.308	0.618 and -0.294

## Experimental section

### 1. General procedure for the reactions of $\alpha,\beta$ -unsaturated aldimines and MBH maleimides of

**isatins:** A mixture of  $\alpha,\beta$ -unsaturated *N*-arylaldehyde (0.10 mmol), MBH maleimide of isatin (0.10 mmol), and DABCO (20 mmol%) in CH<sub>3</sub>CN (10.0 mL) was stirred at 50 °C for 16 hours. After removing the solvent by rotatory evaporation, the resulting residue was purified by column chromatography on silica gel with ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent to afford the desired products **3a-3v** and minor isomers **3g'** and **3i'**.

***cis*-1-Benzyl-5-chloro-7'-methyl-4'-phenyl-1'-(*p*-tolyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3a):** ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.526 g, 90%, m.p. 182-184 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$ : 7.30-7.26 (m, 5H), 7.24 (t,  $J$  = 6.4 Hz, 2H), 7.17-7.15 (m, 2H), 7.11-7.07 (m, 2H), 7.05-7.01 (m, 2H), 6.97-6.94 (m, 3H), 6.54 (dd,  $J_1$  = 8.8 Hz,  $J_2$  = 2.0 Hz, 1H), 6.23 (d,  $J$  = 8.4 Hz, 1H), 5.61 (dd,  $J_1$  = 8.8 Hz,  $J_2$  = 2.0 Hz, 1H), 4.97 (d,  $J$  = 16.0 Hz, 1H), 4.49 (d,  $J$  = 16.0 Hz, 1H), 3.97 (dd,  $J_1$  = 5.6 Hz,  $J_2$  = 2.0 Hz, 1H), 2.77 (s, 3H), 2.43 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$ : 176.2, 168.8, 165.6, 141.5, 141.1, 140.5, 138.0, 138.0, 135.0, 134.9, 133.6, 130.2, 128.9, 128.7, 128.0, 127.8, 127.6, 127.4, 127.1, 126.0, 123.6, 115.5, 109.7, 106.3, 58.0, 50.6, 44.3, 23.6, 21.3; IR(KBr)  $\nu$ : 3746, 3028, 1761, 1709, 1648, 1611, 1487, 1468, 1451, 1380, 1348, 1270, 1221, 1176, 1130, 1102, 1084, 1041, 1030, 1009, 947, 862, 841, 811, 779, 754 cm<sup>-1</sup>; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For C<sub>36</sub>H<sub>28</sub>ClN<sub>3</sub>NaO<sub>3</sub> ([M+Na]<sup>+</sup>): 608.1711, found: 608.1735.

***cis*-1-Benzyl-7'-methyl-1',4'-diphenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3b):** ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.462 g, 86%, m.p. 258-260 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$ : 7.45 (d,  $J$  = 8.8 Hz, 2H), 7.33-7.28 (m, 4H), 7.23 (d,  $J$  = 6.8 Hz, 1H), 7.18 (d,  $J$  = 7.6 Hz, 2H), 7.13 (d,  $J$  = 6.8 Hz, 1H), 7.08-6.93 (m, 8H), 6.52 (dd,  $J_1$  = 8.8 Hz,  $J_2$  = 2.0 Hz, 1H), 6.35 (d,  $J$  = 7.6 Hz, 1H), 5.66 (dd,  $J_1$  = 9.2 Hz,  $J_2$  = 3.6 Hz, 1H), 4.96 (d,  $J$  = 16.0 Hz, 1H), 4.54 (d,  $J$  = 16.0 Hz, 1H), 4.01-4.00 (m, 1H), 2.76 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$ : 176.4, 168.66, 165.8, 142.7, 141.9, 140.7, 138.1, 135.4, 134.0, 133.7, 131.8, 129.7, 129.0, 128.6, 128.3, 127.7, 127.5, 127.3, 127.2, 127.0, 123.2, 122.3, 116.4, 108.9, 108.38, 57.9, 50.6, 44.3, 23.6; IR(KBr)  $\nu$ : 3746, 3028, 1760, 1709, 1648, 1611, 1487, 1468, 1451, 1380, 1347, 1270, 1221, 1176, 1130, 1101, 1083, 1041, 1030, 1008, 947, 862, 841, 811, 779, 754 cm<sup>-1</sup>; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd.

For C<sub>35</sub>H<sub>27</sub>N<sub>3</sub>NaO<sub>3</sub> ([M+Na]<sup>+</sup>): 560.1945, found: 560.1960.

**cis-1-Benzyl-7'-methyl-4'-phenyl-1'-(p-tolyl)-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (3c):** ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.479 g, 87%, m.p. 240-242 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.28 (s, 5H), 7.20-7.13 (m, 4H), 7.05-6.96 (m, 8H), 6.54 (d, *J* = 8.8 Hz, 1H), 6.35 (d, *J* = 7.2 Hz, 1H), 5.62 (s, 1H), 4.96 (d, *J* = 16.0 Hz, 1H), 4.56 (d, *J* = 16.0 Hz, 1H), 4.00 (s, 1H), 2.75 (s, 3H), 2.43 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 176.6, 168.9, 165.9, 142.0, 141.8, 141.0, 138.4, 137.9, 135.5, 134.6, 132.1, 130.2, 129.0, 128.6, 128.2, 127.7, 127.3, 127.3, 1267.0, 126.0, 123.2, 122.3, 115.8, 108.8, 107.3, 58.0, 50.8, 44.3, 23.5, 21.3; IR(KBr) ν: 3726, 1903, 1854, 1819, 1773, 1758, 1734, 1734, 1710, 1700, 1676, 1653, 1611, 1560, 1546, 1491, 1467, 1449, 1380, 1350, 1175, 1009, 948, 811, 751 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>36</sub>H<sub>29</sub>N<sub>3</sub>NaO<sub>3</sub> ([M+Na]<sup>+</sup>): 574.2101, found: 574.2114.

**cis-1-Butyl-5,7'-dimethyl-4'-phenyl-1'-(p-tolyl)-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (3d):** ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.398 g, 75%, m.p. 212-214 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.28 (d, *J* = 9.2 Hz, 3H), 7.24 (d, *J* = 5.2 Hz, 1H), 7.04-7.00 (m, 3H), 6.96-6.94 (m, 2H), 6.91 (s, 1H), 6.89 (d, *J* = 8.0 Hz, 1H), 6.52 (d, *J* = 9.2 Hz, 1H), 6.38 (d, *J* = 8.0 Hz, 1H), 5.60 (dd, *J*<sub>1</sub> = 9.2 Hz, *J*<sub>2</sub> = 3.6 Hz, 1H), 3.93-3.91 (m, 1H), 3.61-3.54 (m, 1H), 3.46-3.39 (m, 1H), 2.72 (s, 3H), 2.42 (s, 3H), 2.32 (s, 3H), 1.54-1.45 (m, 2H), 1.36-1.29 (m, 2H), 0.92 (t, *J* = 7.2 Hz, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 176.2, 168.8, 167.0, 141.8, 140.9, 139.8, 138.5, 137.8, 134.5, 132.2, 131.3, 130.1, 129.1, 128.4, 127.5, 126.8, 126.0, 124.0, 115.9, 107.6, 107.6, 57.9, 50.6, 40.3, 29.2, 23.5, 21.2, 20.2, 13.7; IR(KBr) ν: 3726, 1704, 1603, 1504, 1439, 1386, 1357, 1256, 1202, 1132, 814, 757 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>34</sub>H<sub>34</sub>N<sub>3</sub>O<sub>3</sub> ([M+H]<sup>+</sup>): 532.2595, found: 532.2615.

**cis-1-Butyl-5-chloro-7'-methyl-4'-phenyl-1'-(p-tolyl)-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (3e):** ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.441 g, 80%, m.p. 238-240 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.28 (d, *J* = 8.4 Hz, 3H), 7.24 (s, 1H), 7.09-7.06 (m, 5H), 6.97-6.95 (m, 2H), 6.52 (d, *J* = 8.8 Hz, 1H), 6.41 (d, *J* = 8.0 Hz, 1H), 5.57 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 3.6 Hz, 1H), 3.92 (dd, *J*<sub>1</sub> = 5.2 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 3.62-3.55 (m, 1H), 3.45-3.38 (m, 1H), 2.74 (s, 3H), 2.43 (s, 3H), 1.55-1.44

(m, 2H), 1.37-1.29 (m, 2H), 0.92 (t,  $J = 7.2$  Hz, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 175.9, 168.7, 165.6, 141.6, 141.1, 140.8, 138.0, 137.9, 134.7, 133.7, 130.2, 128.9, 127.9, 127.7, 127.3, 127.1, 126.0, 123.6, 115.4, 108.7, 106.4, 57.8, 50.6, 40.4, 29.1, 23.5, 21.2, 20.2, 13.7; IR(KBr)  $\nu$ : 1712, 1698, 1661, 1607, 1508, 1488, 1439, 1386, 1349, 1256, 1113, 815, 757  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{33}\text{H}_{30}\text{ClN}_3\text{NaO}_5$  ( $[\text{M}+\text{Na}]^+$ ): 574.1868, found: 574.1896.

***cis*-1-Benzyl-6-chloro-7'-methyl-4'-phenyl-1'-(*p*-tolyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3f)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.521 g, 89%, m.p. 230-232 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.32-7.27 (m, 6H), 7.24 (s, 1H), 7.18 (d,  $J = 7.6$  Hz, 2H), 7.10-7.00 (m, 4H), 6.94 (t,  $J = 8.4$  Hz, 1H), 6.52 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 2.0$  Hz, 1H), 6.35 (s, 1H), 5.58 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 3.6$  Hz, 1H), 4.93 (d,  $J = 16.0$  Hz, 1H), 4.51 (d,  $J = 16.0$  Hz, 1H), 3.96 (dd,  $J_1 = 5.6$  Hz,  $J_2 = 2.0$  Hz, 1H), 2.76 (s, 3H), 2.43 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.6, 168.8, 165.7, 143.1, 141.6, 141.1, 138.1, 138.0, 134.9, 134.7, 133.7, 130.5, 130.2, 128.9, 128.7, 127.8, 127.5, 127.2, 126.0, 124.0, 122.2, 115.5, 109.5, 109.4, 106.5, 57.6, 50.7, 44.4, 23.5, 21.2; IR(KBr)  $\nu$ : 1760, 1704, 1607, 1507, 1489, 1441, 1373, 1253, 1171, 1110, 1071, 1014, 814, 753  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{36}\text{H}_{28}\text{ClN}_3\text{NaO}_3$  ( $[\text{M}+\text{Na}]^+$ ): 608.1711, found: 608.1711.

***cis*-1-Benzyl-1'-(4-methoxyphenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3g)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.431 g, 76%, m.p. 204-206 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.30-7.28 (m, 3H), 7.23 (s, 1H), 7.19 (d,  $J = 7.2$  Hz, 2H), 7.13 (d,  $J = 7.2$  Hz, 1H), 7.07-6.93 (m, 10H), 6.47 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 1.6$  Hz, 1H), 6.35 (d,  $J = 8.0$  Hz, 1H), 5.59 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 5.2$  Hz, 1H), 4.96 (d,  $J = 16.0$  Hz, 1H), 4.56 (d,  $J = 16.0$  Hz, 1H), 3.98 (d,  $J = 5.2$  Hz, 1H), 3.86 (s, 3H), 2.75 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.6, 168.9, 165.9, 159.1, 141.9, 141.0, 138.4, 137.1, 135.5, 134.8, 132.1, 129.0, 128.6, 128.1, 127.6, 127.5, 127.3, 127.2, 126.9, 123.2, 122.3, 115.5, 114.6, 108.8, 106.7, 58.1, 55.5, 50.8, 44.2, 23.5; IR(KBr)  $\nu$ : 1762, 1704, 1620, 1507, 1437, 1386, 1333, 1245, 1169, 1080, 1008, 840, 809  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{36}\text{H}_{29}\text{N}_3\text{NaO}_4$  ( $[\text{M}+\text{Na}]^+$ ): 590.2050, found: 590.2084.

***trans*-1-Benzyl-1'-(4-methoxyphenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3g')**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.034 g, 6%, m.p. 213-215 °C;  $^1\text{H}$

NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$ : 7.38 (d,  $J = 7.2$  Hz, 1H), 7.29 (d,  $J = 8.4$  Hz, 2H), 7.23 (d,  $J = 7.2$  Hz, 1H), 7.15-7.06 (m, 2H), 7.00 (d,  $J = 8.8$  Hz, 2H), 6.78 (d,  $J = 7.6$  Hz, 2H), 6.62-6.61 (m, 2H), 6.40 (dd,  $J_1 = 9.2$  Hz,  $J_2 = 2.4$  Hz, 1H), 6.33 (d,  $J = 7.6$  Hz, 1H), 5.36 (dd,  $J_1 = 9.2$  Hz,  $J_2 = 5.2$  Hz, 1H), 4.73 (d,  $J = 16.4$  Hz, 1H), 4.49 (d,  $J = 16.4$  Hz, 1H), 4.34 (dd,  $J_1 = 4.8$  Hz,  $J_2 = 2.0$  Hz, 1H), 3.87 (s, 3H), 2.81 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (150 MHz, CDCl<sub>3</sub>)  $\delta$ : 174.8, 168.9, 165.9, 159.2, 144.7, 140.5, 138.5, 136.9, 135.4, 132.3, 129.1, 128.5, 128.0, 127.8, 127.4, 126.9, 126.5, 124.7, 122.1, 115.9, 114.6, 108.9, 57.0, 55.5, 49.4, 44.0, 29.7, 29.3, 27.2, 23.6, 22.7; IR(KBr)  $\nu$ : 1761, 1703, 1618, 1506, 1438, 1384, 1313, 1225, 1167, 1081, 1007, 841, 806 cm<sup>-1</sup>; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For C<sub>36</sub>H<sub>29</sub>N<sub>3</sub>NaO<sub>4</sub> ([M+Na]<sup>+</sup>): 590.2050, found: 590.2086.

**cis-1-Benzyl-1'-(4-methoxyphenyl)-5,7'-dimethyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'H)-trione (3h)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.482 g, 83%, m.p. 203-205 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$ : 7.30 (d,  $J = 8.4$  Hz, 2H), 7.25-7.22 (m, 2H), 7.18 (d,  $J = 7.6$  Hz, 2H), 7.08-6.94 (m, 9H), 6.79 (d,  $J = 8.0$  Hz, 1H), 6.49 (d,  $J = 8.8$  Hz, 1H), 6.22 (d,  $J = 8.0$  Hz, 1H), 5.60 (dd,  $J_1 = 8.0$  Hz,  $J_2 = 5.6$  Hz, 1H), 4.96 (d,  $J = 16.0$  Hz, 1H), 4.51 (d,  $J = 16.0$  Hz, 1H), 3.96 (d,  $J = 4.8$  Hz, 1H), 3.86 (s, 3H), 2.76 (s, 3H), 2.29 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$ : 176.6, 168.9, 166.0, 159.0, 140.9, 139.5, 138.5, 137.1, 135.6, 134.9, 132.2, 131.6, 129.0, 128.6, 128.4, 127.6, 127.5, 127.2, 127.1, 126.9, 123.9, 115.8, 114.6, 108.5, 107.0, 58.2, 55.5, 50.7, 44.2, 23.5, 21.3; IR(KBr)  $\nu$ : 1764, 1702, 1620, 1508, 1437, 1386, 1336, 1301, 1246, 1187, 1119, 1081, 1011, 839, 809, 769, 751 cm<sup>-1</sup>; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For C<sub>37</sub>H<sub>32</sub>N<sub>3</sub>O<sub>4</sub> ([M+H]<sup>+</sup>): 582.2387, found: 582.2402.

**cis-1-Benzyl-5-chloro-1'-(4-methoxyphenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'H)-trione (3i)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.481 g, 80%, m.p. 199-201 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>)  $\delta$ : 7.29 (d,  $J = 8.4$  Hz, 3H), 7.23 (d,  $J = 7.2$  Hz, 1H), 7.16 (d,  $J = 7.2$  Hz, 2H), 7.11-6.95 (m, 10H), 6.50 (d,  $J = 8.8$  Hz, 1H), 6.24 (d,  $J = 8.4$  Hz, 1H), 5.59 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 5.2$  Hz, 1H), 4.98 (d,  $J = 16.0$  Hz, 1H), 4.49 (d,  $J = 16.0$  Hz, 1H), 3.96 (d,  $J = 3.6$  Hz, 1H), 3.87 (s, 3H), 2.78 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>)  $\delta$ : 176.3, 168.8, 165.7, 159.2, 141.1, 140.5, 138.0, 136.9, 135.1, 135.0, 133.7, 129.0, 128.7, 128.0, 127.8, 127.7, 127.5, 127.5, 127.1, 123.6, 115.3, 114.6, 109.7, 105.9, 58.0, 55.5, 50.7, 44.3, 23.6; IR(KBr)  $\nu$ : 1761, 1701, 1629, 1506,

1435, 1381, 1335, 1309, 1242, 1189, 1110, 1084, 1014, 840, 801, 764, 752 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>36</sub>H<sub>28</sub>ClN<sub>3</sub>NaO<sub>4</sub> ([M+Na]<sup>+</sup>): 624.1661, found: 624.1660.

***trans*-1-Benzyl-5-chloro-1'-(4-methoxyphenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3i')**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.030 g, 5%, m.p. 219-221 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.35 (s, 1H), 7.30 (d, *J* = 8.0 Hz, 2H), 7.15-7.07 (m, 6H), 7.01 (d, *J* = 6.8 Hz, 2H), 6.82 (d, *J* = 7.6 Hz, 2H), 6.60 (d, *J* = 6.4 Hz, 2H), 6.44 (d, *J* = 8.8 Hz, 1H), 6.24 (d, *J* = 8.4 Hz, 1H), 5.39 (dd, *J*<sub>1</sub> = 9.2 Hz, *J*<sub>2</sub> = 5.2 Hz, 1H), 4.71 (d, *J* = 16.0 Hz, 1H), 4.46 (d, *J* = 16.0 Hz, 1H), 4.34 (s, 1H), 3.87 (s, 3H), 2.81 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 174.4, 168.9, 165.6, 159.3, 143.3, 140.6, 138.0, 136.6, 135.1, 134.9, 134.1, 129.1, 128.6, 128.4, 128.2, 127.8, 127.6, 127.4, 127.1, 126.5, 125.0, 115.7, 114.7, 110.2, 108.0, 57.2, 55.5, 49.3, 44.1, 23.6; IR(KBr) ν: 1766, 1709, 1639, 1508, 1435, 1381, 1335, 1319, 1241, 1180, 1119, 1081, 1016, 849, 798, 764, 758 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>36</sub>H<sub>28</sub>ClN<sub>3</sub>NaO<sub>4</sub> ([M+Na]<sup>+</sup>): 624.1661, found: 624.1666.

***cis*-1-Butyl-1'-(4-methoxyphenyl)-5,7'-dimethyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3j)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.394 g, 72%, m.p. 201-203 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.29 (d, *J* = 8.8 Hz, 2H), 7.04-6.94 (m, 7H), 6.89 (d, *J* = 11.6 Hz, 2H), 6.47 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 6.38 (d, *J* = 7.6 Hz, 1H), 5.58 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 3.6 Hz, 1H), 3.91 (dd, *J*<sub>1</sub> = 5.6 Hz, *J*<sub>2</sub> = 2.4 Hz, 1H), 3.68 (s, 3H), 3.61-3.54 (m, 1H), 3.46-3.39 (m, 1H), 2.73 (s, 3H), 2.32 (s, 3H), 1.54-1.47 (m, 2H), 1.34-1.29 (m, 2H), 0.92 (t, *J* = 7.2 Hz, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 176.2, 168.8, 166.0, 159.0, 140.9, 139.8, 138.6, 137.2, 134.7, 132.3, 131.2, 129.1, 128.4, 127.5, 127.4, 126.8, 124.0, 115.7, 114.5, 107.6, 107.1, 58.0, 55.5, 50.6, 40.2, 29.2, 23.5, 21.3, 20.2, 13.7; IR(KBr) ν: 2934, 1763, 1701, 1654, 1604, 1561, 1508, 1438, 1383, 1357, 1299, 1245, 1197, 1168, 1131, 1106, 1032, 1008, 835, 813, 772, 757 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>34</sub>H<sub>34</sub>N<sub>3</sub>O<sub>4</sub> ([M+H]<sup>+</sup>): 548.2544, found: 548.2566.

***cis*-1-Benzyl-1'-(4-(tert-butyl)phenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3k)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.498 g, 84%, m.p. 199-201 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.48 (d, *J* = 7.2 Hz, 2H), 7.28 (s, 3H), 7.25-7.15 (m, 5H), 7.05-6.96 (m, 7H), 6.56

(d,  $J = 7.2$  Hz, 1H), 6.35 (d,  $J = 5.6$  Hz, 1H), 4.96 (d,  $J = 16.0$  Hz, 1H), 4.56 (d,  $J = 16.0$  Hz, 1H), 4.00 (s, 1H), 2.76 (s, 3H), 1.38 (s, 9H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.6, 168.9, 165.9, 150.9, 141.9, 141.6, 141.0, 138.4, 135.5, 134.8, 132.1, 129.0, 128.6, 128.1, 127.6, 127.3, 127.2, 126.9, 126.4, 125.6, 123.1, 122.3, 115.9, 108.8, 107.4, 58.0, 50.7, 44.3, 34.7, 31.4, 23.6; IR(KBr)  $\nu$ : 1764, 1701, 1604, 1561, 1493, 1438, 1379, 1269, 1172, 839, 758  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{39}\text{H}_{36}\text{N}_3\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 594.2751, found: 594.2746.

***cis*-1-Benzyl-1'-(4-chlorophenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3l)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.423 g, 74%, m.p. 211-213  $^{\circ}\text{C}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.51-7.47 (m, 2H), 7.44-7.38 (m, 3H), 7.30-7.26 (m, 1H), 7.25-7.24 (m, 1H), 7.20-7.18 (m, 2H), 7.15-7.13 (m, 1H), 7.08-7.03 (m, 1H), 7.01-6.94 (m, 6H), 6.58 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 1.6$  Hz, 1H), 6.35 (dd,  $J_1 = 7.2$  Hz,  $J_2 = 2.4$  Hz, 1H), 5.64 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 5.2$  Hz, 1H), 4.96 (d,  $J = 16.0$  Hz, 1H), 4.55 (d,  $J = 16.0$  Hz, 1H), 4.01 (dd,  $J_1 = 5.2$  Hz,  $J_2 = 2.0$  Hz, 1H), 2.75 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.5, 168.8, 165.8, 144.2, 142.0, 141.0, 138.3, 135.5, 134.5, 132.0, 129.5, 129.0, 128.6, 128.2, 127.9, 127.7, 127.3, 127.3, 127.0, 126.2, 123.2, 122.3, 116.0, 108.9, 107.8, 58.0, 50.7, 44.3, 23.5; IR(KBr)  $\nu$ : 1764, 1703, 1609, 1490, 1467, 1453, 1435, 1380, 1353, 1263, 1173, 1134, 1074, 1031, 1010, 783, 774, 751  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{35}\text{H}_{26}\text{ClN}_3\text{NaO}_3$  ( $[\text{M}+\text{Na}]^+$ ): 594.1555, found: 594.1571.

***cis*-1-Benzyl-5-chloro-1'-(4-chlorophenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3m)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.563 g, 93%, m.p. 228-230  $^{\circ}\text{C}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.45 (d,  $J = 8.4$  Hz, 2H), 7.33-7.28 (m, 4H), 7.23 (d,  $J = 6.4$  Hz, 1H), 7.15 (d,  $J = 7.2$  Hz, 2H), 7.11 (s, 1H), 7.07 (d,  $J = 6.8$  Hz, 1H), 7.03 (t,  $J = 7.2$  Hz, 2H), 6.95 (d,  $J = 5.6$  Hz, 3H), 6.51 (d,  $J = 9.2$  Hz, 1H), 6.24 (d,  $J = 9.2$  Hz, 1H), 5.66 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 5.2$  Hz, 1H), 4.97 (d,  $J = 16.0$  Hz, 1H), 4.47 (d,  $J = 16.0$  Hz, 1H), 3.98 (d,  $J = 3.6$  Hz, 1H), 2.78 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.1, 168.6, 165.6, 142.5, 140.8, 140.6, 137.7, 134.9, 134.3, 133.8, 133.4, 129.8, 129.0, 128.7, 128.2, 127.9, 127.7, 127.6, 127.5, 127.3, 127.2, 123.6, 116.2, 109.9, 107.5, 57.9, 50.6, 44.4, 23.7; IR(KBr)  $\nu$ : 1707, 1650, 1616, 1486, 1447, 1387, 1331, 1268, 1167, 1120, 1100, 1080, 1013, 963, 842, 805, 756  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{35}\text{H}_{25}\text{Cl}_2\text{N}_3\text{NaO}_3$  ( $[\text{M}+\text{Na}]^+$ ): 628.1165, found: 628.1173.

**cis-1-Benzyl-5-chloro-1'-(3-chlorophenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (3n):** ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.502 g, 83%, m.p. 219-221 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.44-7.39 (m, 3H), 7.30-7.26 (m, 3H), 7.23 (d, *J* = 5.2 Hz, 1H), 7.16-7.14 (m, 2H), 7.12 (d, *J* = 2.0 Hz, 1H), 7.08 (d, *J* = 7.2 Hz, 1H), 7.03 (t, *J* = 7.6 Hz, 2H), 6.97-6.94 (m, 3H), 6.56 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 6.24 (d, *J* = 8.0 Hz, 1H), 5.66 (dd, *J*<sub>1</sub> = 9.2 Hz, *J*<sub>2</sub> = 5.6 Hz, 1H), 4.98 (d, *J* = 16.0 Hz, 1H), 4.47 (d, *J* = 16.0 Hz, 1H), 3.99-3.97 (m, 1H), 2.79 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 176.0, 168.6, 165.4, 144.9, 140.78, 140.6, 137.6, 135.0, 134.9, 134.2, 133.3, 130.5, 128.9, 128.7, 128.2, 127.9, 127.7, 127.5, 127.2, 127.1, 126.6, 124.4, 123.6, 116.4, 109.8, 108.0, 63.8, 57.8, 50.5, 44.3, 23.6; IR(KBr) ν: 1705, 1657, 1589, 1485, 1454, 1378, 1244, 1171, 990, 817, 758 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>35</sub>H<sub>26</sub>Cl<sub>2</sub>N<sub>3</sub>O<sub>3</sub> ([M+H]<sup>+</sup>): 606.1346, found: 606.1361.

**cis-1-Benzyl-1'-(4-bromophenyl)-5-chloro-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (3o):** ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.493 g, 76%, m.p. 234-236 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.61 (d, *J* = 8.8 Hz, 2H), 7.30-7.27 (m, 3H), 7.23 (d, *J* = 10.0 Hz, 2H), 7.14 (d, *J* = 6.8 Hz, 2H), 7.11 (s, 1H), 7.08 (d, *J* = 7.2 Hz, 1H), 7.03 (t, *J* = 7.6 Hz, 2H), 6.97-6.94 (m, 3H), 6.53 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 6.24 (d, *J* = 8.4 Hz, 1H), 5.65 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 5.2 Hz, 1H), 4.97 (d, *J* = 16.0 Hz, 1H), 4.47 (d, *J* = 16.0 Hz, 1H), 3.97 (dd, *J*<sub>1</sub> = 5.2 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 2.78 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 176.0, 168.6, 165.6, 143.0, 140.8, 140.5, 137.7, 134.9, 134.2, 133.3, 132.8, 128.9, 128.7, 128.1, 127.9, 127.8, 127.7, 127.5, 127.2, 127.1, 123.6, 121.7, 116.2, 107.6, 57.9, 50.6, 44.3, 23.6; IR(KBr) ν: 1760, 1712, 1612, 1485, 1454 1380, 1331, 1267, 1172, 1119, 1078, 1009, 839, 813, 757 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>35</sub>H<sub>26</sub>BrClN<sub>3</sub>NaO<sub>3</sub> ([M+Na]<sup>+</sup>): 672.0660, found: 672.0668.

**cis-1-Benzyl-1'-mesityl-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (3p):** ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.504 g, 87%, m.p. 223-225 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.29-7.27 (m, 2H), 7.23 (d, *J* = 5.2 Hz, 1H), 7.19 (d, *J* = 6.8 Hz, 2H), 7.09 (d, *J* = 7.2 Hz, 1H), 7.06-6.93 (m, 9H), 6.36 (d, *J* = 7.2 Hz, 1H), 6.12 (dd, *J*<sub>1</sub> = 9.2 Hz, *J*<sub>2</sub> = 2.4 Hz, 1H), 5.61 (dd, *J*<sub>1</sub> = 9.2 Hz, *J*<sub>2</sub> = 4.8 Hz, 1H), 4.94 (d, *J* = 16.0 Hz, 1H), 4.56 (d, *J* = 16.0 Hz, 1H), 3.97 (dd, *J*<sub>1</sub> = 5.2



Hz,  $J_2 = 2.4$  Hz, 1H), 2.76 (s, 3H), 2.35 (s, 3H), 2.32 (s, 3H), 2.26 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.4, 168.9, 165.5, 142.0, 141.0, 139.9, 139.6, 138.8, 138.1, 135.5, 134.9, 134.5, 132.1, 129.6, 129.2, 129.0, 128.6, 128.0, 127.6, 127.3, 126.8, 123.2, 122.1, 116.1, 105.5, 58.1, 51.3, 51.3, 44.2, 23.5, 21.1, 17.9, 17.7; IR(KBr)  $\nu$ : 1762, 1717, 1598, 1492, 1438, 1378, 1177, 1011, 851, 753  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{38}\text{H}_{33}\text{N}_3\text{NaO}_3$  ( $[\text{M}+\text{Na}]^+$ ): 602.2414, found: 602.2412.

**cis-1-Benzyl-4'-(4-methoxyphenyl)-7'-methyl-1'-(*p*-tolyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3q)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.523 g, 90%, m.p. 226-228 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.30-7.26 (m, 5H), 7.25-7.24 (m, 2H), 7.20-7.18 (m, 2H), 7.13-7.11 (m, 1H), 7.04-6.96 (m, 2H), 6.85 (d,  $J = 8.8$  Hz, 2H), 6.53-6.49 (m, 3H), 6.39 (d,  $J = 8.0$  Hz, 1H), 5.58 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 5.2$  Hz, 1H), 4.93 (d,  $J = 16.0$  Hz, 1H), 4.62 (d,  $J = 16.0$  Hz, 1H), 3.95 (d,  $J = 2.0$  Hz, 1H), 3.68 (s, 3H), 2.75 (s, 3H), 2.43 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.7, 168.9, 165.9, 158.4, 142.0, 141.8, 140.9, 137.8, 135.5, 134.3, 132.3, 130.5, 130.1, 130.0, 128.6, 128.1, 127.3, 127.3, 126.0, 123.1, 122.2, 116.1, 113.0, 108.9, 107.2, 58.1, 55.1, 49.8, 49.8, 44.2, 23.5, 21.2; IR(KBr)  $\nu$ : 1759, 1712, 1651, 1611, 1513, 1489, 1468, 1445, 1381, 1347, 1304, 1251, 1174, 1129, 1103, 1038, 1009, 822  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{37}\text{H}_{32}\text{N}_3\text{O}_4$  ( $[\text{M}+\text{H}]^+$ ): 582.2387, found: 582.2408.

**cis-1-Benzyl-5-chloro-4'-(4-methoxyphenyl)-7'-methyl-1'-(4-nitrophenyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3r)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.433 g, 67%, m.p. 234-236 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 8.34 (d,  $J = 8.8$  Hz, 2H), 7.49 (d,  $J = 8.8$  Hz, 2H), 7.30-7.27 (m, 3H), 7.14 (d,  $J = 8.4$  Hz, 3H), 7.00 (d,  $J = 9.6$  Hz, 1H), 6.86 (d,  $J = 8.4$  Hz, 2H), 6.74 (d,  $J = 8.4$  Hz, 1H), 6.55 (d,  $J = 8.4$  Hz, 2H), 6.27 (d,  $J = 8.4$  Hz, 1H), 5.80 (dd,  $J_1 = 8.4$  Hz,  $J_2 = 5.2$  Hz, 1H), 4.95 (d,  $J = 16.0$  Hz, 1H), 4.52 (d,  $J = 16.0$  Hz, 1H), 4.01 (d,  $J = 6.0$  Hz, 1H), 3.70 (s, 3H), 2.81 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 175.8, 168.5, 165.4, 142.9, 140.7, 140.5, 136.3, 134.8, 133.2, 133.1, 132.8, 130.3, 128.7, 128.4, 128.1, 127.8, 127.6, 127.3, 123.5, 121.6, 115.4, 110.1, 107.4, 61.5, 57.7, 55.7, 49.7, 29.7, 23.6; IR(KBr)  $\nu$ : 1705, 1590, 1516, 1488, 1434, 1341, 1251, 1177  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{36}\text{H}_{27}\text{ClN}_4\text{NaO}_6$  ( $[\text{M}+\text{Na}]^+$ ): 669.1511, found: 669.1541.

***cis*-1-Benzyl-5-chloro-1'-(2,6-dimethylphenyl)-4'-(4-methoxyphenyl)-7'-methyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3s):** ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.554 g, 88%, m.p. 249-251 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.31-7.27 (m, 3H), 7.24 (s, 1H), 7.19-7.16 (m, 4H), 7.06 (d, *J* = 2.0 Hz, 1H), 6.98 (dd, *J*<sub>1</sub> = 8.4 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 6.86 (d, *J* = 8.4 Hz, 2H), 6.53 (d, *J* = 8.8 Hz, 2H), 6.29 (d, *J* = 8.4 Hz, 1H), 6.12 (dd, *J*<sub>1</sub> = 9.2 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 5.57 (dd, *J*<sub>1</sub> = 9.2 Hz, *J*<sub>2</sub> = 4.4 Hz, 1H), 4.93 (d, *J* = 16.0 Hz, 1H), 4.55 (d, *J* = 16.0 Hz, 1H), 3.91 (dd, *J*<sub>1</sub> = 4.8 Hz, *J*<sub>2</sub> = 1.6 Hz, 1H), 3.70 (s, 3H), 2.78 (s, 3H), 2.35 (s, 3H), 2.29 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 176.1, 168.9, 165.3, 158.5, 141.9, 140.9, 140.6, 135.3, 135.1, 134.8, 133.8, 130.4, 130.0, 128.9, 128.7, 128.5, 127.9, 127.6, 127.4, 127.3, 123.5, 116.3, 113.1, 109.9, 104.7, 58.1, 55.1, 55.1, 50.3, 44.3, 23.6, 17.9, 17.9; IR(KBr) ν: 1709, 1610, 1515, 1483, 1380, 1255, 1170 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>38</sub>H<sub>32</sub>ClN<sub>3</sub>NaO<sub>4</sub> ([M+Na]<sup>+</sup>): 652.1974, found: 652.1993.

***cis*-1-Benzyl-1'-(4-bromophenyl)-5-chloro-4'-(4-chlorophenyl)-7'-methyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3t):** ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.546 g, 80%, m.p. 262-264 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.60 (d, *J* = 6.8 Hz, 2H), 7.32-7.29 (m, 4H), 7.23 (s, 1H), 7.13 (d, *J* = 7.6 Hz, 2H), 7.10 (s, 1H), 7.02 (d, *J* = 8.0 Hz, 1H), 6.96 (d, *J* = 6.8 Hz, 2H), 6.86 (d, *J* = 6.8 Hz, 2H), 6.51 (d, *J* = 9.2 Hz, 1H), 6.34 (d, *J* = 8.4 Hz, 1H), 5.55-5.51 (m, 1H), 4.90 (d, *J* = 16.0 Hz, 1H), 4.58 (d, *J* = 16.0 Hz, 1H), 3.95-3.94 (m, 1H), 2.78 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 175.8, 168.5, 165.4, 142.9, 140.7, 140.5, 136.3, 134.8, 134.4, 133.2, 133.1, 132.8, 130.2, 128.7, 128.4, 128.1, 127.8, 127.6, 127.3, 123.5, 121.9, 115.4, 110.1, 107.4, 61.5, 57.7, 49.7, 23.6; IR(KBr) ν: 3726, 1709, 1611, 1484, 1445, 1379, 1337 1171, 1009, 814 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>35</sub>H<sub>25</sub>BrCl<sub>2</sub>N<sub>3</sub>O<sub>3</sub> ([M+H]<sup>+</sup>): 684.0451, found: 684.0447.

***cis*-1-Benzyl-5-chloro-4'-(4-chlorophenyl)-7'-methyl-1'-(*o*-tolyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3u):** ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.551 g, 89%, m.p. 247-249 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.40-7.27 (m, 7H), 7.15 (d, *J* = 8.0 Hz, 2H), 7.07 (s, 1H), 7.02 (dd, *J*<sub>1</sub> = 8.4 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 6.97 (d, *J* = 8.0 Hz, 2H), 6.87 (d, *J* = 8.4 Hz, 2H), 6.36 (d, *J* = 8.4 Hz, 1H), 6.25 (d, *J* = 8.8 Hz, 1H), 5.49 (s, 1H), 4.90 (d, *J* = 16.0 Hz, 1H), 4.60 (d, *J* = 16.0 Hz, 1H), 3.94 (s, 1H), 2.77 (s, 3H), 2.38 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 175.8, 168.7, 165.3,

140.5, 134.9, 133.4, 133.1, 130.3, 128.7, 128.3, 128.0, 127.8, 127.6, 127.3, 123.5, 114.8, 111.0, 105.1, 57.9, 55.4, 50.2, 44.4, 23.6, 17.7; IR(KBr)  $\nu$ : 1712, 1615, 1489, 1439, 1380, 1337, 1166, 1014, 836, 807, 742  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{36}\text{H}_{28}\text{Cl}_2\text{N}_3\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 620.1502, found: 620.1508.

***cis*-1-Benzyl-5-chloro-4'-(4-methoxyphenyl)-7'-methyl-1'-(naphthalen-1-yl)-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'H)-trione (3v)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.534 g, 82%, m.p. 230-232 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.97-7.92 (m, 3H), 7.66-7.56 (m, 4H), 7.32-7.27 (m, 3H), 7.20-7.15 (m, 3H), 7.02-6.98 (m, 1H), 6.94-6.89 (m, 2H), 6.58-6.43 (m, 3H), 6.30 (d,  $J = 7.6$  Hz, 1H), 5.57 (s, 1H), 4.98 (d,  $J = 16.0$  Hz, 1H), 4.58 (d,  $J = 16.0$  Hz, 1H), 4.07 (s, 1H), 3.70 (s, 3H), 2.69 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (150 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.3, 168.9, 168.8, 165.2, 158.6, 141.7, 140.7, 140.5, 135.1, 134.7, 134.6, 133.7, 130.3, 130.1, 129.2, 128.9, 128.7, 127.9, 127.8, 127.7, 127.5, 127.3, 126.7, 125.8, 125.4, 125.2, 124.2, 123.6, 122.3, 121.3, 115.6, 115.2, 113.2, 109.9, 58.2, 55.2, 50.3, 44.4, 23.6; IR(KBr)  $\nu$ : 1703, 1594, 1513, 1485, 1440, 1381, 1328, 1244, 1168, 810, 778  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{40}\text{H}_{31}\text{ClN}_3\text{O}_4$  ( $[\text{M}+\text{H}]^+$ ): 652.1998, found: 652.1980.

***cis*-1-benzyl-5-chloro-4'-(2-methoxyphenyl)-7'-methyl-1'-(thiazol-2-yl)-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'H)-trione (3w)**: ethyl acetate, dichloromethane and petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.498 g, 82%, m.p. 229-231 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 8.34 (d,  $J = 7.6$  Hz, 1H), 7.20 (d,  $J = 7.2$  Hz, 3H), 7.13-7.09 (m, 4H), 6.98-6.89 (m, 3H), 6.84-6.79 (m, 2H), 6.52 (d,  $J = 8.4$  Hz, 1H), 6.37 (d,  $J = 7.2$  Hz, 1H), 5.84 (dd,  $J_1 = 16.0$  Hz,  $J_2 = 8.0$  Hz, 1H), 5.02 (d,  $J = 16.0$  Hz, 1H), 4.71 (d,  $J = 16.0$  Hz, 1H), 4.57 (d,  $J = 8.0$  Hz, 1H), 3.66 (s, 3H), 2.98 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 172.8, 167.3, 165.6, 157.3, 157.0, 143.0, 135.6, 134.9, 131.8, 129.7, 128.9, 128.7, 128.6, 127.8, 127.4, 127.3, 127.1, 125.7, 125.5, 125.3, 122.5, 120.5, 115.4, 110.9, 110.2, 106.8, 65.6, 58.2, 55.3, 44.6, 24.2; IR(KBr)  $\nu$ : 3121, 2834, 1709, 1656, 1612, 1486, 1455, 1369, 1341, 1247, 1215, 1194, 1085, 977, 831, 759  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{33}\text{H}_{25}\text{ClN}_4\text{NaO}_4\text{S}$  ( $[\text{M}+\text{Na}]^+$ ): 631.1177, found: 631.1151.

***cis*-1'-(benzo[*d*]thiazol-2-yl)-1-benzyl-5-chloro-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'H)-trione (3x)**: ethyl acetate, dichloromethane and

petroleum ether (V/V/V = 1:10:10) as eluent, yellow solid, 0.458 g, 73%, m.p. 214-216 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.92 (d, *J* = 8.0 Hz, 1H), 7.82 (d, *J* = 8.4 Hz, 1H), 7.49 (d, *J* = 8.0 Hz, 1H), 7.37 (d, *J* = 8.0 Hz, 1H), 7.30-7.27 (m, 3H), 7.14-6.96 (m, 10H), 6.21 (d, *J* = 8.4 Hz, 1H), 6.11 (t, *J* = 7.2 Hz, 1H), 4.98 (d, *J* = 16.0 Hz, 1H), 4.44 (d, *J* = 16.0 Hz, 1H), 4.26 (d, *J* = 5.2 Hz, 1H), 2.89 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 175.2, 168.2, 164.6, 161.8, 150.2, 141.3, 140.9, 136.0, 134.7, 133.5, 133.1, 132.3, 128.9, 128.7, 128.0, 127.9, 127.6, 127.1, 126.7, 125.0, 123.7, 122.5, 122.5, 121.5, 118.8, 110.1, 56.4, 49.2, 44.4, 23.9; IR(KBr) ν: 3448, 2925, 2854, 1711, 1637, 1439, 1376, 1020 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>36</sub>H<sub>25</sub>ClN<sub>4</sub>NaO<sub>3</sub>S ([M+Na]<sup>+</sup>): 651.1218, found: 651.1190.

**2. General procedure for the reactions of  $\alpha,\beta$ -unsaturated *N*-benzylaldimines and MBH maleimides of isatins:** A mixture of  $\alpha,\beta$ -unsaturated *N*-benzylaldimine (0.10 mmol), MBH maleimide of isatin (0.10 mmol) and DABCO (20 mmol%) in CH<sub>3</sub>CN (10.0 mL) was stirred at 50 °C for 8 hours. After removing the solvent by rotatory evaporation, the resulting residue was purified by column chromatography on silica gel with ethyl acetate and petroleum ether (V/V = 1:8) as eluent to afford the desired products **4a-4k** and minor isomers **4c'**, **4e'** and **4h'**.

***cis*-1,1'-Dibenzyl-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4a):** ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.474 g, 86%, m.p. 178-180 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.42-7.27 (m, 7H), 7.25-7.22 (m, 1H), 7.18 (d, *J* = 7.6 Hz, 2H), 7.05-6.92 (m, 6H), 6.86 (d, *J* = 7.2 Hz, 2H), 6.33-6.30 (m, 1H), 6.31 (d, *J* = 7.6 Hz, 1H), 5.86 (d, *J* = 15.6 Hz, 1H), 5.71 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 3.2 Hz, 1H), 5.10 (d, *J* = 15.6 Hz, 1H), 4.96 (d, *J* = 16.0 Hz, 1H), 4.51 (d, *J* = 16.0 Hz, 1H), 3.78 (dd, *J*<sub>1</sub> = 5.6 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 2.85 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 176.8, 168.6, 167.2, 141.8, 141.3, 138.1, 137.6, 135.5, 134.3, 132.3, 128.9, 128.9, 128.6, 128.0, 127.7, 127.6, 127.2, 127.2, 126.9, 122.9, 122.2, 117.9, 108.7, 106.3, 58.6, 55.2, 50.6, 44.1, 23.6; IR(KBr) ν: 3726, 1697, 1576, 1489, 1442, 1384, 1328, 1254, 879 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>36</sub>H<sub>30</sub>N<sub>3</sub>O<sub>3</sub> ([M+H]<sup>+</sup>): 552.2282, found: 552.2273.

***cis*-1,1'-Dibenzyl-5,7'-dimethyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4b):** ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.412 g, 73%, m.p. 190-192 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.42-7.31 (m, 6H), 7.24-7.21 (m, 2H), 7.17 (d, *J* = 7.2 Hz, 2H), 7.04 (t, *J* = 7.2 Hz, 1H), 6.96 (t, *J* = 7.6 Hz, 2H), 6.87

(d,  $J = 7.6$  Hz, 2H), 6.81 (s, 1H), 6.75 (d,  $J = 8.0$  Hz, 1H), 6.30 (d,  $J = 8.8$  Hz, 1H), 6.18 (d,  $J = 8.0$  Hz, 1H), 5.88 (d,  $J = 15.6$  Hz, 1H), 5.70 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 3.2$  Hz, 1H), 5.08 (d,  $J = 15.6$  Hz, 1H), 4.95 (d,  $J = 16.0$  Hz, 1H), 4.47 (d,  $J = 16.0$  Hz, 1H), 3.75 (d,  $J = 7.2$  Hz, 1H), 2.85 (s, 3H), 2.25 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.7, 168.7, 167.3, 141.2, 139.5, 138.3, 137.6, 135.6, 134.3, 132.4, 131.6, 129.0, 128.8, 128.5, 128.4, 127.7, 127.6, 127.3, 127.2, 126.8, 123.7, 118.0, 116.6, 115.9, 108.9, 108.4, 108.3, 106.5, 58.6, 55.2, 50.6, 44.1, 23.6, 21.2; IR(KBr)  $\nu$ : 1698, 1597, 1489, 1454, 753  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{37}\text{H}_{31}\text{N}_3\text{NaO}_3$  ( $[\text{M}+\text{Na}]^+$ ): 588.2258, found: 588.2253.

***cis*-1,1'-Dibenzyl-5-chloro-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4c)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.379 g, 65%, m.p. 194-196  $^\circ\text{C}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.40-7.27 (m, 7H), 7.23 (s, 1H), 7.15 (d,  $J = 5.6$  Hz, 2H), 7.06 (d,  $J = 5.2$  Hz, 1H), 7.01-6.99 (m, 3H), 6.93-6.87 (m, 3H), 6.32 (d,  $J = 8.8$  Hz, 1H), 6.20 (d,  $J = 7.6$  Hz, 1H), 5.86 (d,  $J = 15.6$  Hz, 1H), 5.67 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 3.2$  Hz, 1H), 5.10 (d,  $J = 15.6$  Hz, 1H), 4.97 (d,  $J = 16.0$  Hz, 1H), 4.45 (d,  $J = 16.0$  Hz, 1H), 3.74 (s, 1H), 2.87 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.4, 168.6, 167.0, 141.3, 140.4, 137.7, 137.3, 135.0, 134.4, 133.9, 128.9, 128.8, 128.7, 127.9, 127.8, 127.7, 127.6, 127.4, 127.3, 127.1, 127.0, 123.3, 117.7, 109.7, 105.4, 58.5, 55.2, 50.5, 44.25, 23.7; IR(KBr)  $\nu$ : 1704, 1694, 1669, 1595, 1488, 1454, 1423, 1387, 1331, 1259, 1162, 1019, 996, 961, 818, 755  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{36}\text{H}_{29}\text{ClN}_3\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 586.1892, found: 586.1883.

***trans*-1,1'-Dibenzyl-5-chloro-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4c')**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.105 g, 18%, m.p. 203-205  $^\circ\text{C}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.43-7.34 (m, 5H), 7.26-7.22 (m, 2H), 7.14-7.04 (m, 6H), 6.76 (d,  $J = 7.6$  Hz, 2H), 6.57 (d,  $J = 6.8$  Hz, 2H), 6.29 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 2.0$  Hz, 1H), 6.21 (d,  $J = 9.2$  Hz, 1H), 5.81 (d,  $J = 15.6$  Hz, 1H), 5.47 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 5.2$  Hz, 1H), 5.20 (d,  $J = 15.6$  Hz, 1H), 4.67 (d,  $J = 16.4$  Hz, 1H), 4.43 (d,  $J = 16.4$  Hz, 1H), 4.15 (dd,  $J_1 = 5.2$  Hz,  $J_2 = 2.4$  Hz, 1H), 2.92 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 174.4, 168.7, 167.0, 143.3, 140.9, 137.8, 137.1, 135.0, 134.2, 129.0, 128.5, 128.3, 128.2, 127.9, 127.6, 127.3, 127.2, 127.1, 126.5, 125.1, 117.9, 110.2, 107.4, 57.3, 55.1, 49.2, 44.1, 23.8; IR(KBr)  $\nu$ : 1701, 1695, 1668, 1591, 1489, 1451, 1420, 1381, 1330, 1261, 1160, 1018, 995, 960, 817, 756  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{36}\text{H}_{29}\text{ClN}_3\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 586.1892, found:

586.1885.

***cis*-1-Benzyl-7'-methyl-1'-(4-methylbenzyl)-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4d)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.480 g, 85%, m.p. 197-199 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.28-7.27 (m, 2H), 7.25-7.17 (m, 7H), 7.05-6.90 (m, 6H), 6.87 (d, *J* = 7.6 Hz, 2H), 6.31 (d, *J* = 8.8 Hz, 1H), 6.30 (d, *J* = 8.8 Hz, 1H), 5.83 (d, *J* = 15.2 Hz, 1H), 5.69 (dd, *J*<sub>1</sub> = 9.2 Hz, *J*<sub>2</sub> = 3.2 Hz, 1H), 5.03 (d, *J* = 15.2 Hz, 1H), 4.95 (d, *J* = 16.0 Hz, 1H), 4.51 (d, *J* = 16.0 Hz, 1H), 3.78 (dd, *J*<sub>1</sub> = 5.6 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 2.84 (s, 3H), 2.36 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 176.8, 168.7, 167.3, 141.9, 141.3, 138.2, 137.5, 135.5, 134.5, 132.4, 129.5, 128.9, 128.6, 128.0, 127.6, 127.3, 127.2, 126.9, 122.9, 122.2, 117.9, 108.7, 106.2, 58.6, 55.0, 50.6, 44.2, 23.6, 21.2; IR(KBr) ν: 1701, 1697, 1595, 1488, 1481, 1331, 1264, 1150, 1018, 752 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>37</sub>H<sub>32</sub>N<sub>3</sub>O<sub>3</sub> ([M+H]<sup>+</sup>): 566.2438, found: 566.2439.

***cis*-1-Benzyl-5-chloro-7'-methyl-1'-(4-methylbenzyl)-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4e)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.450 g, 75%, m.p. 198-200 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.29-7.26 (m, 2H), 7.25-7.19 (m, 5H), 7.15 (d, *J* = 6.8 Hz, 2H), 7.06 (t, *J* = 7.2 Hz, 1H), 7.01-6.98 (m, 3H), 6.91 (dd, *J*<sub>1</sub> = 8.4 Hz, *J*<sub>2</sub> = 2.4 Hz, 1H), 6.88 (d, *J* = 7.2 Hz, 2H), 6.31 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 6.19 (d, *J* = 8.4 Hz, 1H), 6.19 (d, *J* = 8.4 Hz, 1H), 5.86 (d, *J* = 15.6 Hz, 1H), 5.67 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 3.2 Hz, 1H), 5.02 (d, *J* = 15.6 Hz, 1H), 4.96 (d, *J* = 16.0 Hz, 1H), 4.45 (d, *J* = 16.0 Hz, 1H), 3.72 (dd, *J*<sub>1</sub> = 5.6 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 2.36 (s, 3H), 2.87 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 176.4, 168.6, 167.0, 141.4, 140.4, 137.8, 137.6, 135.1, 134.4, 134.3, 134.0, 129.6, 128.9, 128.7, 127.9, 127.8, 127.6, 127.4, 127.3, 127.1, 123.4, 117.6, 109.7, 105.3, 58.5, 55.0, 50.5, 44.3, 23.7, 21.2; IR(KBr) ν: 3786, 3726, 3703, 3629, 1753, 1735, 1696, 1592, 1561, 1554, 1513, 1484, 1438, 1387, 1332, 1258, 1157, 1130, 995, 817, 756 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>37</sub>H<sub>31</sub>ClN<sub>3</sub>O<sub>3</sub> ([M+H]<sup>+</sup>): 600.2048, found: 600.2038.

***trans*-1-Benzyl-5-chloro-7'-methyl-1'-(4-methylbenzyl)-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4e')**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.060 g, 10%, m.p. 211-213 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.25-7.20 (m, 6H), 7.16-7.08 (m, 5H), 7.05 (dd, *J*<sub>1</sub> = 8.4 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 6.75 (d, *J* = 7.6 Hz, 2H), 6.57 (dd, *J*<sub>1</sub> = 7.6 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 6.29 (dd, *J*<sub>1</sub> = 8.4 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 6.20 (d, *J* =

8.4 Hz, 1H), 5.74 (d,  $J = 15.6$  Hz, 1H), 5.45 (dd,  $J_1 = 9.2$  Hz,  $J_2 = 3.6$  Hz, 1H), 5.17 (d,  $J = 15.6$  Hz, 1H), 4.67 (d,  $J = 16.0$  Hz, 1H), 4.43 (d,  $J = 16.0$  Hz, 1H), 4.14 (dd,  $J_1 = 5.6$  Hz,  $J_2 = 2.4$  Hz, 1H), 2.91 (s, 3H), 2.37 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 174.4, 168.7, 167.0, 143.2, 141.0, 137.8, 137.7, 135.0, 134.3, 134.1, 129.6, 129.0, 128.5, 128.3, 128.1, 127.5, 127.3, 127.2, 127.1, 126.5, 125.1, 117.8, 107.3, 57.5, 49.2, 44.1, 29.7, 21.1; IR(KBr)  $\nu$ : 3788, 3726, 3703, 3609, 1751, 1736, 1699, 1592, 1560, 1552, 1511, 1482, 1436, 1388, 1131, 995, 816, 754  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{37}\text{H}_{31}\text{ClN}_3\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 600.2048, found: 600.2040.

***cis*-1-Butyl-5-chloro-7'-methyl-1'-(4-methylbenzyl)-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indolin e-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4f)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.497 g, 88%, m.p. 206-208 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.24 (d,  $J = 8.0$  Hz, 2H), 7.20 (d,  $J = 8.0$  Hz, 2H), 7.05-7.01 (m, 4H), 6.95 (d,  $J = 2.0$  Hz, 1H), 6.89-6.87 (m, 2H), 6.38 (d,  $J = 8.4$  Hz, 1H), 6.30 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 2.0$  Hz, 1H), 5.86 (d,  $J = 15.2$  Hz, 1H), 5.66 (dd,  $J_1 = 8.4$  Hz,  $J_2 = 5.2$  Hz, 1H), 5.08 (d,  $J = 15.2$  Hz, 1H), 3.68 (dd,  $J_1 = 5.2$  Hz,  $J_2 = 2.0$  Hz, 1H), 3.60-3.53 (m, 1H), 3.42-3.35 (m, 1H), 2.83 (s, 3H), 2.36 (s, 3H), 1.51-1.46 (m, 2H), 1.33-1.25 (m, 2H), 0.91 (t,  $J = 7.2$  Hz, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.1, 168.5, 167.1, 141.4, 140.8, 137.8, 137.5, 134.4, 134.3, 134.0, 129.6, 128.9, 127.9, 127.6, 127.3, 127.2, 127.0, 123.4, 117.6, 108.6, 105.4, 58.3, 55.0, 50.5, 40.3, 29.1, 23.6, 21.2, 20.0, 13.7; IR(KBr)  $\nu$ : 1715, 1696, 1591, 1487, 1423, 1340, 1257, 820, 756  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{34}\text{H}_{33}\text{ClN}_3\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 566.2205, found: 566.2203.

***cis*-1-Benzyl-1'-(4-methoxybenzyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4g)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.523 g, 90%, m.p. 161-163 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.32-7.28 (m, 3H), 7.25-7.17 (m, 4H), 7.05-6.86 (m, 10H), 6.32 (m, 2H), 5.79 (d,  $J = 15.2$  Hz, 1H), 5.70 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 5.2$  Hz, 1H), 5.02 (d,  $J = 15.2$  Hz, 1H), 4.95 (d,  $J = 16.0$  Hz, 1H), 4.51 (d,  $J = 16.0$  Hz, 1H), 3.82 (s, 3H), 3.76 (d,  $J = 5.2$  Hz, 1H), 2.85 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 176.8, 168.7, 167.3, 159.2, 141.8, 141.3, 138.2, 135.5, 134.1, 132.4, 129.6, 128.9, 128.8, 128.6, 128.0, 127.6, 127.2, 127.1, 126.9, 122.9, 122.1, 117.9, 114.2, 108.7, 106.2, 58.6, 55.3, 54.6, 50.5, 44.2, 23.6; IR(KBr)  $\nu$ : 1753, 1700, 1654, 1610, 1512, 1487, 1454, 1386, 1357, 1301, 1248, 1175, 1029, 993, 812  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{37}\text{H}_{32}\text{N}_3\text{O}_4$  ( $[\text{M}+\text{H}]^+$ ): 582.2387, found: 582.2374.

**cis-1-Benzyl-5-chloro-1'-(4-methoxybenzyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (4h)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.461 g, 75%, m.p. 160-162 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.30 (d, *J* = 8.4 Hz, 2H), 7.26-7.21 (m, 3H), 7.15 (d, *J* = 6.8 Hz, 2H), 7.05 (d, *J* = 7.2 Hz, 1H), 7.01-6.97 (m, 3H), 6.94-6.87 (m, 5H), 6.31 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 6.19 (d, *J* = 8.0 Hz, 1H), 5.80 (d, *J* = 15.2 Hz, 1H), 5.68 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 5.2 Hz, 1H), 5.01 (d, *J* = 15.2 Hz, 1H), 4.96 (d, *J* = 16.0 Hz, 1H), 4.45 (d, *J* = 16.0 Hz, 1H), 3.82 (s, 3H), 3.71 (dd, *J*<sub>1</sub> = 5.2 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 2.87 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 176.4, 168.6, 167.1, 159.3, 141.4, 140.4, 137.8, 135.0, 133.9, 129.4, 128.9, 128.8, 128.7, 127.9, 127.8, 127.6, 127.4, 127.1, 123.4, 117.6, 114.3, 109.6, 105.3, 58.5, 55.3, 55.3, 54.6, 50.5, 44.2, 23.7; IR(KBr) ν: 1751, 1706, 1652, 1611, 1512, 1488, 1456 1385, 1356, 1300, 1247, 1172, 1028, 991, 812, 754 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>37</sub>H<sub>30</sub>ClN<sub>3</sub>NaO<sub>4</sub> ([M+Na]<sup>+</sup>): 638.1817, found: 638.1842.

**trans-1-Benzyl-5-chloro-1'-(4-methoxybenzyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (4h')**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.068 g, 11%, m.p. 183-185 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.29 (d, *J* = 8.4 Hz, 2H), 7.24-7.23 (m, 2H), 7.14-7.08 (m, 4H), 7.06-7.03 (m, 1H), 6.93 (d, *J* = 8.4 Hz, 2H), 6.75 (d, *J* = 7.6 Hz, 2H), 6.56 (d, *J* = 7.4 Hz, 2H), 6.30 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 2.0 Hz, 1H), 6.20 (d, *J* = 8.4 Hz, 1H), 5.74 (d, *J* = 15.2 Hz, 1H), 5.45 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 5.2 Hz, 1H), 5.13 (d, *J* = 15.2 Hz, 1H), 4.77 (s, 1H), 4.67 (d, *J* = 15.6 Hz, 1H), 4.42 (d, *J* = 15.6 Hz, 1H), 4.11 (dd, *J*<sub>1</sub> = 5.6 Hz, *J*<sub>2</sub> = 2.4 Hz, 1H), 3.83 (s, 3H), 2.92 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 174.5, 168.7, 167.1, 159.4, 143.2, 141.0, 137.8, 135.0, 134.3, 134.1, 129.1, 129.0, 128.7, 128.5, 128.3, 128.1, 127.5, 127.3, 127.1, 126.8, 126.5, 125.0, 117.9, 114.4, 110.1, 108.0, 107.3, 102.8, 57.5, 55.3, 54.6, 49.2, 44.1, 29.7, 23.8; IR(KBr) ν: 1698, 1604, 1513, 1485, 1454, 1388, 1249, 1175, 1010, 982, 971, 861, 758 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>37</sub>H<sub>30</sub>ClN<sub>3</sub>NaO<sub>4</sub> ([M+Na]<sup>+</sup>): 638.1817, found: 638.1836.

**cis-1-Benzyl-1'-(3,4-dimethoxyphenethyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (4i)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.575 g, 92%, m.p. 160-162 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.25-7.19 (m, 3H), 7.14 (d, *J* = 6.4 Hz, 2H), 7.04 (t, *J* = 7.6 Hz, 1H), 6.98-6.89 (m, 4H), 6.86-6.83 (m, 5H), 6.76 (d, *J* = 6.8 Hz, 1H), 6.28-6.24 (m, 2H), 5.72 (dd, *J*<sub>1</sub> = 8.8 Hz, *J*<sub>2</sub> = 5.2 Hz, 1H),



5.23-5.16 (m, 1H), 4.93 (d,  $J = 16.0$  Hz, 1H), 4.46 (d,  $J = 16.0$  Hz, 1H), 3.93-3.88 (m, 1H), 3.91 (s, 3H), 3.86 (s, 3H), 3.52 (dd,  $J_1 = 5.2$  Hz,  $J_2 = 2.0$  Hz, 1H), 3.09-3.04 (m, 1H), 3.00-2.94 (m, 1H), 2.83 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 177.1, 168.6, 167.5, 149.1, 147.9, 141.7, 141.5, 138.0, 135.5, 133.8, 132.2, 130.6, 128.9, 128.5, 128.0, 127.6, 127.2, 127.1, 126.9, 122.7, 122.1, 121.2, 118.6, 112.4, 111.2, 108.6, 105.4, 67.4, 58.7, 55.9, 55.8, 52.4, 49.9, 44.1, 36.1, 23.5; IR(KBr)  $\nu$ : 1752, 1735, 1701, 1654, 1577, 1561, 1513, 1438, 1261, 750  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{39}\text{H}_{36}\text{N}_3\text{O}_5$  ( $[\text{M}+\text{H}]^+$ ): 626.2649, found: 626.2644.

***cis*-1-Benzyl-1'-butyl-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4j)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.481 g, 93%, m.p. 162-164 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.28 (s, 1H), 7.25-7.21 (m, 2H), 7.17 (d,  $J = 7.2$  Hz, 2H), 7.06-6.89 (m, 8H), 6.30 (d,  $J = 7.2$  Hz, 1H), 6.26 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 3.2$  Hz, 1H), 5.69 (dd,  $J_1 = 8.8$  Hz,  $J_2 = 5.2$  Hz, 1H), 4.95 (d,  $J = 16.0$  Hz, 1H), 4.65-6.58 (m, 1H), 4.51 (d,  $J = 16.0$  Hz, 1H), 3.82-3.75 (m, 2H), 2.85 (s, 3H), 1.81-1.73 (m, 2H), 1.48-1.43 (m, 2H), 1.01 (t,  $J = 7.2$  Hz, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 177.0, 168.7, 167.2, 141.8, 141.4, 138.3, 135.6, 134.0, 132.5, 128.9, 128.6, 128.0, 127.6, 127.2, 126.9, 122.9, 122.2, 117.5, 108.7, 105.1, 58.7, 52.4, 50.5, 44.2, 32.5, 23.6, 20.0, 13.9; IR(KBr)  $\nu$ : 1723, 1695, 1656, 1613, 1483, 1466, 1428, 1410, 1381, 1357, 1340, 1303, 1265, 1204, 1078, 983, 762  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{33}\text{H}_{32}\text{N}_3\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 518.2438, found: 518.2435.

***cis*-1,1'-Dibenzyl-7'-methyl-4'-(*p*-tolyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4k)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, yellow solid, 0.429 g, 76%, m.p. 200-202 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.45-7.28 (m, 10H), 7.20-7.05 (m, 5H), 6.96-6.839 (m, 3H), 6.69 (d,  $J = 7.6$  Hz, 1H), 6.12 (s, 1H), 5.58 (d,  $J = 15.2$  Hz, 1H), 5.46 (d,  $J = 15.2$  Hz, 1H), 5.04 (d,  $J = 15.6$  Hz, 1H), 4.83 (d,  $J = 15.6$  Hz, 1H), 3.24 (s, 1H), 2.89 (s, 3H), 1.68 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (150 MHz,  $\text{CDCl}_3$ )  $\delta$ : 174.5, 168.7, 167.3, 153.3, 152.7, 142.3, 139.8, 139.6, 137.2, 136.3, 135.1, 129.2, 128.9, 128.8, 128.7, 128.6, 128.1, 128.0, 127.9, 127.7, 127.6, 127.4, 127.2, 126.2, 123.6, 122.1, 109.0, 105.0, 58.1, 55.6, 53.5, 44.2, 29.7, 25.8, 23.8; IR(KBr)  $\nu$ : 1714, 1693, 1650, 1592, 1485, 1463, 1350, 1262, 1224, 1098, 1070, 758  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{37}\text{H}_{32}\text{N}_3\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 566.2438, found: 566.2432.

**3. General procedure for the reactions of  $\alpha,\beta$ -unsaturated *N*-aryldimine and MBH esters/nitriles of isatins**: A mixture of  $\alpha,\beta$ -unsaturated *N*-aryldimine (0.40 mmol), MBH esters

or nitrile of isatin (0.40 mmol) in CH<sub>3</sub>CN (10.0 mL) was stirred at 80 °C for 72 hours. After removing the solvent by rotatory evaporation, the resulting residue was purified by column chromatography on silica gel with ethyl acetate and petroleum ether (V/V = 1:8) as eluent to afford the desired products **6a-6n**.

**Methyl-*cis*-1-benzyl-2-oxo-1'-phenyl-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6a):** ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.266 g, 52%, m.p. 197-199 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 8.05 (s, 1H), 7.32-7.28 (m, 7H), 7.18-7.00 (m, 9H), 6.85 (t, *J* = 7.6 Hz, 2H), 6.62-6.51 (m, 2H), 6.38 (t, *J* = 16.4 Hz, 1H), 5.16 (d, *J* = 8.0 Hz, 1H), 5.14 (d, *J* = 15.6 Hz, 1H), 4.58 (d, *J* = 15.6 Hz, 1H), 3.54 (s, 3H); <sup>13</sup>C {<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 175.1, 164.5, 147.4, 143.1, 140.6, 135.7, 134.4, 132.8, 129.4, 128.7, 128.6, 128.4, 128.3, 127.2, 127.1, 127.0, 125.6, 123.1, 122.9, 122.8, 117.3, 109.0, 107.2, 74.6, 62.2, 50.9, 50.8, 43.9; IR(KBr) ν: 1713, 1692, 1650, 1585, 1483, 1462, 1351, 1269, 1224, 1098, 1070, 754 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>34</sub>H<sub>29</sub>N<sub>2</sub>O<sub>3</sub> ([M+H]<sup>+</sup>): 513.2173, found: 513.2172.

**Methyl-*cis*-1-benzyl-5-chloro-2-oxo-1'-phenyl-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6b):** ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.289 g, 53%, m.p. 201-203 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 8.03 (s, 1H), 7.31-7.27 (m, 8H), 7.13-7.01 (m, 7H), 6.86 (t, *J* = 7.6 Hz, 2H), 6.55-6.48 (m, 2H), 6.41 (d, *J* = 16.4 Hz, 1H), 5.14 (d, *J* = 7.6 Hz, 1H), 5.12 (d, *J* = 16.0 Hz, 1H), 4.55 (d, *J* = 16.0 Hz, 1H), 3.57 (s, 3H); <sup>13</sup>C {<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 174.8, 164.4, 147.7, 141.7, 140.3, 135.5, 135.2, 134.7, 134.5, 129.5, 128.7, 128.6, 128.5, 128.4, 128.2, 127.4, 127.1, 127.0, 125.1, 123.7, 123.1, 117.5, 110.0, 106.6, 74.5, 62.3, 51.0, 44.0; IR(KBr) ν: 1719, 1690, 1657, 1594, 1489, 1460, 1352, 1261, 1225, 1094, 1079, 7528 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>34</sub>H<sub>28</sub>ClN<sub>2</sub>O<sub>3</sub> ([M+H]<sup>+</sup>): 547.1783, found: 547.1789.

**Methyl-*cis*-1-benzyl-5-methyl-2-oxo-2'-((*E*)-styryl)-1'-(*p*-tolyl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6c):** ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.265 g, 49%, m.p. 191-193 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 8.00 (s, 1H), 7.28 (d, *J* = 7.6 Hz, 4H), 7.12-6.93 (m, 10H), 6.85 (t, *J* = 7.2 Hz, 2H), 6.56-6.47 (m, 2H), 6.37 (d, *J* = 16.0 Hz, 1H), 5.12 (d, *J* = 12.4 Hz, 1H), 5.11 (d, *J* = 15.6 Hz, 1H), 4.55 (d, *J* = 15.6 Hz, 1H), 3.54 (s, 3H), 2.31 (s, 3H), 2.27 (s, 3H); <sup>13</sup>C {<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>) δ: 175.2, 164.7, 147.6, 140.7, 138.2, 135.8, 135.7, 134.3, 133.0, 132.6, 132.4, 129.9, 129.0, 128.6, 128.4, 128.2, 127.1, 127.0, 126.9,

125.7, 124.0, 117.6, 108.7, 106.6, 74.9, 62.2, 50.9, 43.9, 21.2, 20.6; IR(KBr)  $\nu$ : 1710, 1696, 1653, 1592, 1484, 1465, 1353, 1269, 1223, 1095, 1070, 750  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{36}\text{H}_{34}\text{N}_2\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 541.2486, found: 541.2490.

**Methyl-*cis*-1-benzyl-1'-(4-(*tert*-butyl)phenyl)-5-methyl-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[*indoline*-3,3'-pyrrole]-4'-carboxylate (6d)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.326 g, 56 %, m.p. 181-183 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 8.03 (s, 1H), 7.34-7.29 (m, 5H), 7.26 (s, 2H), 7.12-7.03 (m, 6H), 6.85 (t,  $J = 8.0$  Hz, 2H), 6.61-6.55 (m, 1H), 6.48 (d,  $J = 8.0$  Hz, 1H), 6.39 (d,  $J = 16.8$  Hz, 1H), 5.13 (d,  $J = 16.0$  Hz, 1H), 5.11 (s, 1H), 4.55 (d,  $J = 16.0$  Hz, 1H), 3.54 (s, 3H), 2.30 (s, 3H), 1.28 (s, 9H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 175.2, 164.7, 147.3, 145.7, 140.6, 138.1, 135.8, 135.8, 134.0, 133.0, 132.4, 128.9, 128.6, 128.4, 128.2, 127.1, 127.0, 126.3, 126.1, 124.9, 116.8, 108.6, 106.7, 74.7, 62.2, 50.8, 43.9, 34.2, 31.3, 21.2; IR(KBr)  $\nu$ : 2958, 1713, 1688, 1595, 1521, 1496, 1450, 1427, 1405, 1346, 1280, 1237, 1215, 1186, 1122, 1106, 982, 815, 773  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{39}\text{H}_{39}\text{N}_2\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 583.2955, found: 583.2943.

**Methyl-*cis*-1-benzyl-1'-(4-chlorophenyl)-5-methyl-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[*indoline*-3,3'-pyrr (6e)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.280 g, 50 %, m.p. 139-141 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.89 (s, 1H), 7.24-6.90 (m, 16H), 6.54 (d,  $J = 15.6$  Hz, 1H), 6.47 (d,  $J = 8.0$  Hz, 1H), 5.84-5.78 (m, 1H), 5.37 (d,  $J = 9.2$  Hz, 1H), 5.06 (d,  $J = 16.0$  Hz, 1H), 4.90 (d,  $J = 16.0$  Hz, 1H), 3.53 (s, 3H), 2.28 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 177.7, 164.2, 147.4, 139.9, 139.5, 135.6, 134.9, 131.8, 129.4, 129.2, 128.6, 128.5, 128.4, 128.3, 127.9, 127.3, 126.9, 126.6, 125.4, 124.4, 119.3, 109.3, 109.0, 73.5, 62.3, 51.1, 44.9, 21.1; IR(KBr)  $\nu$ : 1717, 1589, 1496, 1438, 1189, 812  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{35}\text{H}_{30}\text{ClN}_2\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 561.1939, found: 561.1937.

**Methyl-*cis*-1-benzyl-1'-(3-chlorophenyl)-5-methyl-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[*indoline*-3,3'-pyrrole]-4'-carboxylateole]-4'-carboxylate (6f)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.330 g, 59 %, m.p. 136-138 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.91 (s, 1H), 7.29-7.27 (m, 2H), 7.24-7.08 (m, 8H), 7.03-6.90 (m, 6H), 6.57 (d,  $J = 16.0$  Hz, 1H), 6.47 (d,  $J = 7.6$  Hz, 1H), 5.81 (dd,  $J_1 = 16.0$  Hz,  $J_2 = 8.8$  Hz, 1H), 5.37 (d,  $J = 9.2$  Hz, 1H), 5.05 (d,  $J = 16.0$  Hz, 1H), 4.90 (d,  $J = 16.0$  Hz, 1H), 3.54 (s, 3H), 2.28 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 178.0, 164.3, 148.2, 142.2, 140.9, 135.7, 135.5, 134.6, 129.4, 128.8, 128.6, 128.5, 128.3,

128.2, 127.6, 127.3, 126.9, 126.7, 124.8, 124.6, 123.3, 122.4, 118.1, 109.5, 108.2, 73.5, 62.4, 51.0, 50.9, 44.1; IR(KBr)  $\nu$ : 1716, 1590, 1496, 1424, 1344, 1253, 1092, 875, 821, 753  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{35}\text{H}_{30}\text{ClN}_2\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 561.1939, found: 561.1942.

**Methyl-*cis*-1-benzyl-5-chloro-1'-(4-chlorophenyl)-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6g)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.278 g, 48%, m.p. 149-151 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.98 (s, 1H), 7.38 (d,  $J = 8.8$  Hz, 2H), 7.30 (d,  $J = 2.0$  Hz, 5H), 7.19-7.03 (m, 6H), 6.85 (d,  $J = 8.8$  Hz, 2H), 6.84 (t,  $J = 7.2$  Hz, 2H), 6.61 (d,  $J = 7.6$  Hz, 1H), 6.50 (dd,  $J_1 = 16.0$  Hz,  $J_2 = 8.8$  Hz, 1H), 6.38 (d,  $J = 16.0$  Hz, 1H), 5.14 (d,  $J = 16.0$  Hz, 1H), 5.12 (d,  $J = 8.8$  Hz, 1H), 4.57 (d,  $J = 16.0$  Hz, 1H), 3.54 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 174.6, 164.2, 147.2, 141.7, 138.9, 135.2, 135.2, 135.1, 134.2, 129.5, 128.8, 128.7, 128.6, 128.5, 128.3, 128.2, 127.4, 127.0, 126.9, 124.5, 123.7, 118.7, 110.1, 107.3, 74.6, 62.3, 51.1, 44.0; IR(KBr)  $\nu$ : 1719, 1699, 1657, 1595, 1482, 1461, 1356, 1268, 1224, 1098, 1070, 756  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{34}\text{H}_{27}\text{Cl}_2\text{N}_2\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 581.1393, found: 581.1400.

**Methyl-*cis*-1-benzyl-1'-(4-bromophenyl)-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6h)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.236 g, 40%, m.p. 159-161 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.89 (s, 1H), 7.39 (d,  $J = 9.2$  Hz, 2H), 7.24-7.20 (m, 4H), 7.15-7.07 (m, 3H), 7.03-6.97 (m, 4H), 6.90 (t,  $J = 8.4$  Hz, 1H), 6.54 (d,  $J = 16.0$  Hz, 1H), 6.47 (d,  $J = 8.0$  Hz, 1H), 5.80 (dd,  $J_1 = 16.0$  Hz,  $J_2 = 9.2$  Hz, 1H), 5.36 (d,  $J = 9.2$  Hz, 1H), 5.04 (d,  $J = 16.0$  Hz, 1H), 4.90 (d,  $J = 16.0$  Hz, 1H), 3.53 (s, 3H), 2.27 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 177.7, 164.2, 147.3, 140.0, 139.9, 135.6, 134.9, 132.3, 131.8, 129.2, 128.6, 128.5, 128.3, 127.9, 127.3, 126.9, 126.7, 125.5, 124.4, 119.56, 115.8, 109.3, 109.2, 73.4, 62.4, 51.1, 51.0, 44.1, 21.1; IR(KBr)  $\nu$ : 3062, 1715, 1692, 1654, 1597, 1482, 1465, 1359, 1261, 1224, 1096, 1070, 750  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{34}\text{H}_{28}\text{BrN}_2\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 591.2053, found: 591.2056.

**Methyl-*cis*-1,1'-dibenzyl-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6i)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.316 g, 60%, m.p. 229-231 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.64 (s, 1H), 7.42-7.27 (m, 10H), 7.11-6.95 (m, 6H), 6.83 (t,  $J = 7.6$  Hz, 2H), 6.53-6.47 (m, 2H), 6.20 (d,  $J = 15.6$  Hz, 1H), 5.16 (d,  $J = 16.0$  Hz, 1H), 4.58 (d,  $J = 16.0$  Hz, 1H), 4.46 (d,  $J = 15.2$  Hz, 1H), 4.38 (d,  $J = 9.2$  Hz, 1H), 4.18 (d,  $J =$

15.2 Hz, 1H), 3.47 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 175.8, 164.8, 153.0, 143.0, 136.3, 135.7, 135.6, 135.5, 133.1, 128.9, 128.7, 128.4, 128.3, 128.2, 128.1, 127.1, 127.0, 126.9, 123.2, 123.1, 122.7, 108.7, 103.3, 75.1, 61.5, 51.8, 50.5, 43.7; IR(KBr)  $\nu$ : 1714, 1684, 1604, 1512, 1432, 1347, 1256, 1188, 1110, 1037, 829  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{35}\text{H}_{31}\text{N}_2\text{O}_3$  ( $[\text{M}+\text{H}]^+$ ): 527.2329, found: 527.2313.

**Methyl-*cis*-1-benzyl-1'-(4-methoxyphenyl)-2'-((*E*)-4-methoxystyryl)-5-methyl-2-oxo-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6j)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.328 g, 56%, m.p. 211-213  $^\circ\text{C}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.91 (s, 1H), 7.24 (d,  $J = 8.0$  Hz, 2H), 7.12 (d,  $J = 6.1$  Hz, 3H), 7.06-7.03 (m, 3H), 6.94-6.80 (m, 7H), 6.46 (d,  $J = 8.0$  Hz, 1H), 6.41-6.35 (m, 1H), 6.28 (d,  $J = 16.0$  Hz, 1H), 5.13 (d,  $J = 15.6$  Hz, 1H), 5.04 (d,  $J = 8.8$  Hz, 1H), 4.56 (d,  $J = 15.6$  Hz, 1H), 3.79 (s, 3H), 3.76 (s, 3H), 3.54 (s, 3H), 2.31 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 175.3, 164.8, 159.7, 155.8, 148.4, 140.7, 135.8, 134.2, 134.1, 133.1, 132.3, 128.9, 128.5, 128.4, 128.3, 127.1, 127.0, 123.9, 123.0, 119.8, 114.6, 113.9, 108.6, 105.9, 75.8, 62.2, 55.5, 55.4, 50.8, 43.9, 21.2; IR(KBr)  $\nu$ : 1706, 1678, 1593, 1488, 1467, 1455, 1428, 1354, 1256, 1231, 1179, 1148, 1088, 1017, 977, 941, 897, 752  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{37}\text{H}_{35}\text{N}_2\text{O}_5$  ( $[\text{M}+\text{H}]^+$ ): 587.2540, found: 587.2530.

**Methyl-*cis*-1-benzyl-1'-(2,6-dimethylphenyl)-2'-((*E*)-4-methoxystyryl)-5-methyl-2-oxo-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6k)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.415 g, 71%, m.p. 236-238  $^\circ\text{C}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$ : 7.51 (s, 1H), 7.21 (s, 1H), 7.14-7.02 (m, 8H), 6.91 (d,  $J = 8.4$  Hz, 1H), 6.86 (t,  $J = 7.6$  Hz, 2H), 6.74 (t,  $J = 9.2$  Hz, 2H), 6.45 (d,  $J = 8.0$  Hz, 1H), 6.33 (dd,  $J_1 = 15.6$  Hz,  $J_2 = 9.6$  Hz, 1H), 6.03 (d,  $J = 15.6$  Hz, 1H), 5.14 (d,  $J = 16.0$  Hz, 1H), 5.05 (d,  $J = 9.6$  Hz, 1H), 4.64 (d,  $J = 16.0$  Hz, 1H), 3.76 (s, 3H), 3.50 (s, 3H), 2.51 (s, 3H), 2.40 (s, 3H), 2.35 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$ : 175.8, 165.1, 159.7, 152.7, 141.0, 138.4, 137.2, 136.5, 135.8, 135.1, 133.3, 132.2, 128.9, 128.7, 128.5, 128.4, 128.4, 128.2, 128.0, 127.1, 126.9, 124.0, 120.1, 113.9, 108.6, 103.2, 97.6, 62.0, 55.3, 55.3, 50.6, 50.5, 43.8, 21.3, 19.1, 18.8; IR(KBr)  $\nu$ : 1719, 1681, 1600, 1511, 1435, 1346, 1251, 1189, 1111, 1036, 823  $\text{cm}^{-1}$ ; MS ( $m/z$ ): HRMS (ESI-TOF) Calcd. For  $\text{C}_{38}\text{H}_{37}\text{N}_2\text{O}_4$  ( $[\text{M}+\text{H}]^+$ ): 585.2748, found: 585.2723.

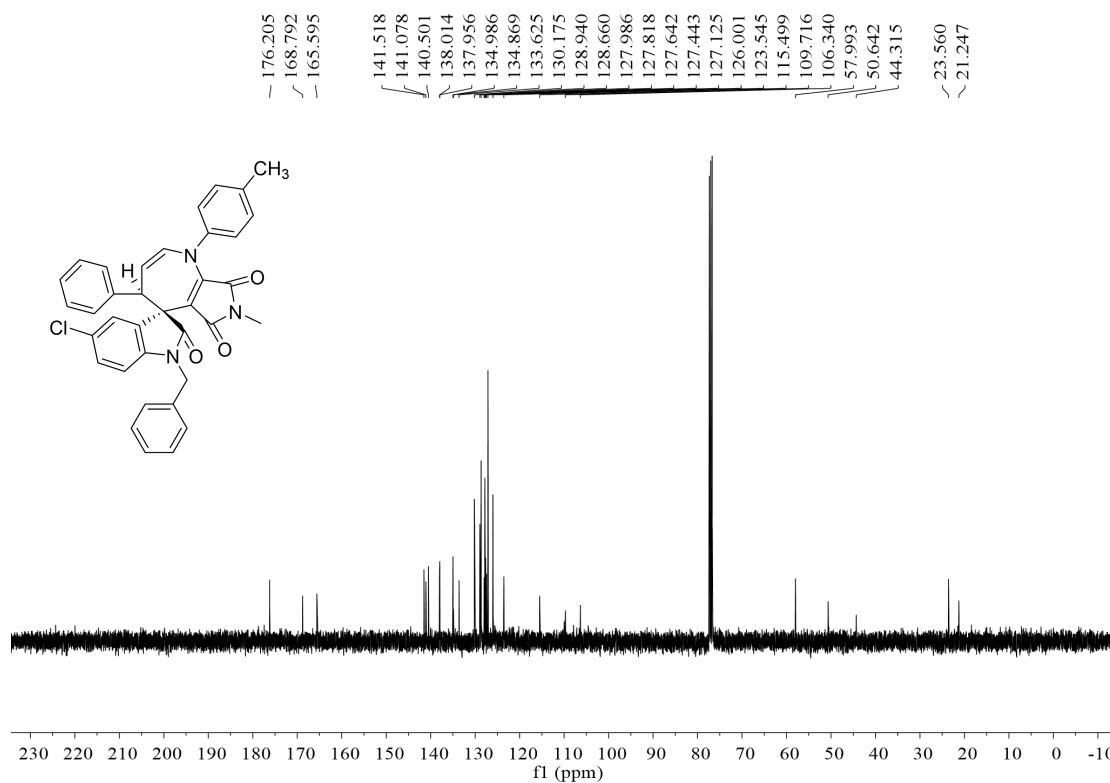
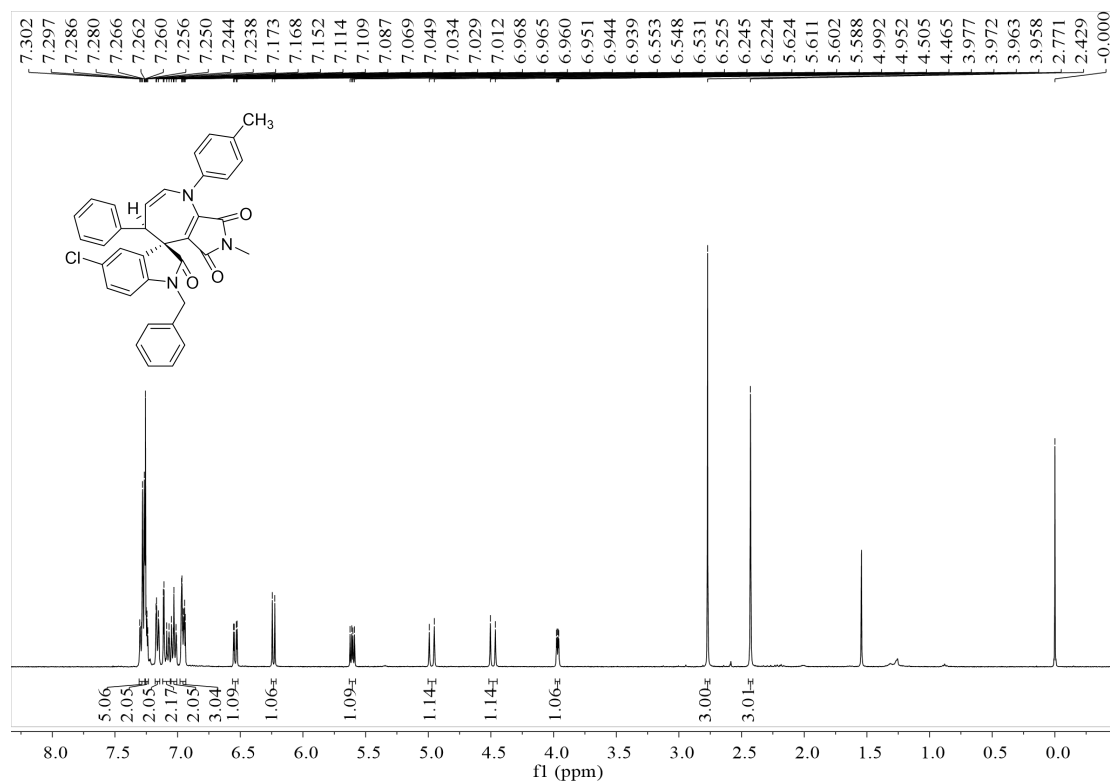
**Methyl-*cis*-1-benzyl-2'-((*E*)-4-chlorostyryl)-5-methyl-2-oxo-1'-(*o*-tolyl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6l)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent,

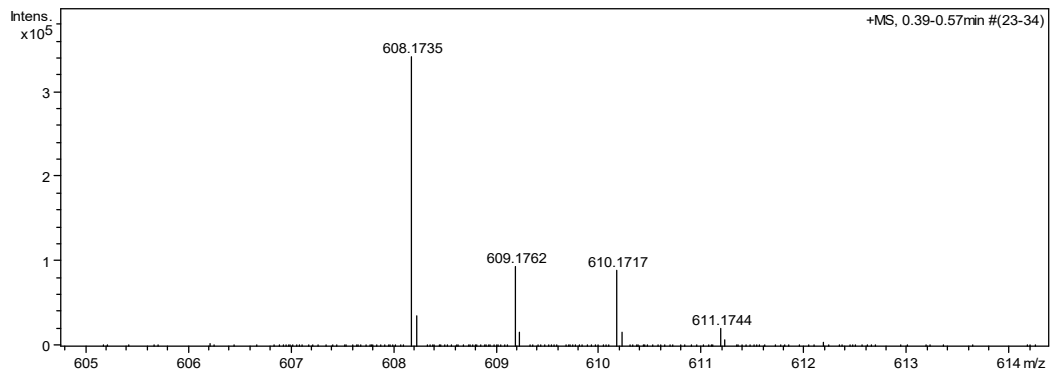
white solid, 0.390 g, 68%, m.p. 219-221 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.57 (s, 1H), 7.23-7.11 (m, 10H), 7.04 (t, *J* = 7.6 Hz, 2H), 6.93 (d, *J* = 7.6 Hz, 1H), 6.83 (d, *J* = 8.4 Hz, 2H), 6.48 (d, *J* = 8.0 Hz, 1H), 6.37 (d, *J* = 16.0 Hz, 1H), 5.72 (dd, *J*<sub>1</sub> = 15.6 Hz, *J*<sub>2</sub> = 6.0 Hz, 1H), 5.32 (d, *J* = 9.2 Hz, 1H), 5.09 (d, *J* = 16.0 Hz, 1H), 4.85 (d, *J* = 16.0 Hz, 1H), 3.52 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ: 177.9, 164.5, 151.5, 139.7, 139.7, 135.7, 134.6, 134.2, 133.9, 133.7, 131.9, 131.5, 129.2, 128.6, 128.5, 128.3, 127.8, 127.2, 126.9, 126.8, 126.6, 124.9, 124.7, 123.9, 109.3, 107.6, 61.9, 50.9, 50.9, 44.0, 21.2, 18.8; IR(KBr) ν: 1718, 1685, 1604, 1510, 1432, 1345, 1252, 1189, 1111, 1036, 823, 752 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>36</sub>H<sub>32</sub>ClN<sub>2</sub>O<sub>3</sub> ([M+H]<sup>+</sup>): 575.2096, found: 575.2102.

**cis-1-Benzyl-5-methyl-2-oxo-2'-((E)-styryl)-1'-(p-tolyl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carbonitrile (6m)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.314 g, 62%, m.p. 198-200 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.68 (s, 1H), 7.27 (s, 4H), 7.23 (s, 1H), 7.09 (d, *J* = 8.4 Hz, 2H), 7.05-6.95 (m, 7H), 6.78 (t, *J* = 7.6 Hz, 2H), 6.51-6.45 (m, 2H), 6.37 (d, *J* = 16.0 Hz, 1H), 5.17 (d, *J* = 16.0 Hz, 1H), 5.14 (d, *J* = 8.8 Hz, 1H), 4.49 (d, *J* = 16.0 Hz, 1H), 2.35 (s, 3H), 2.28 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ: 173.8, 150.5, 140.6, 137.8, 135.6, 135.3, 135.0, 133.5, 133.2, 130.1, 130.0, 129.6, 128.7, 128.52, 128.50, 127.3, 127.0, 126.9, 126.8, 125.0, 124.1, 118.2, 116.4, 109.3, 84.4, 74.3, 62.8, 44.0, 29.7, 21.17, 20.7; IR(KBr) ν: 2901, 1710, 1682, 1606, 1574, 1511, 1497, 1478, 1429, 1398, 1340, 1248, 1222, 1177, 1133, 1106, 1028, 968, 857, 828, 806, 788, 771 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>35</sub>H<sub>30</sub>N<sub>3</sub>O ([M+H]<sup>+</sup>): 508.2383, found: 508.2399.

**cis-1-Benzyl-1'-(4-methoxyphenyl)-5-methyl-2-oxo-2'-((E)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carbonitrile (6n)**: ethyl acetate and petroleum ether (V/V = 1:8) as eluent, white solid, 0.356 g, 68%, m.p. 212-214 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ: 7.60 (s, 1H), 7.27 (s, 4H), 7.25 (s, 1H), 7.05-6.98 (m, 7H), 6.83 (d, *J* = 8.8 Hz, 2H), 6.77 (t, *J* = 7.6 Hz, 2H), 6.49-6.45 (m, 2H), 6.34 (d, *J* = 16.0 Hz, 1H), 5.18 (d, *J* = 15.6 Hz, 1H), 5.07 (d, *J* = 8.8 Hz, 1H), 4.49 (d, *J* = 15.6 Hz, 1H), 3.76 (s, 3H), 2.36 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ: 173.9, 156.5, 151.2, 146.2, 140.7, 135.9, 135.3, 135.0, 133.8, 133.1, 130.0, 129.6, 128.7, 128.6, 128.5, 127.3, 127.0, 126.8, 126.7, 126.7, 125.0, 123.8, 121.3, 120.7, 116.5, 114.7, 83.9, 62.7, 55.5, 44.0, 29.7, 21.7 21.2; IR(KBr) ν: 2195, 1707, 1594, 1519, 1496, 1435, 1352, 1250, 1224, 826 cm<sup>-1</sup>; MS (*m/z*): HRMS (ESI-TOF) Calcd. For C<sub>35</sub>H<sub>30</sub>N<sub>3</sub>O<sub>2</sub> ([M+H]<sup>+</sup>): 524.2333, found: 524.2331.

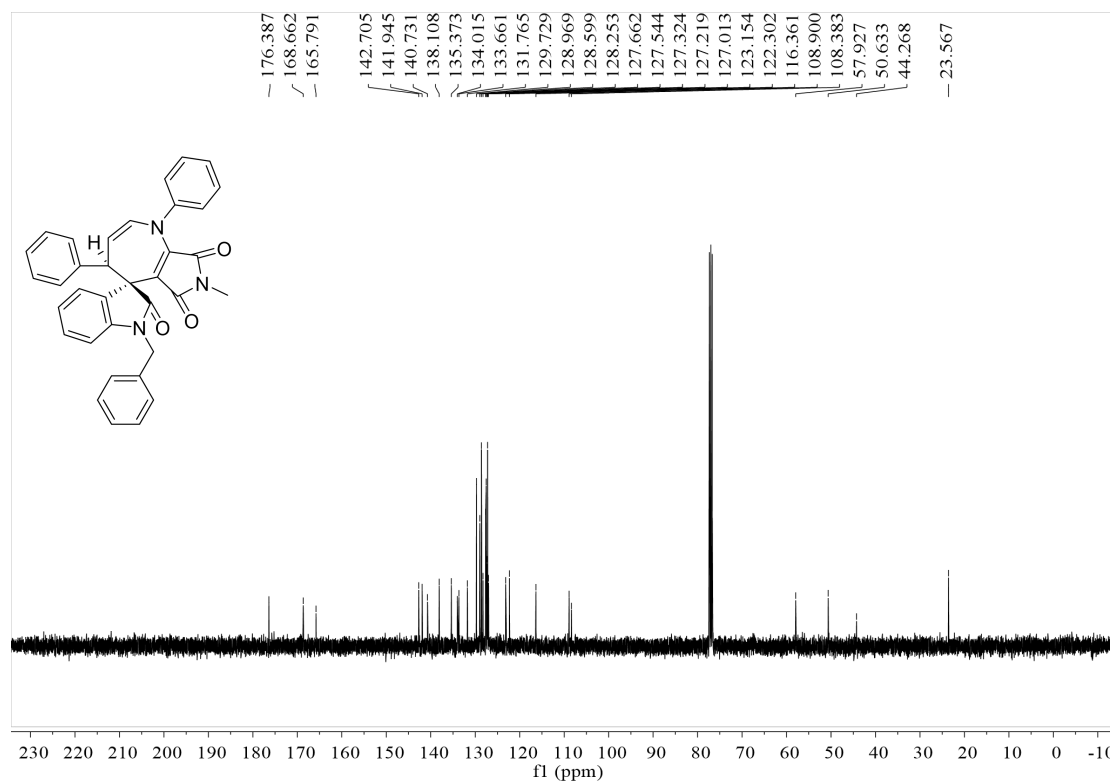
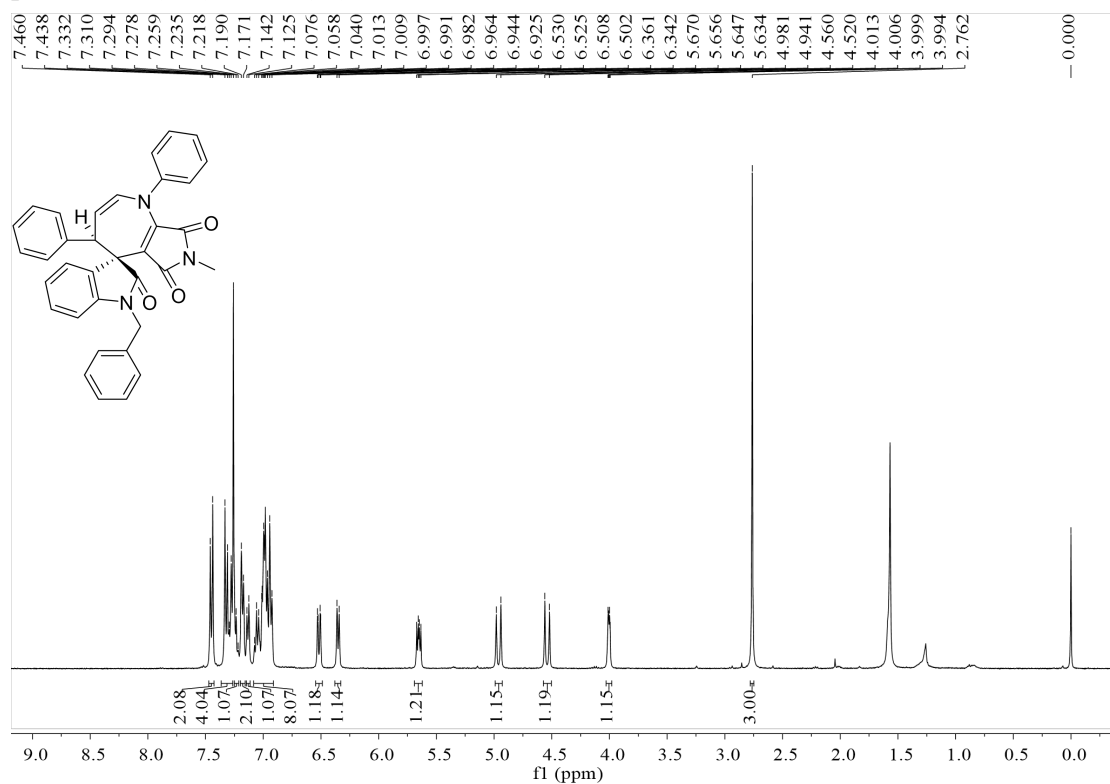
**cis-1-benzyl-5-chloro-7'-methyl-4'-phenyl-1'-(p-tolyl)-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (3a):**

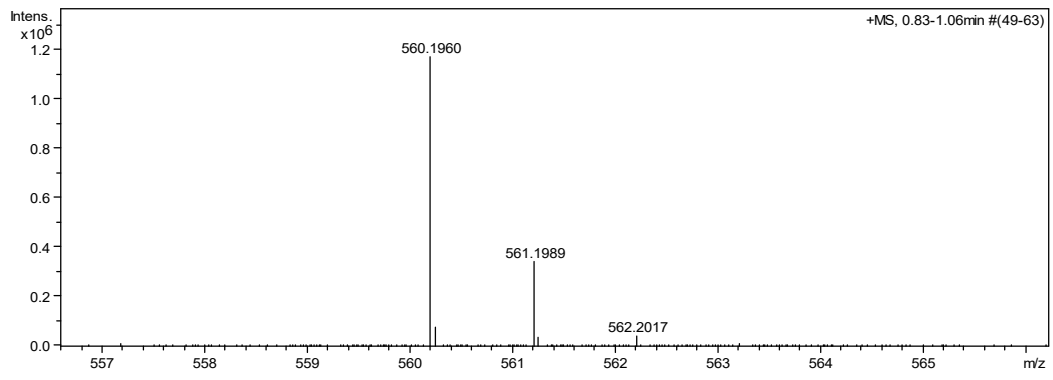




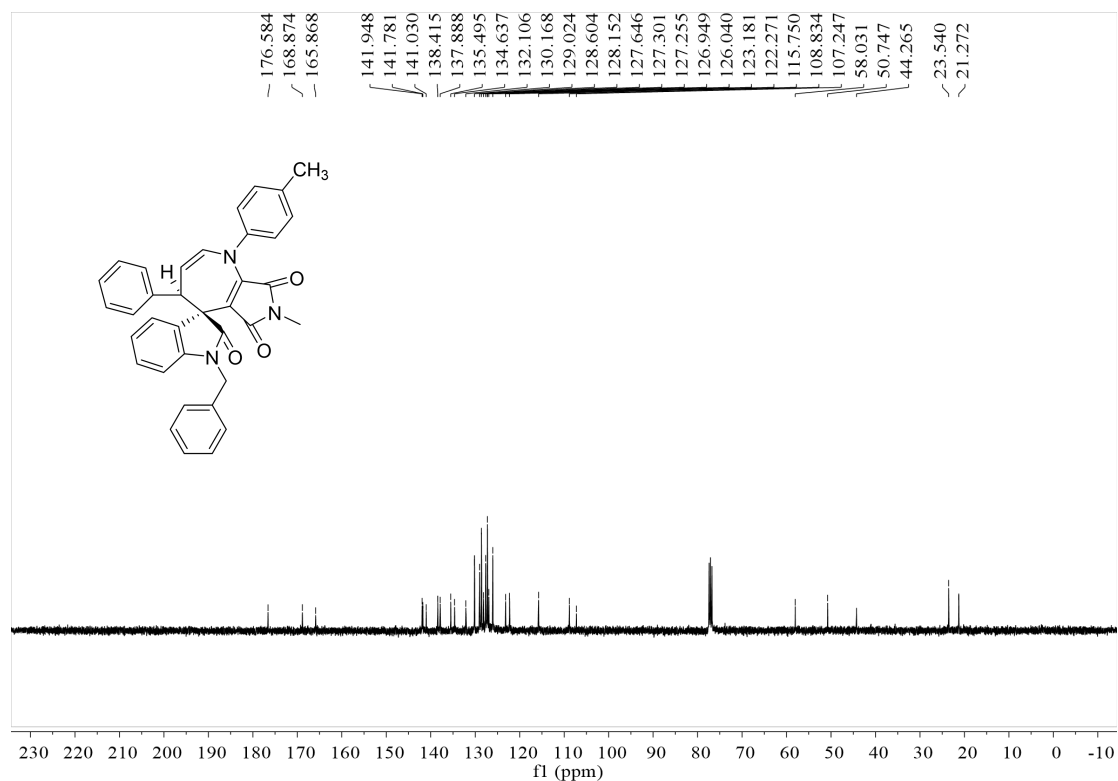
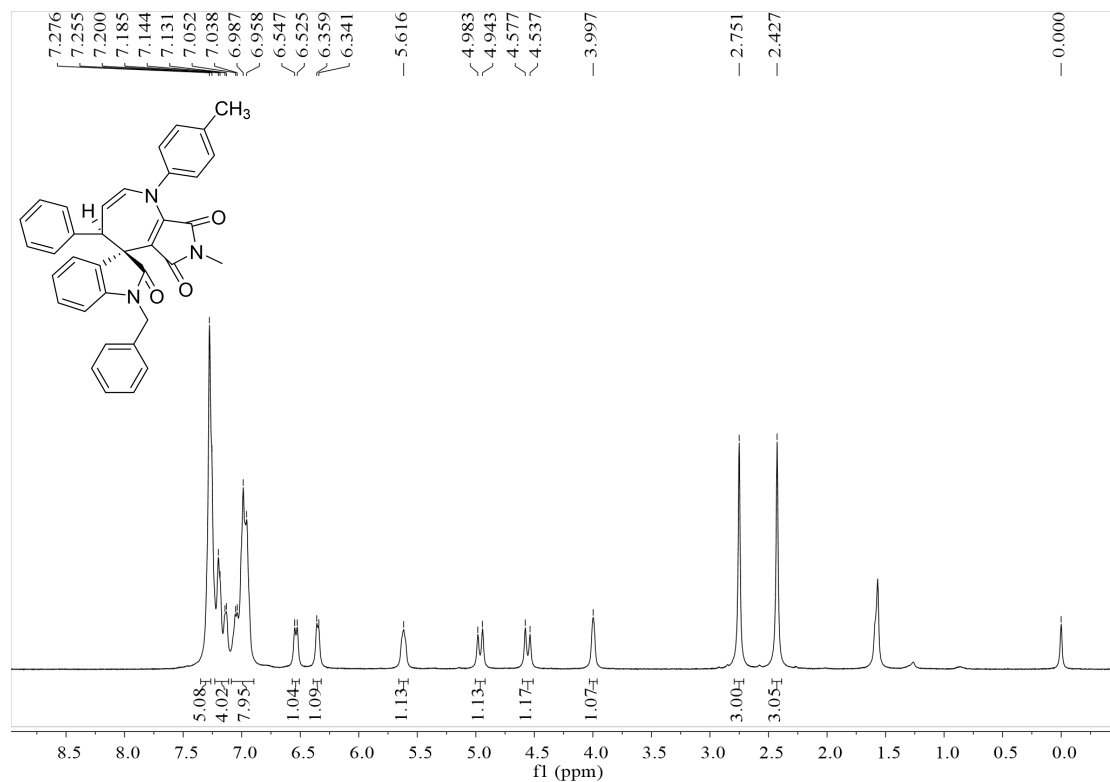


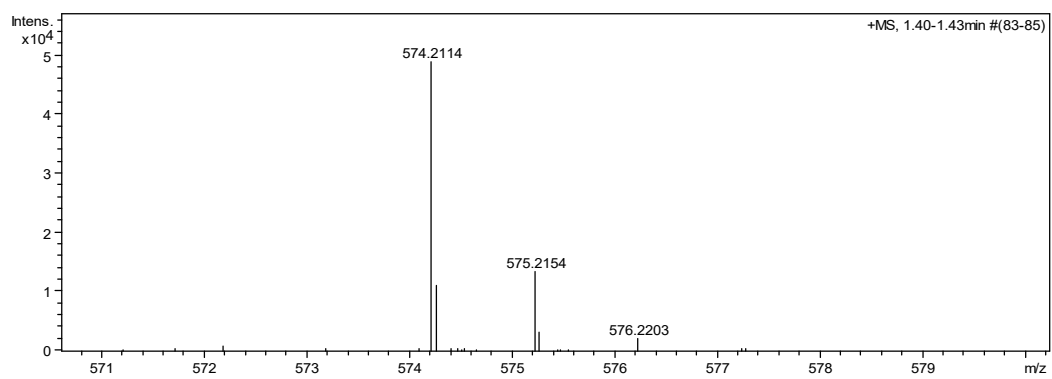
**cis-1-benzyl-7'-methyl-1',4'-diphenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (3b):**



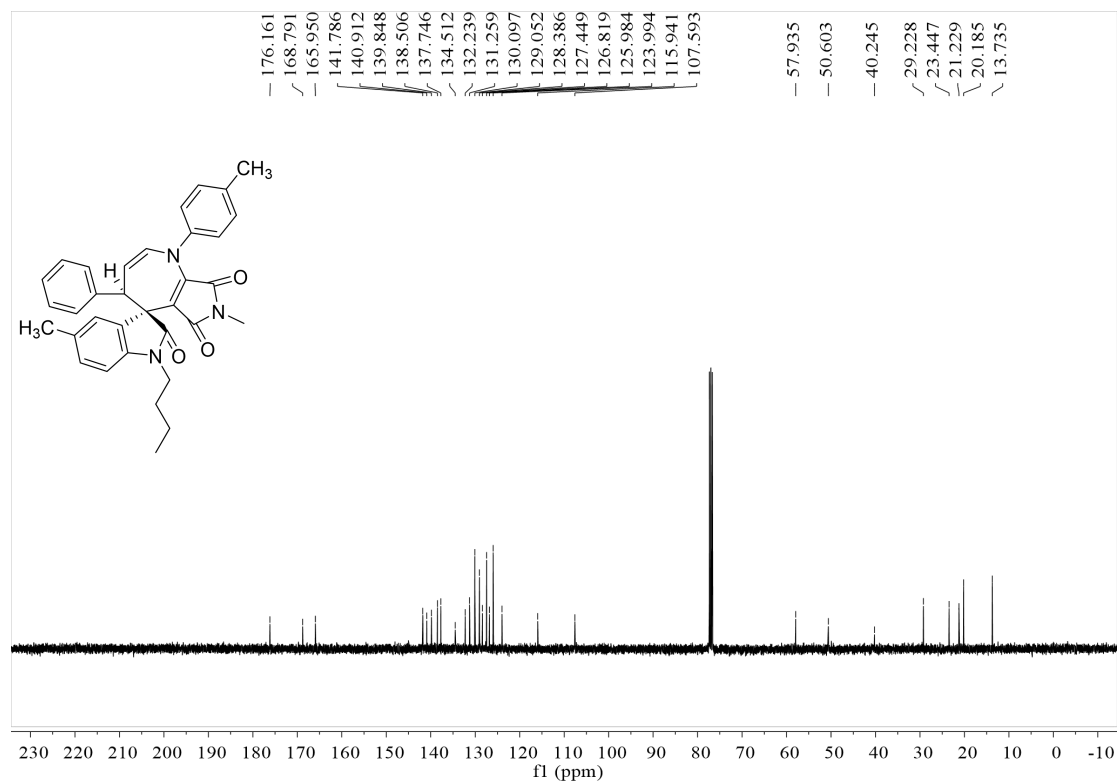
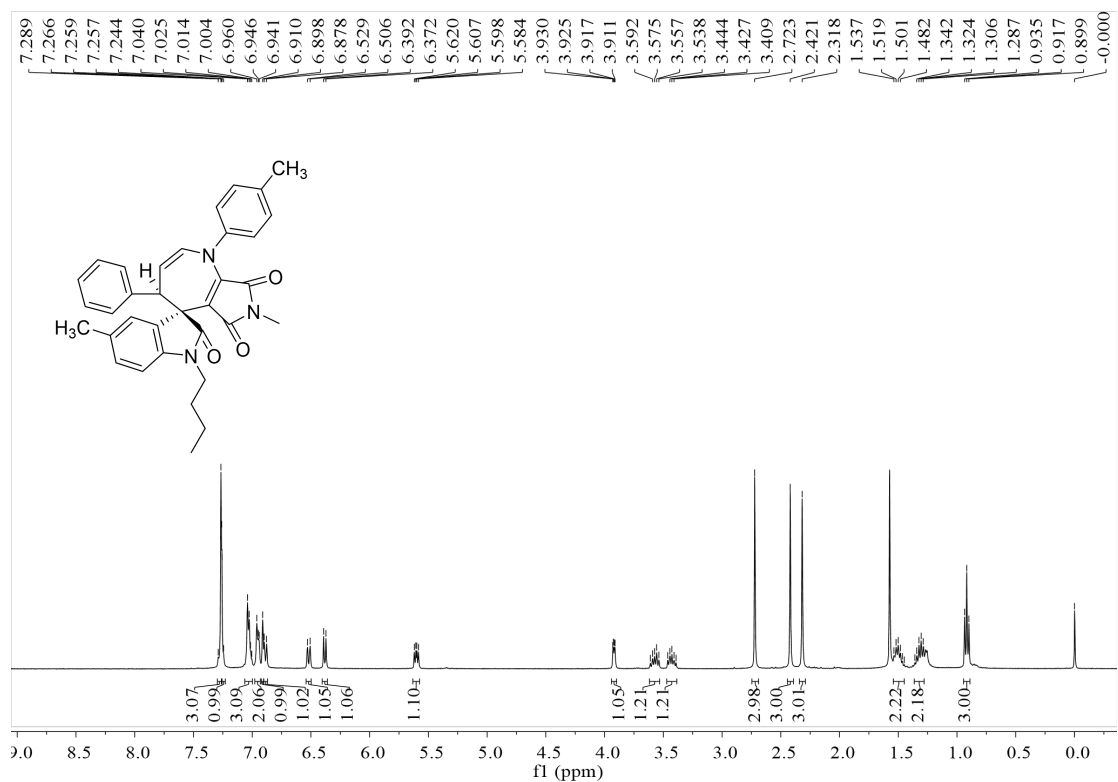


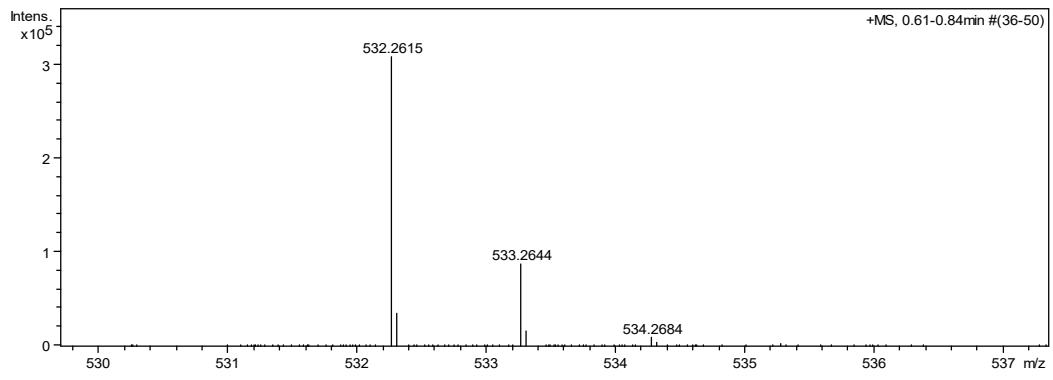
**cis-1-benzyl-7'-methyl-4'-phenyl-1'-(*p*-tolyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3c):**



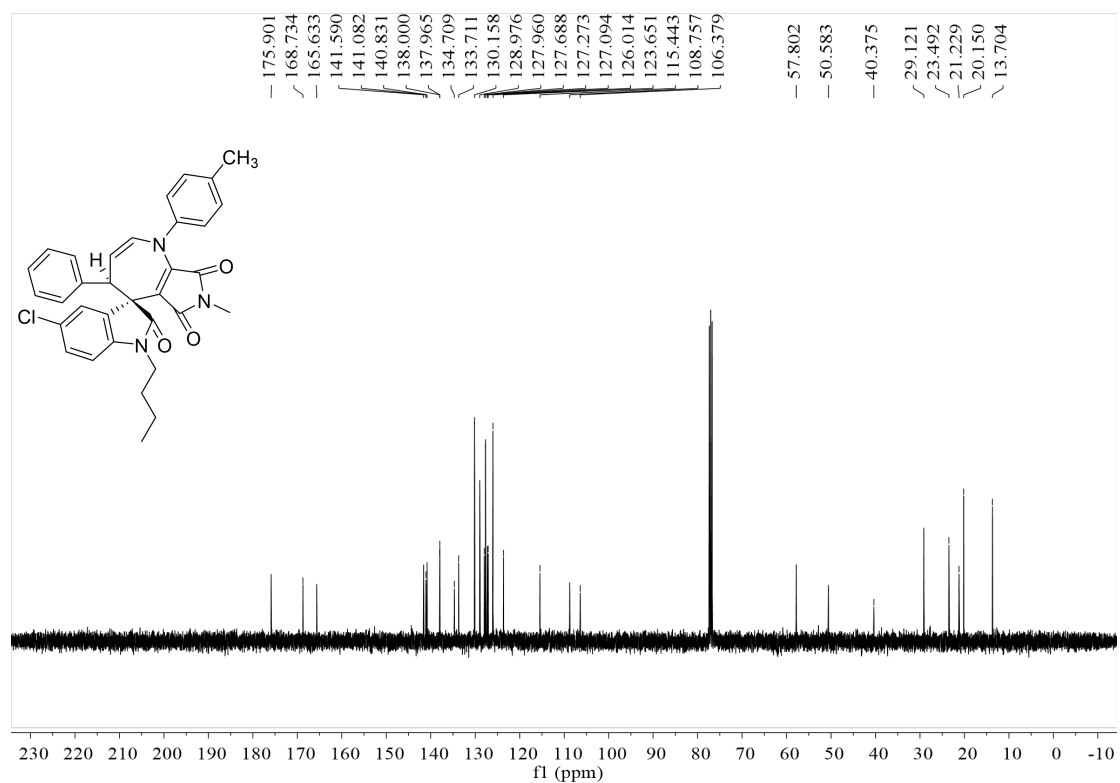
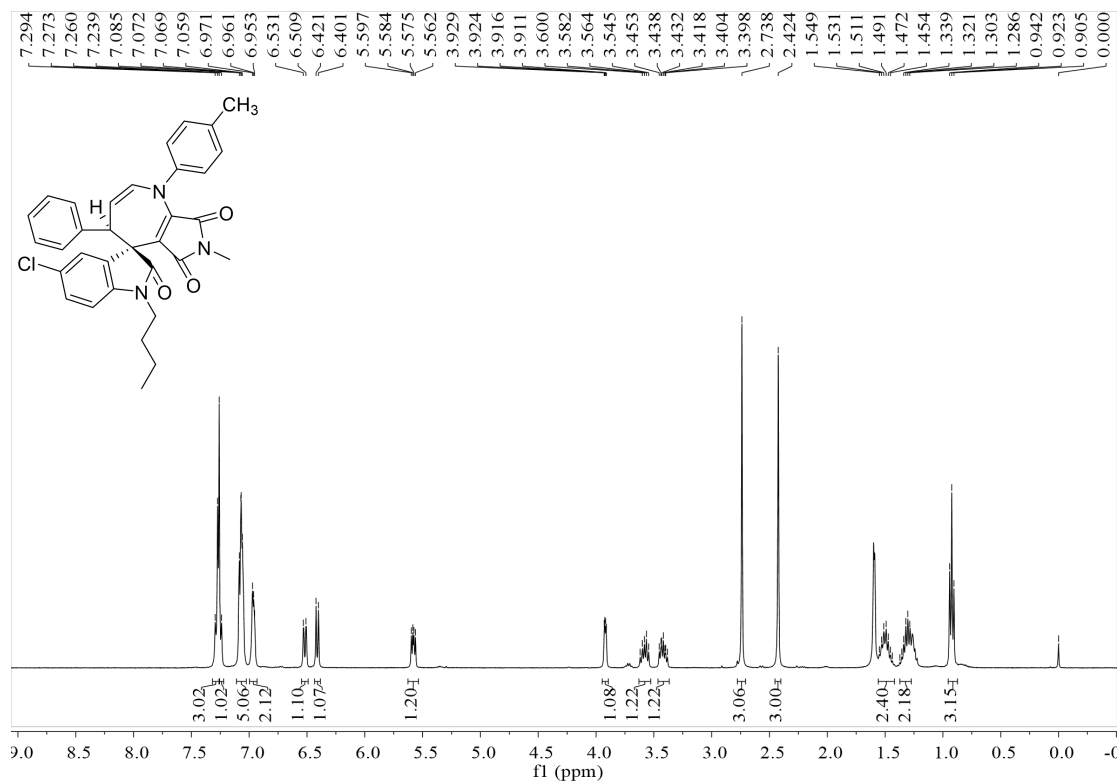


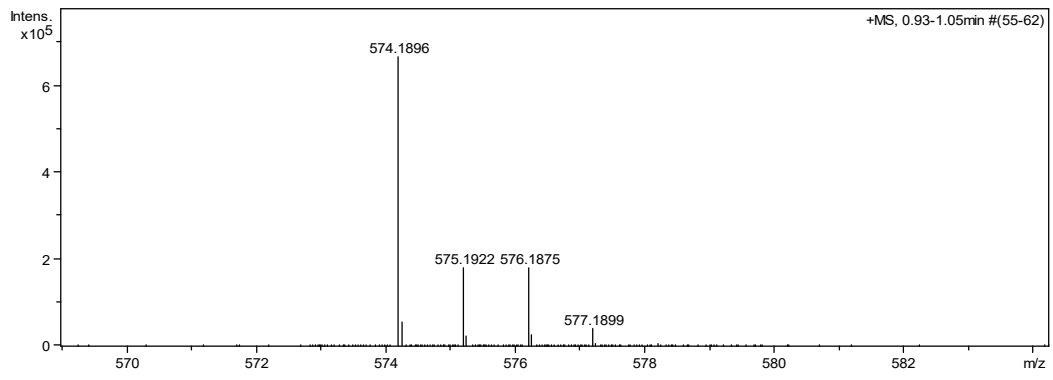
***cis*-1-butyl-5,7'-dimethyl-4'-phenyl-1'-(*p*-tolyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3d):**





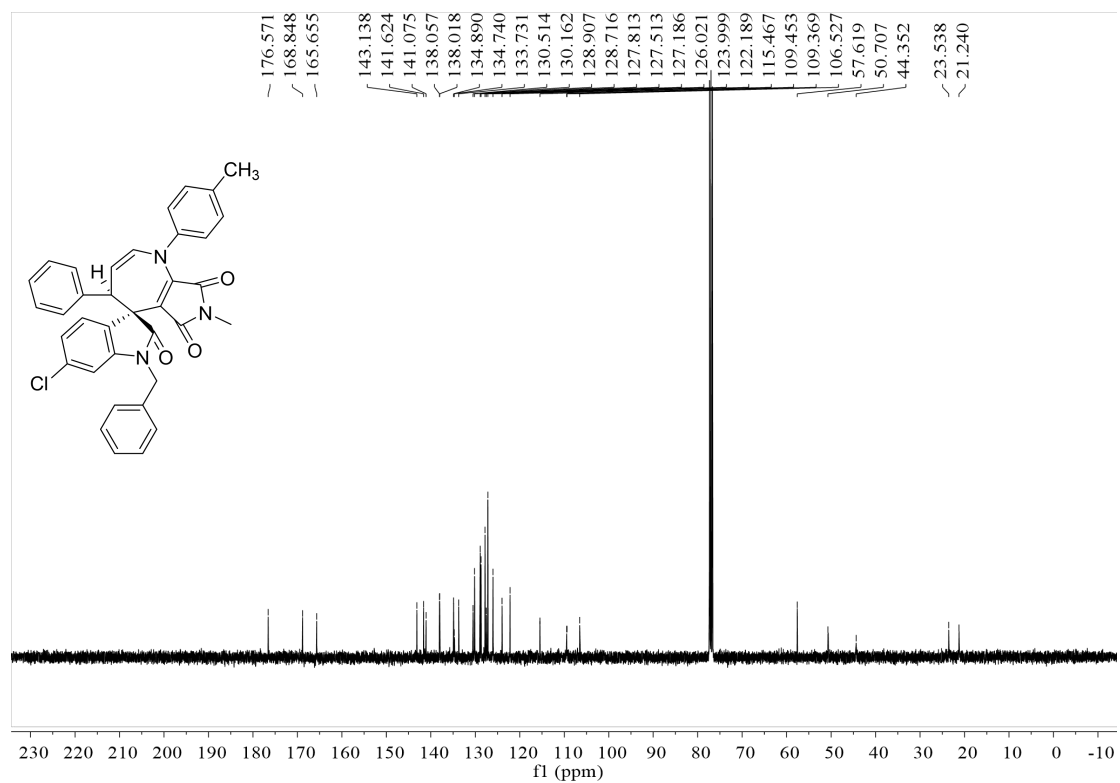
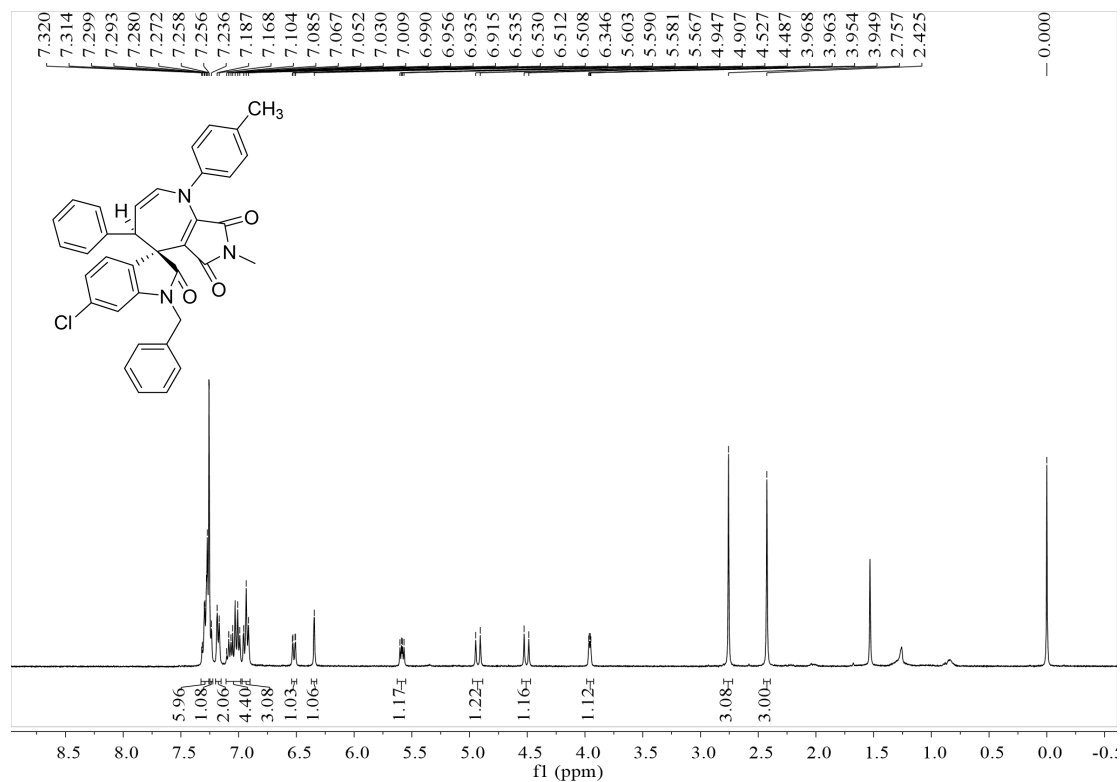
***cis*-1-butyl-5-chloro-7'-methyl-4'-phenyl-1'-(*p*-tolyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3e):**

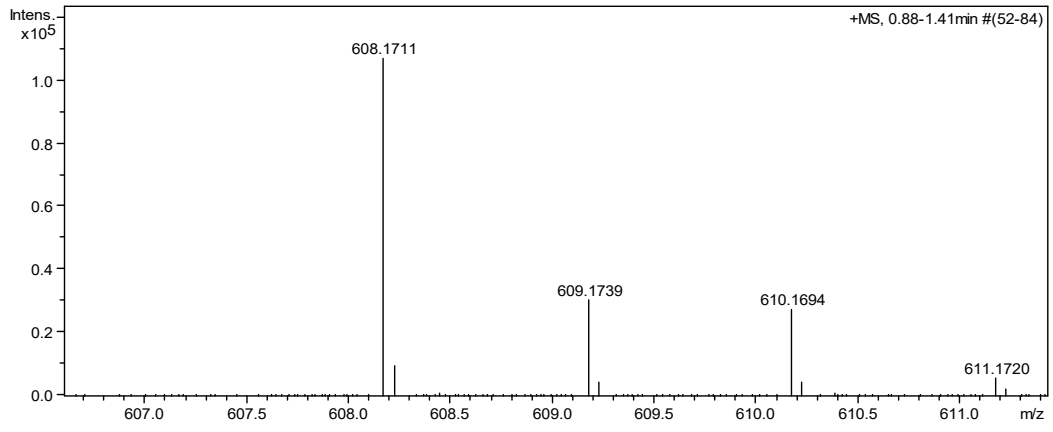




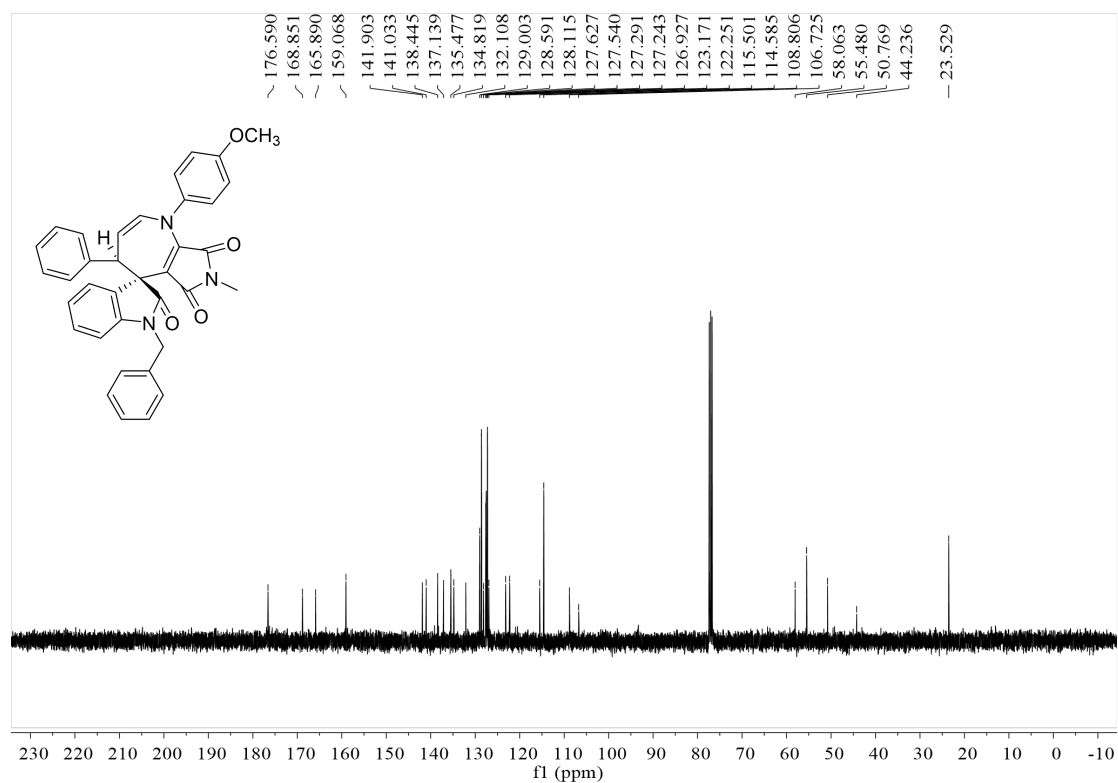
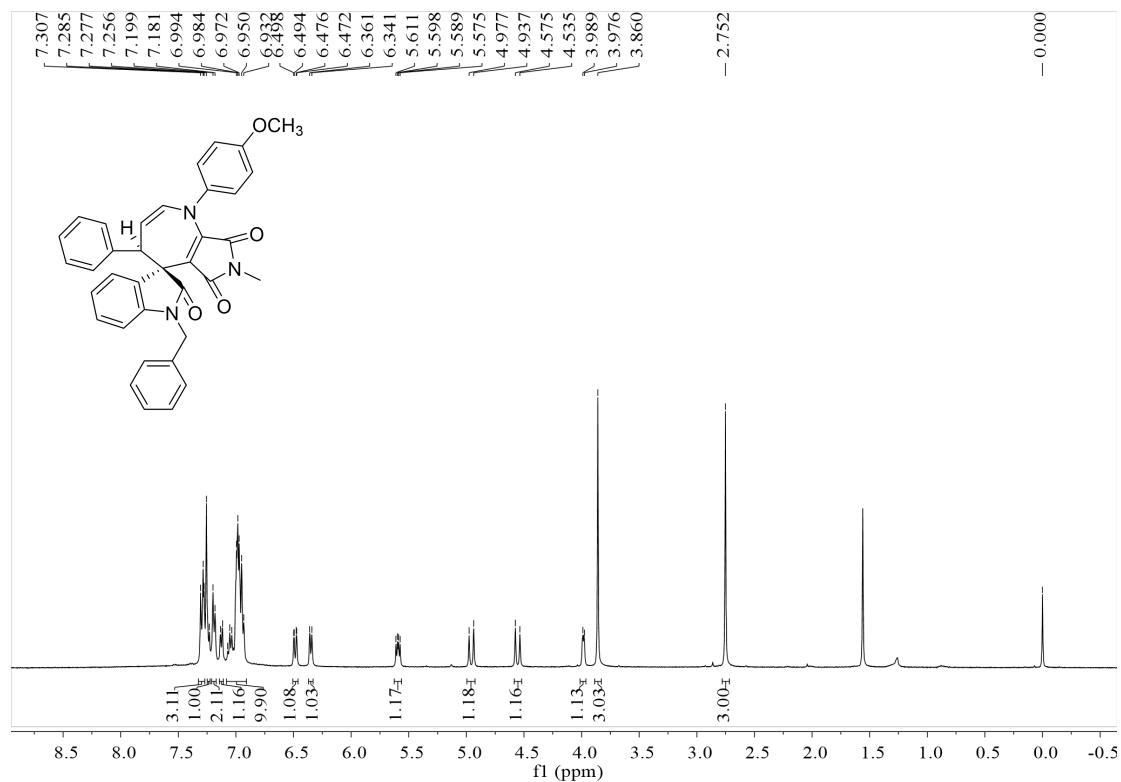


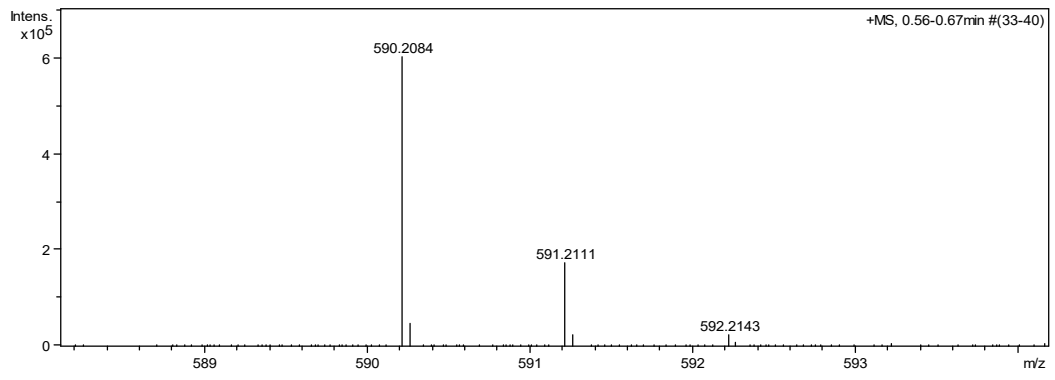
***cis*-1-benzyl-6-chloro-7'-methyl-4'-phenyl-1'-(*p*-tolyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-*p*yrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3f):**



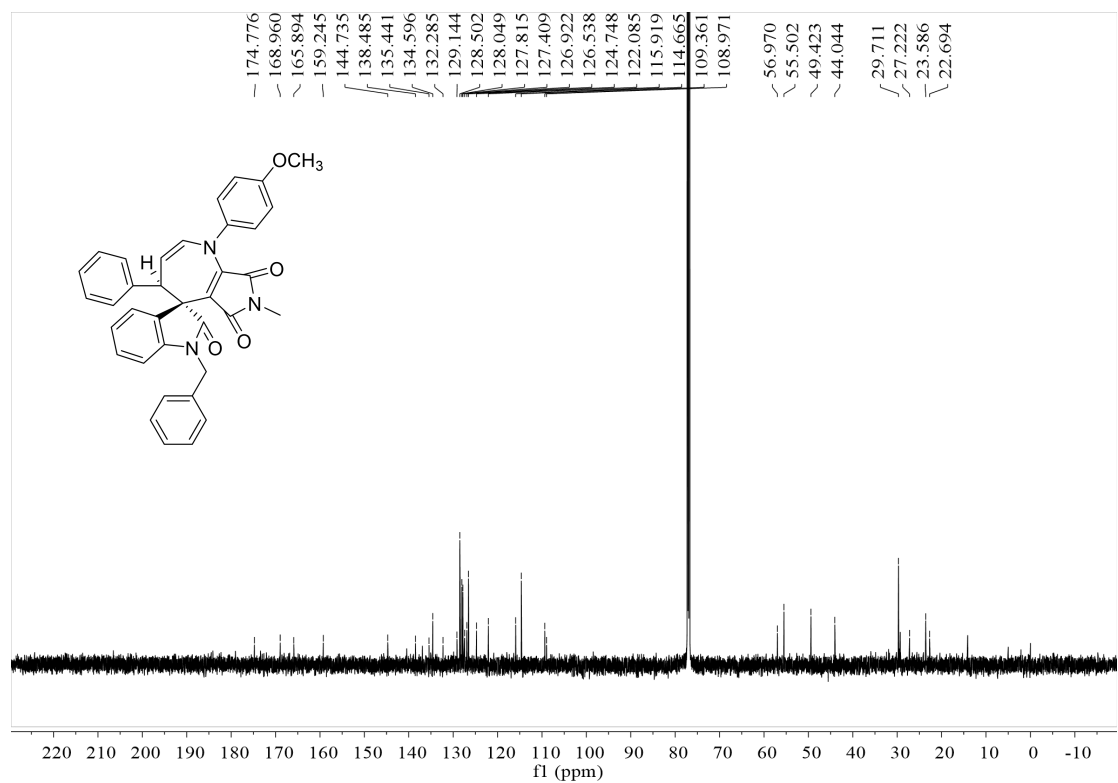
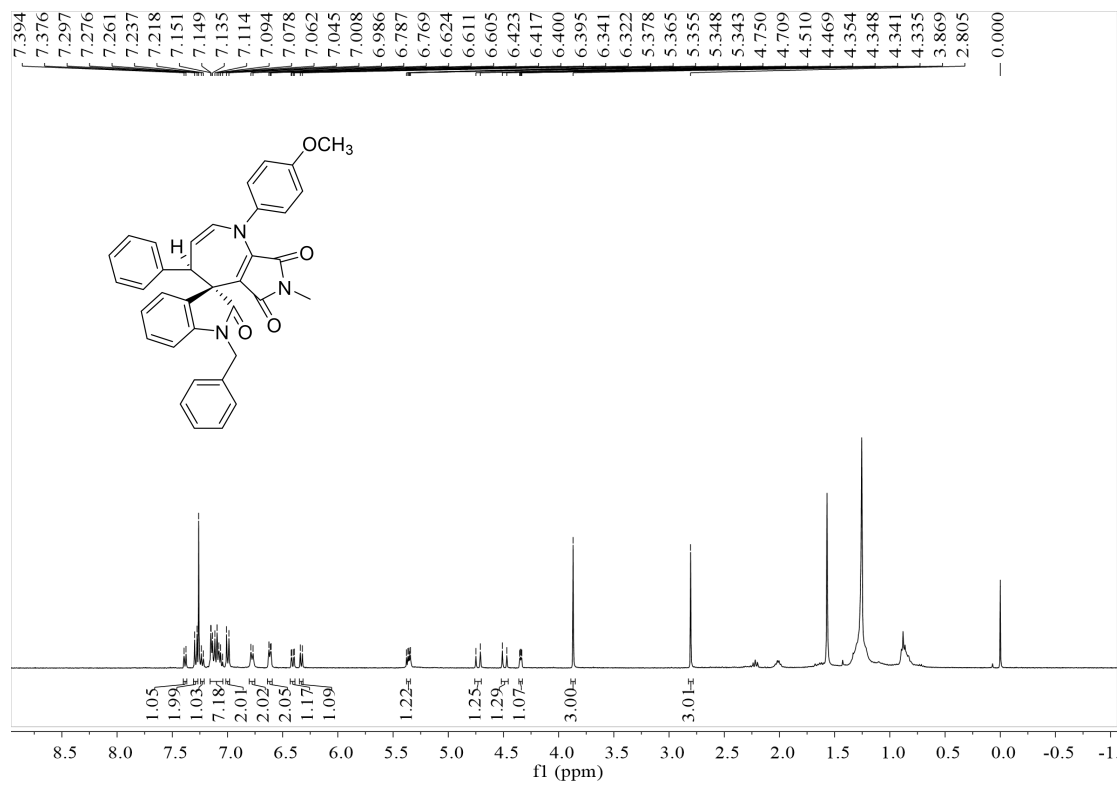


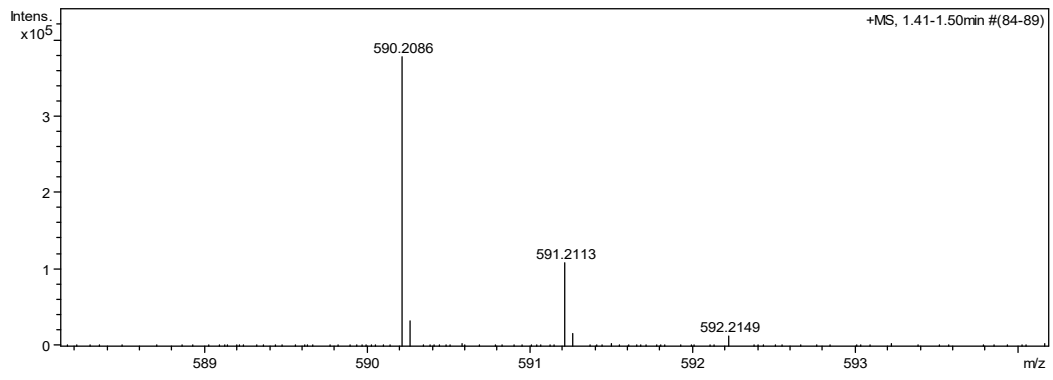
***cis*-1-benzyl-1'-(4-methoxyphenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3g):**



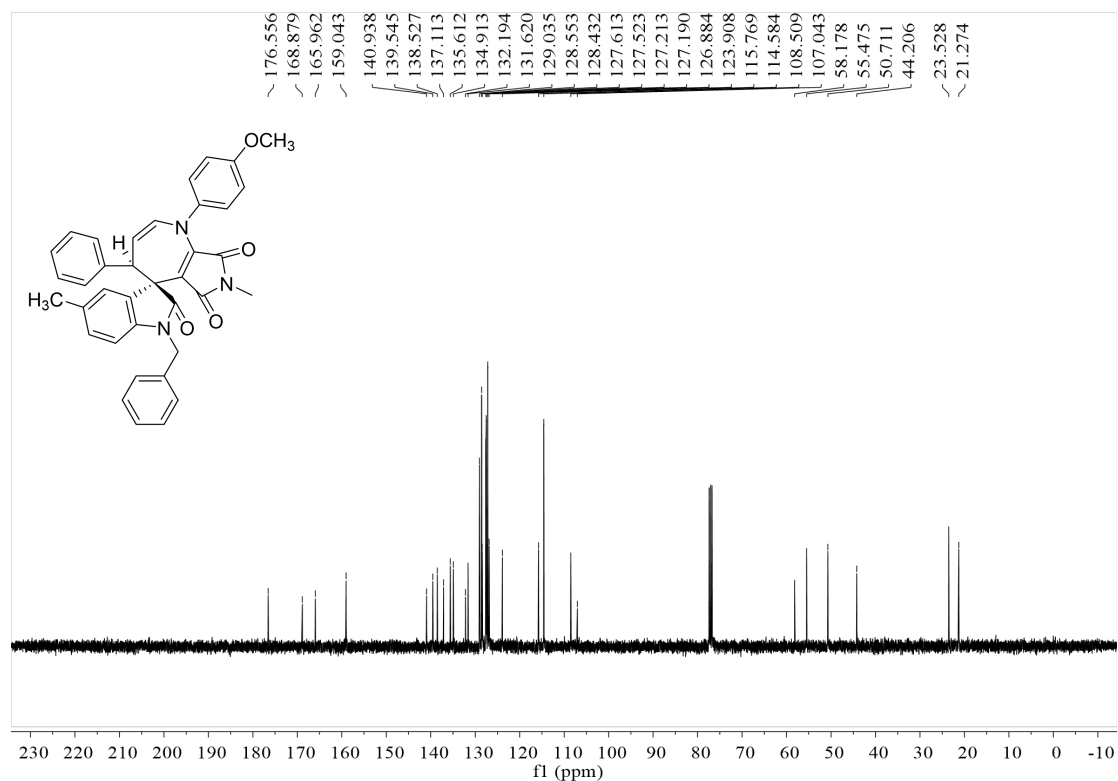
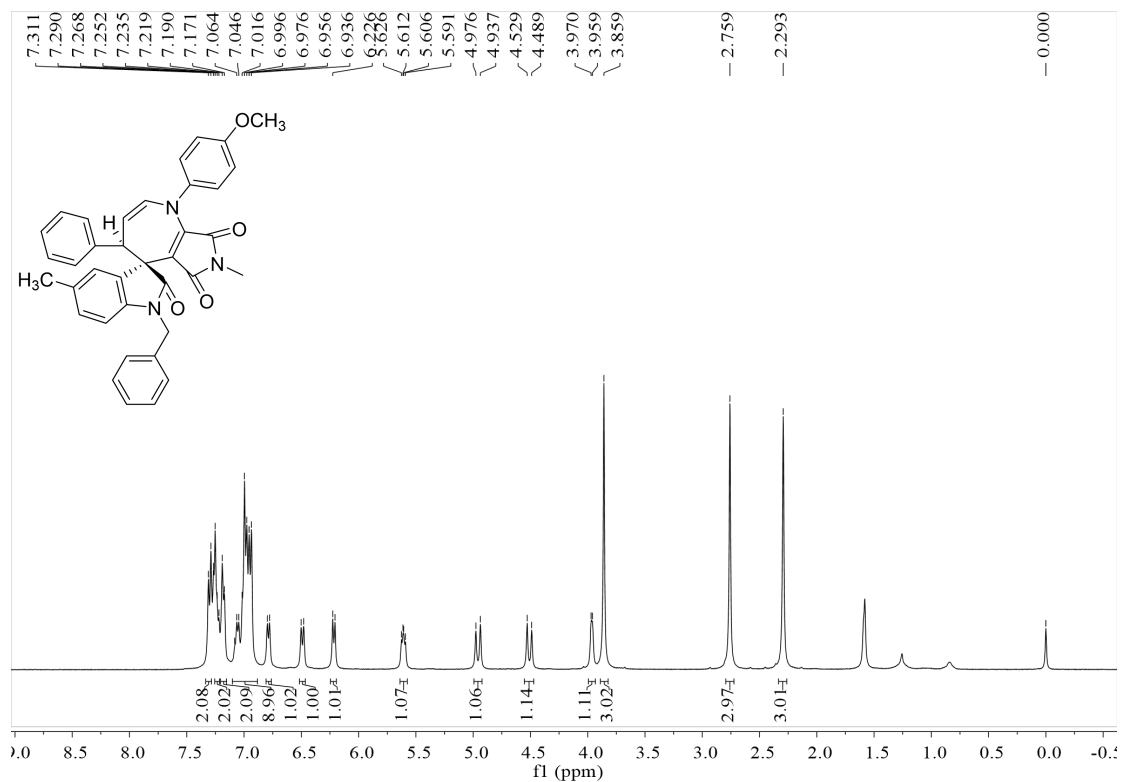


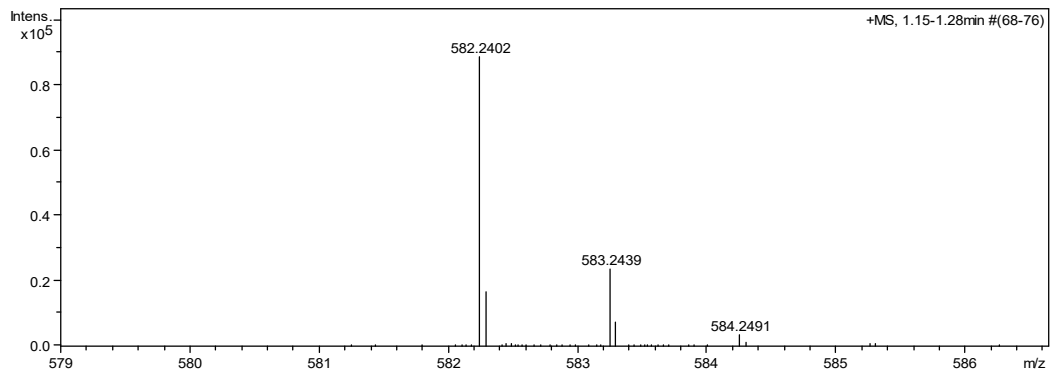
***trans*-1-benzyl-1'-(4-methoxyphenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3, 5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3g')**





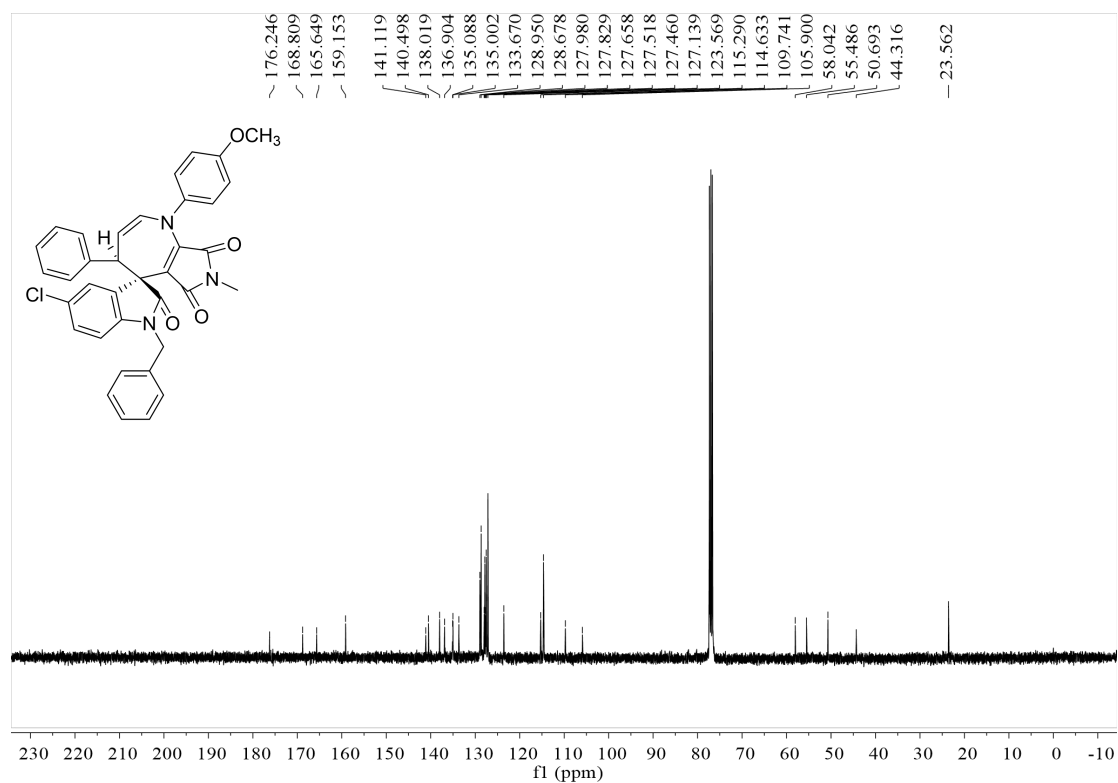
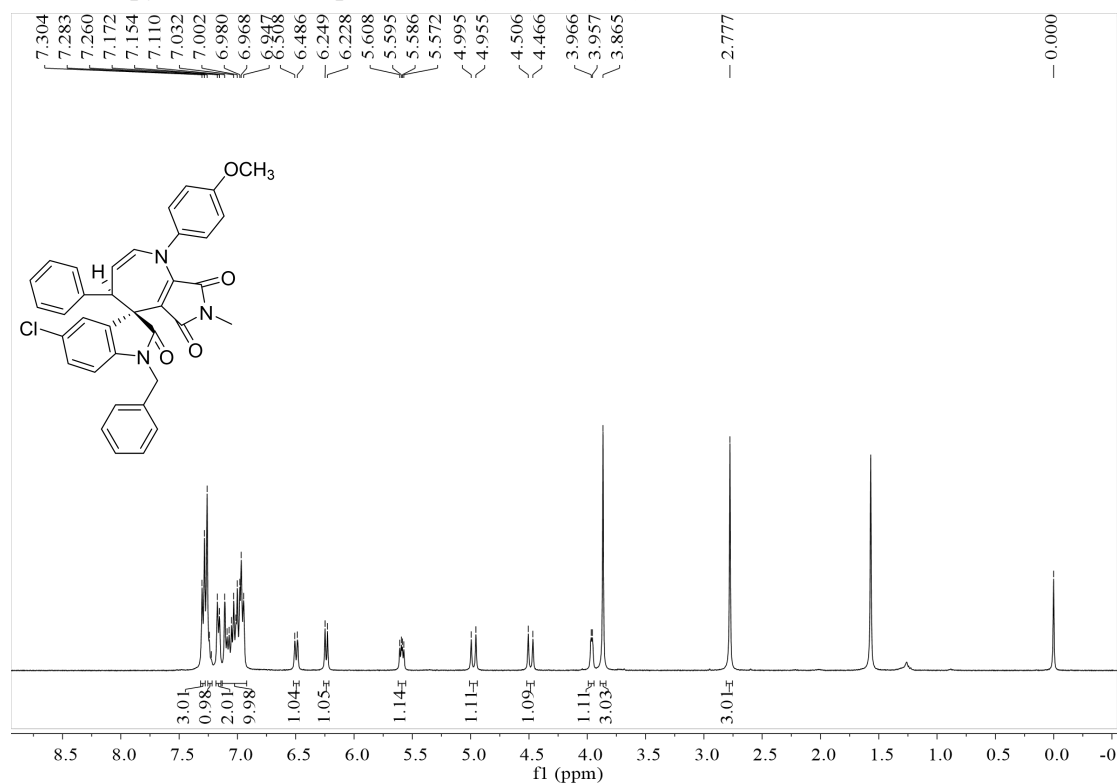
***cis*-1-benzyl-1'-(4-methoxyphenyl)-5,7'-dimethyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3h):**

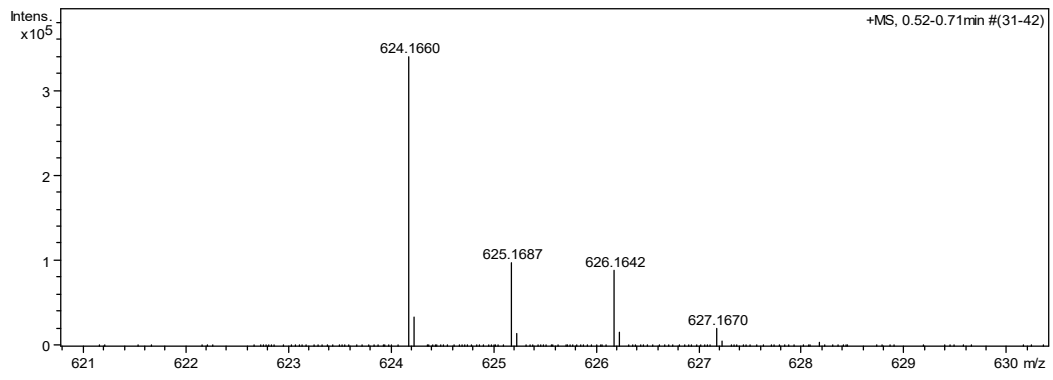




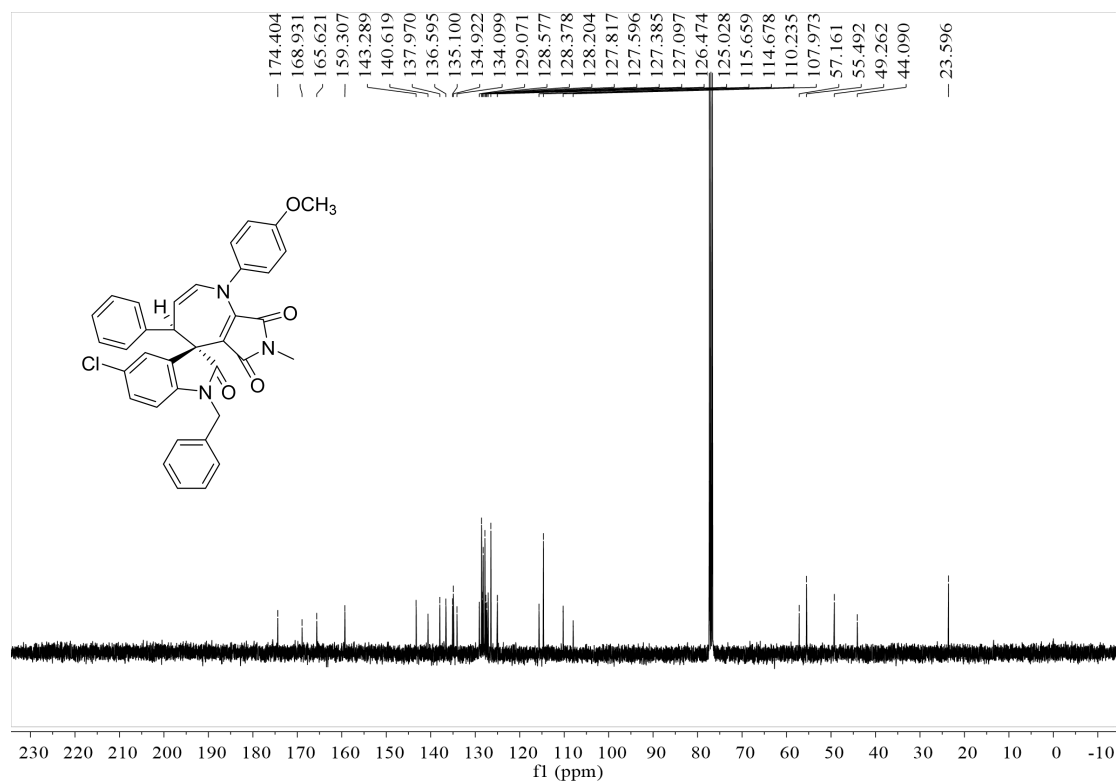
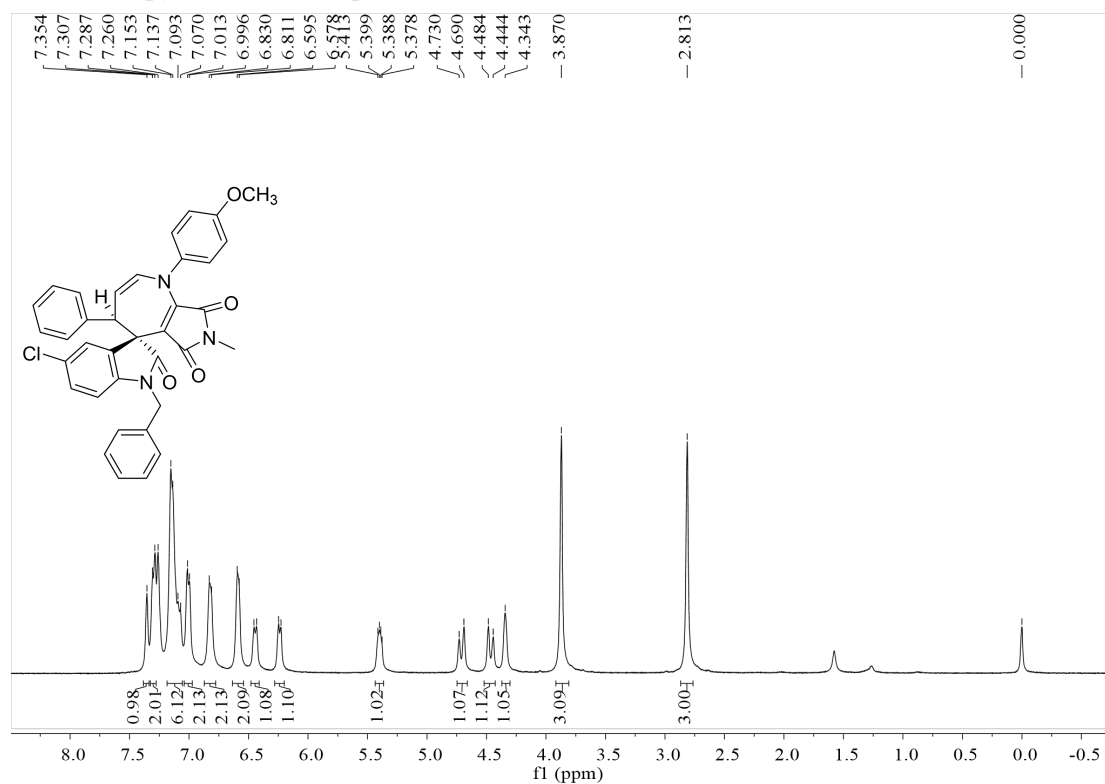


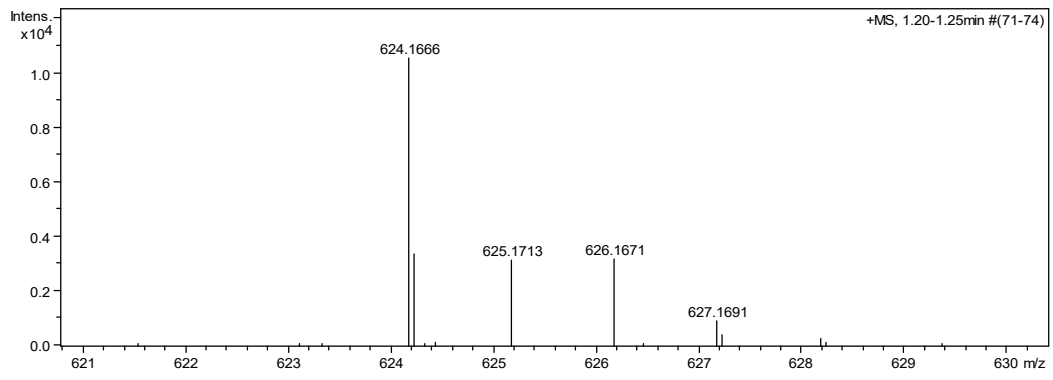
***cis*-1-benzyl-5-chloro-1'-(4-methoxyphenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6,8'(7'*H*)-trione (3i):**



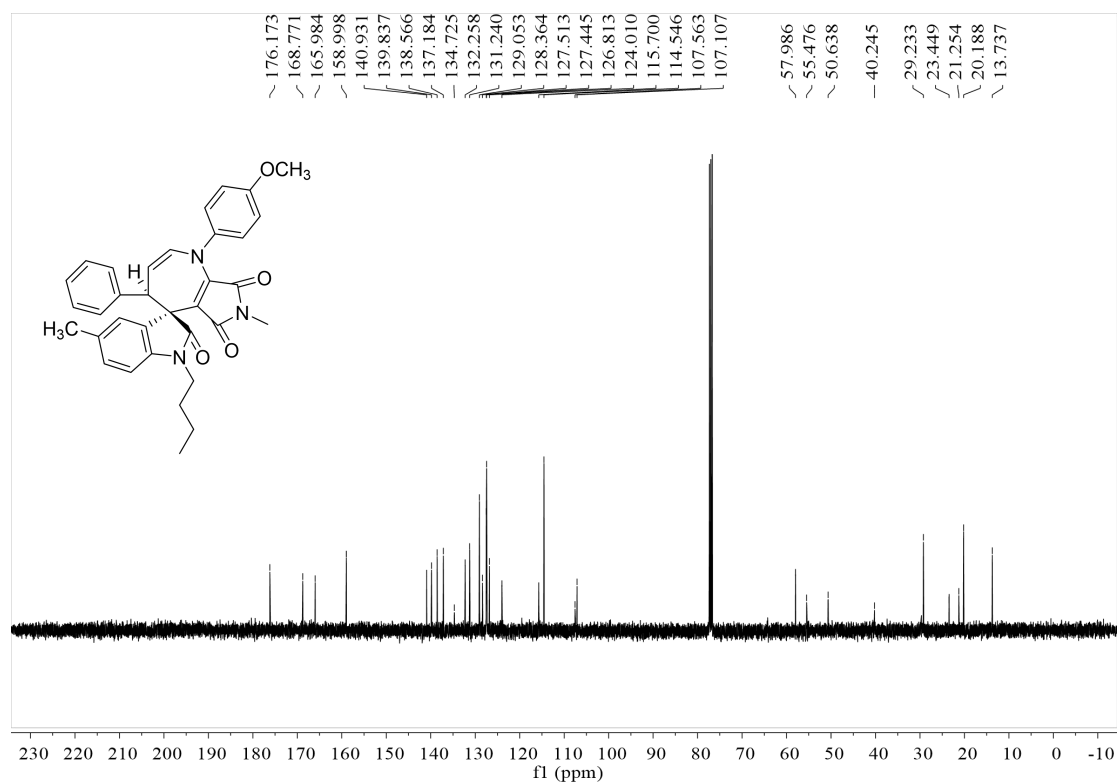
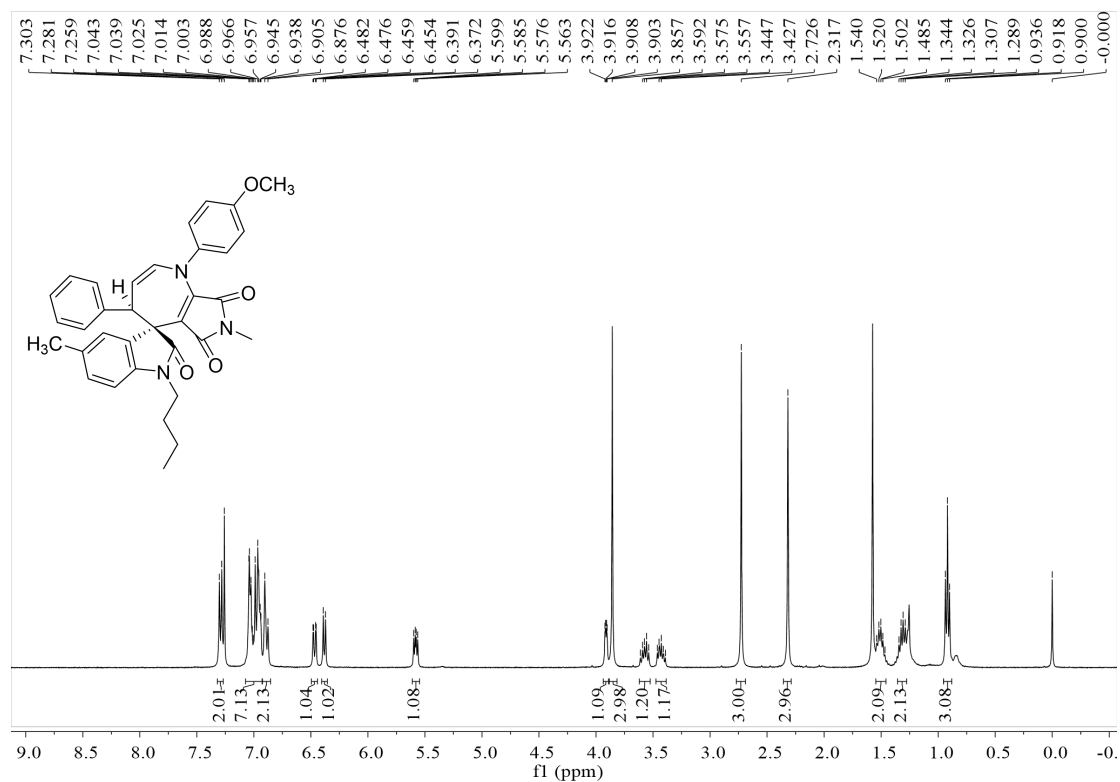


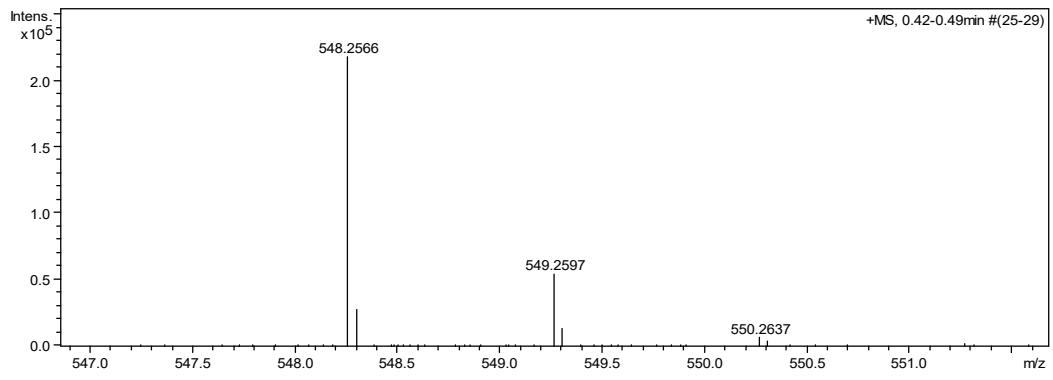
***trans*-1-benzyl-5-chloro-1'-(4-methoxyphenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[*indoline-3,5'*-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3i')**



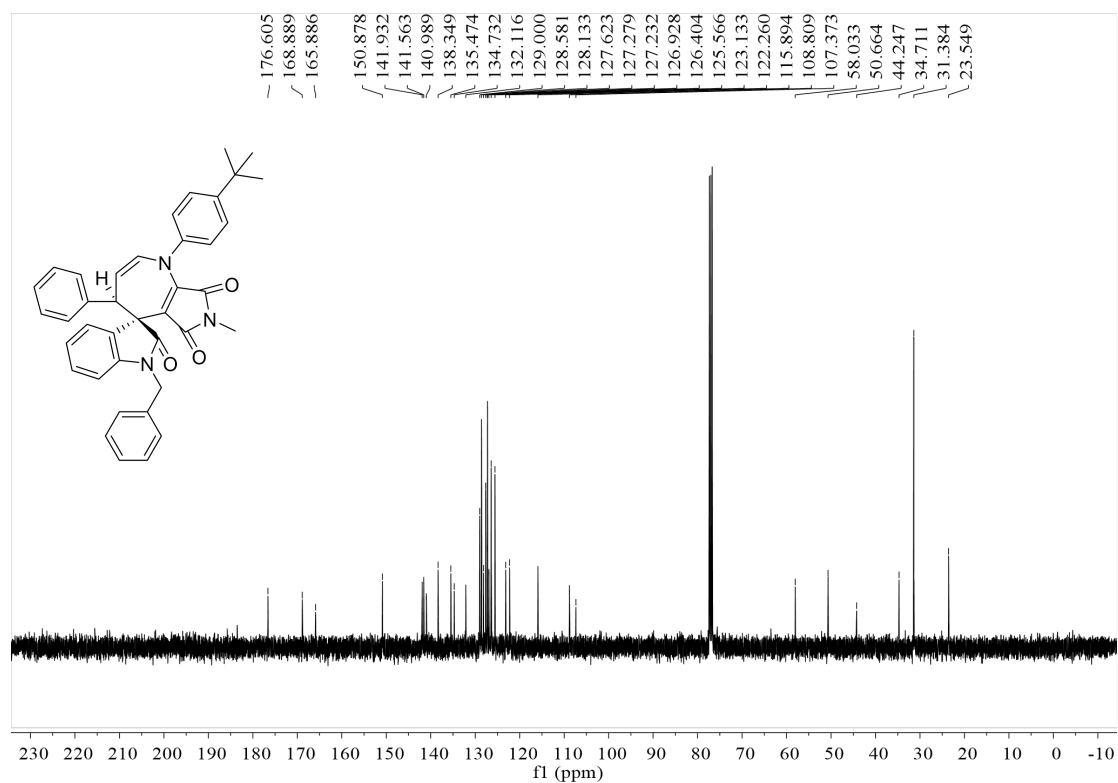
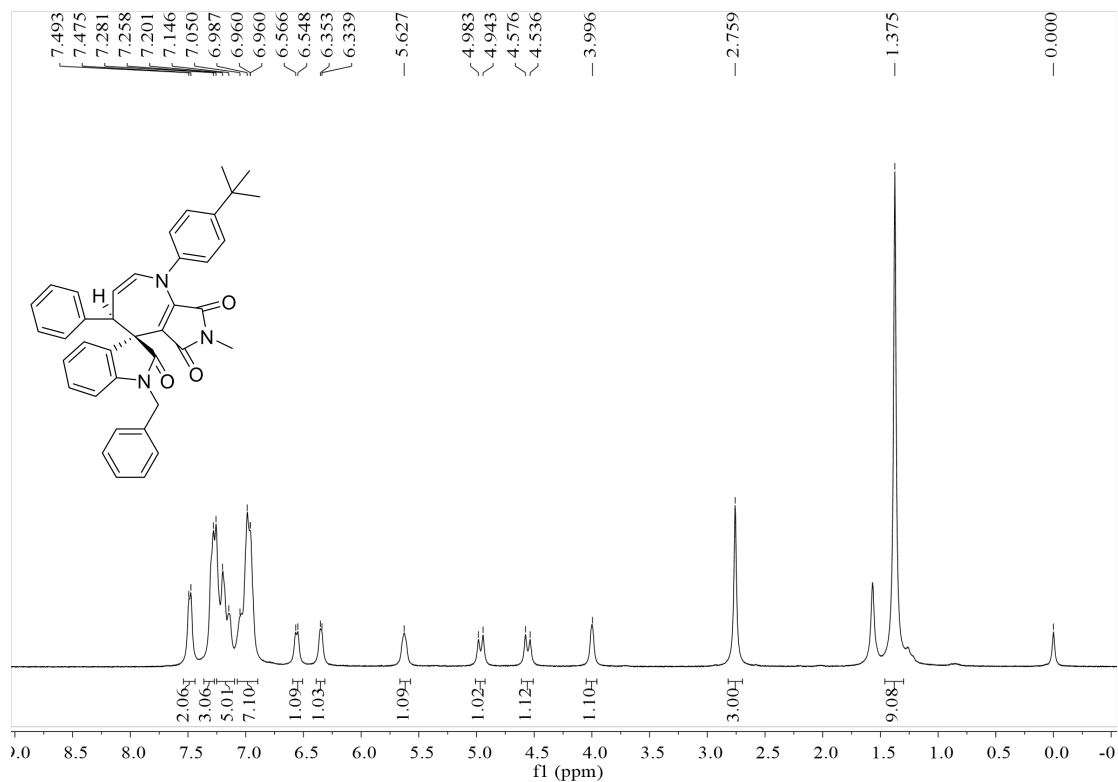


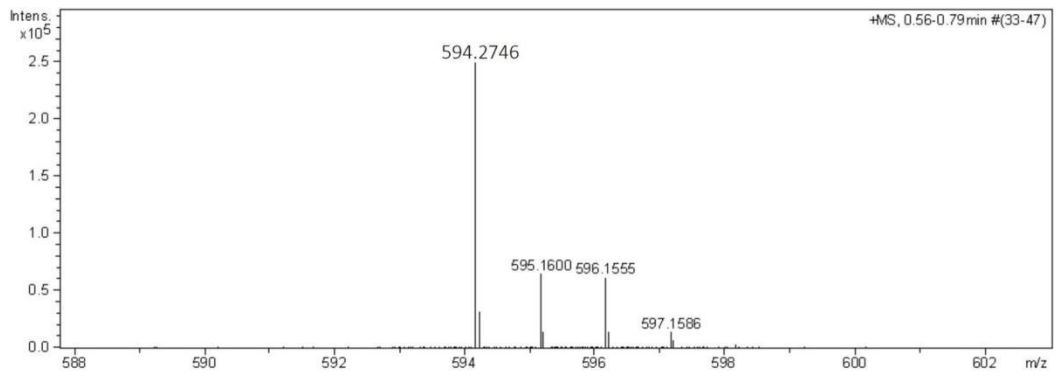
***cis*-1-butyl-1'-(4-methoxyphenyl)-5,7'-dimethyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3j):**





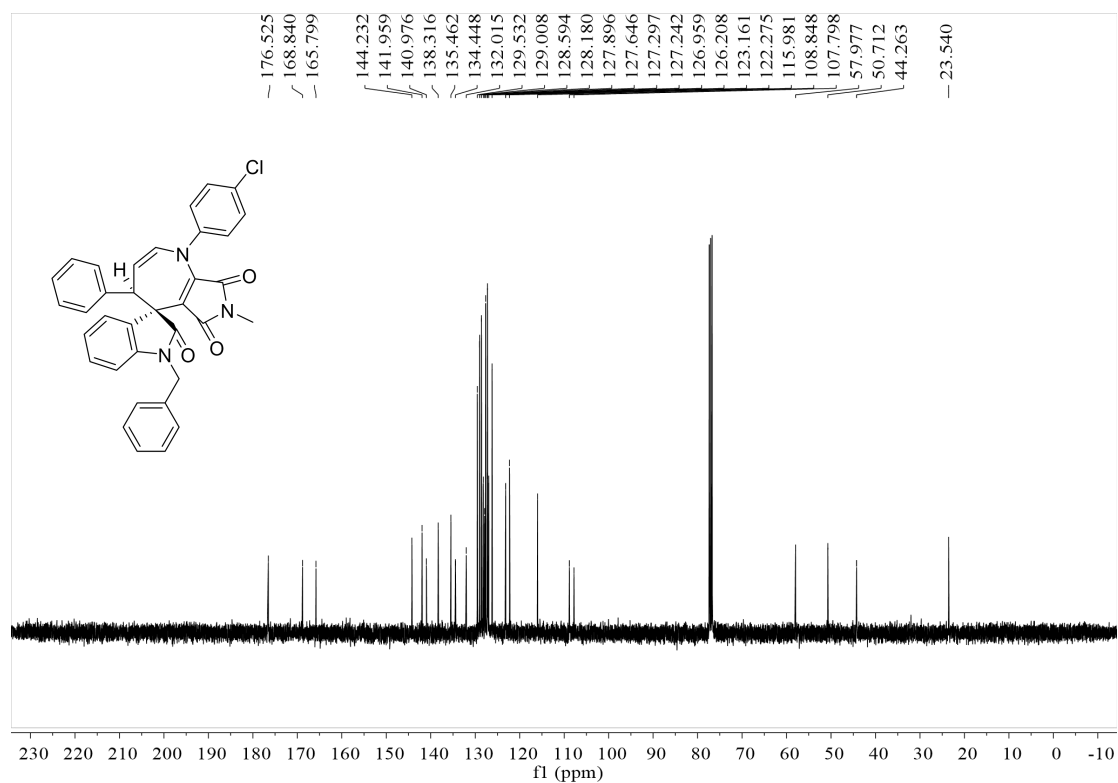
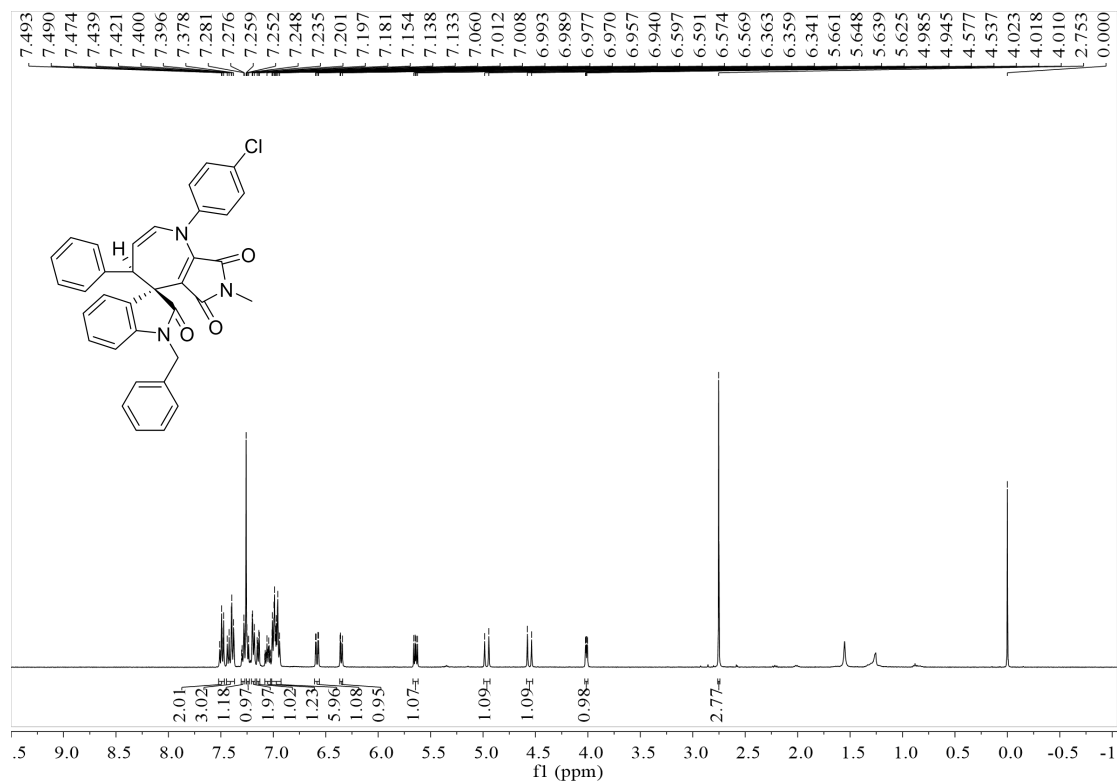
***cis*-1-benzyl-1'-(4-(*tert*-butyl)phenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3k):**

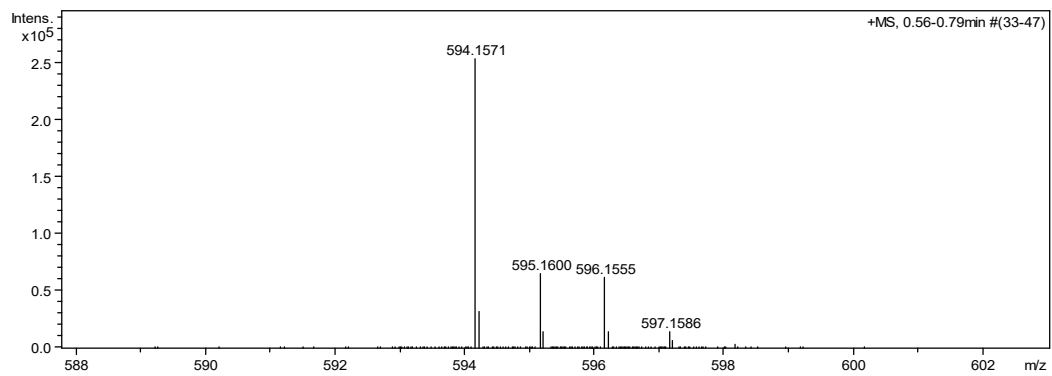




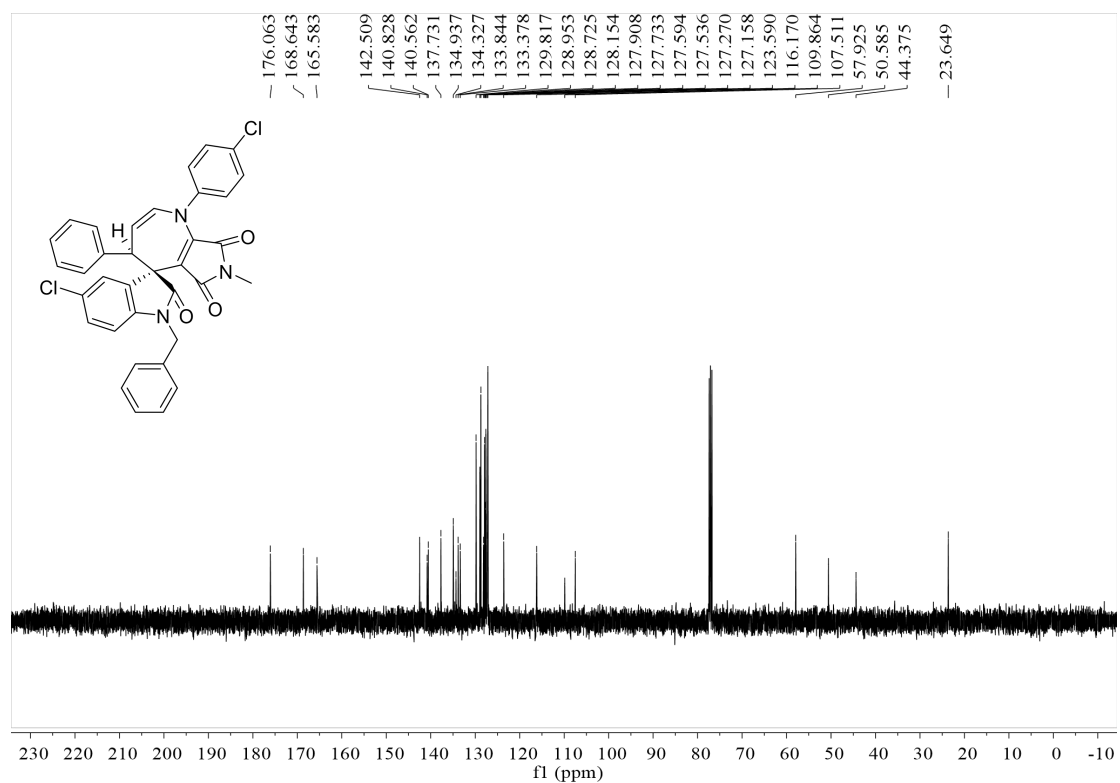
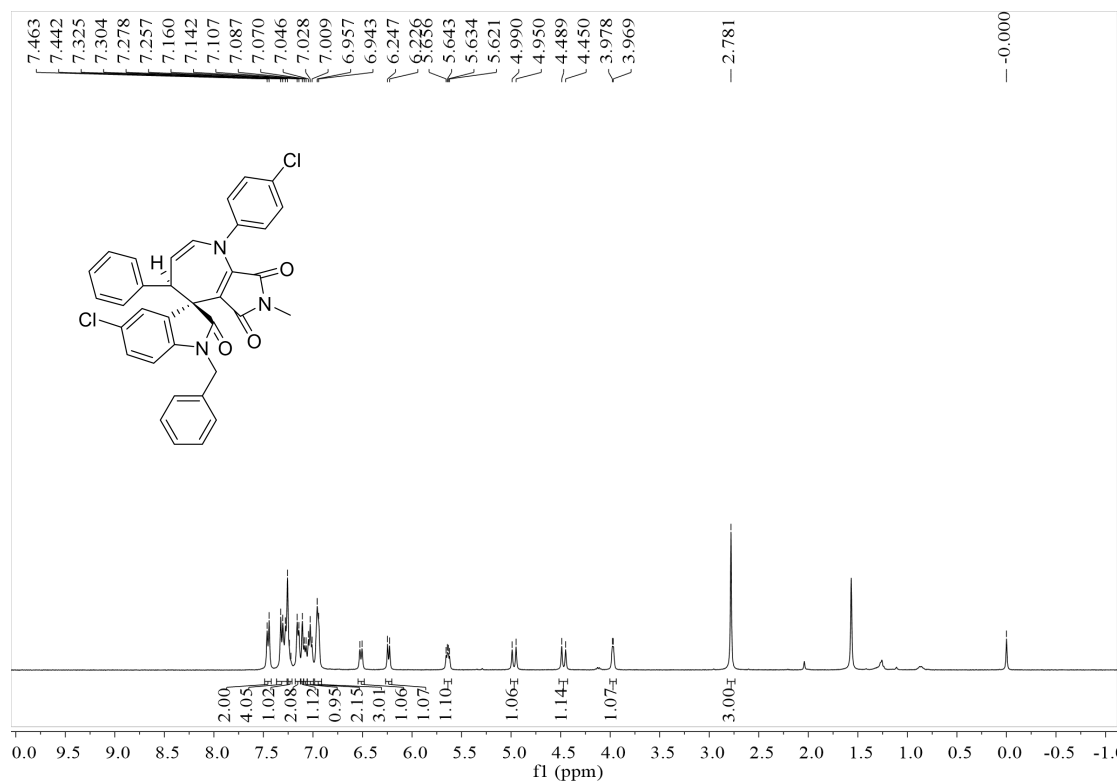


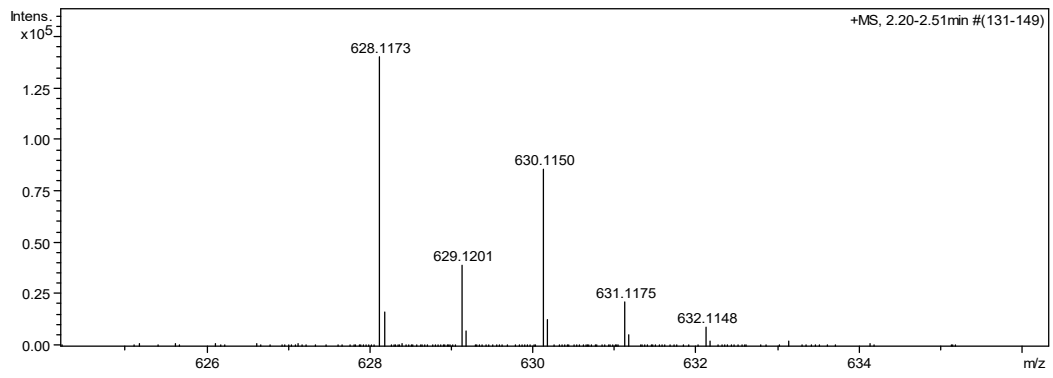
***cis*-1-benzyl-1'-(4-chlorophenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3l):**



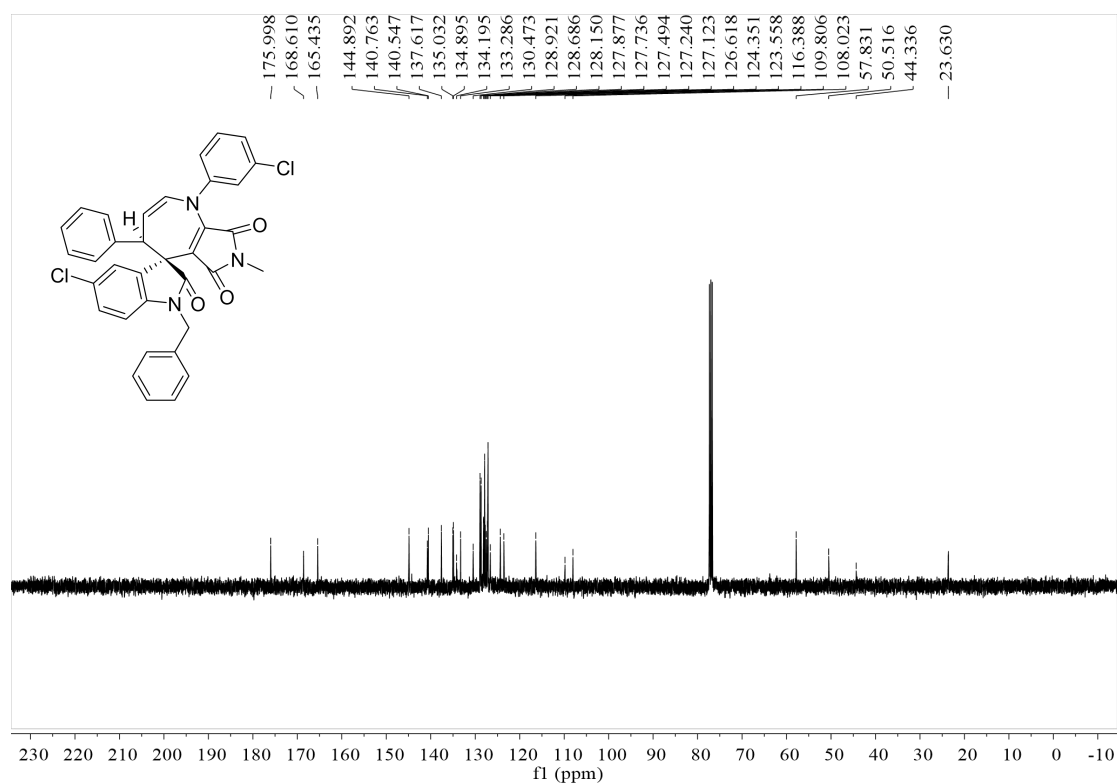
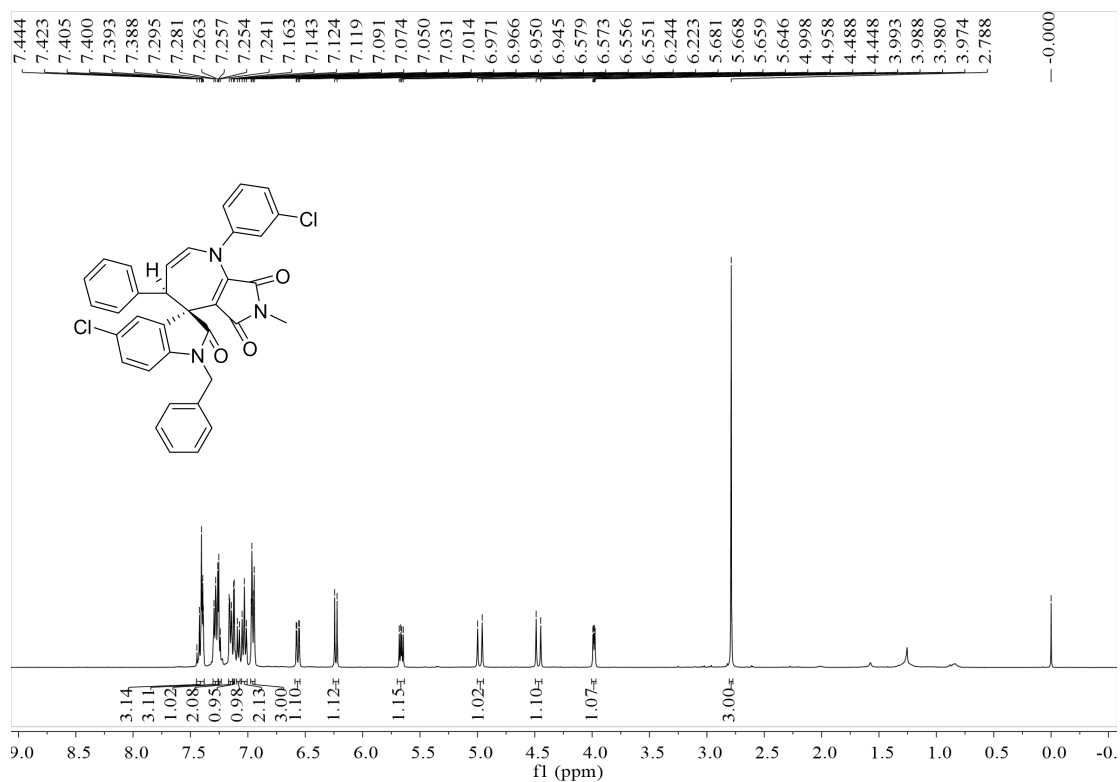


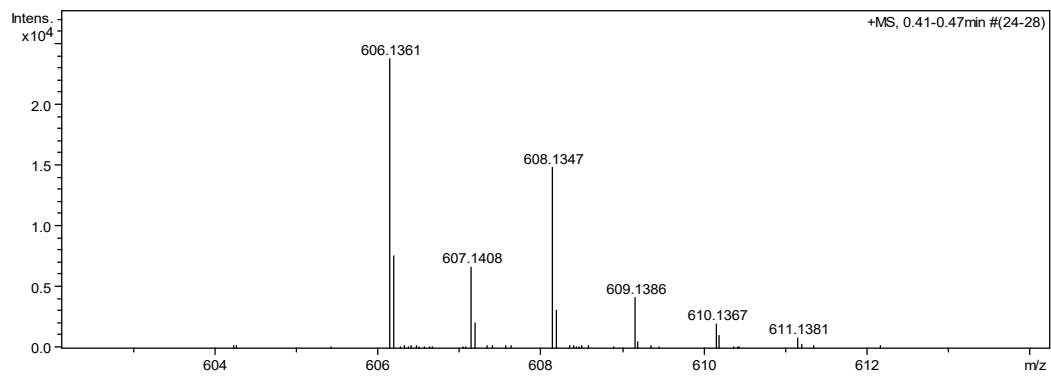
**cis-1-benzyl-5-chloro-1'-(4-chlorophenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6,8'(7'H)-trione (3m):**



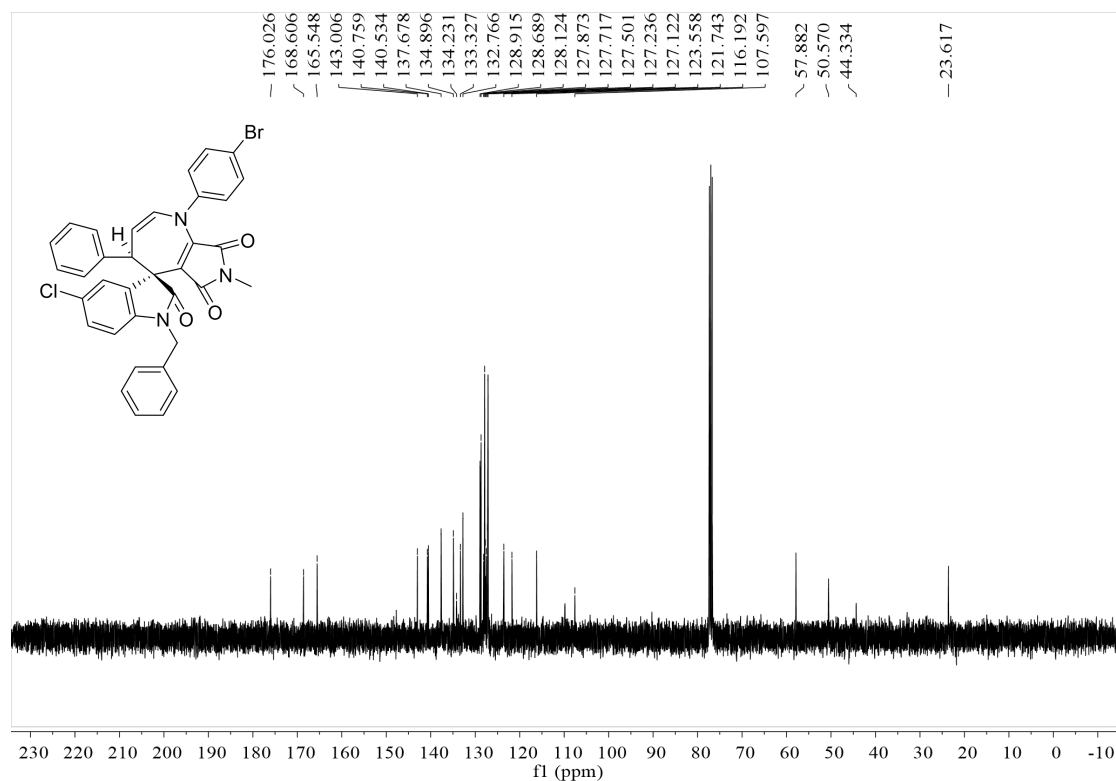
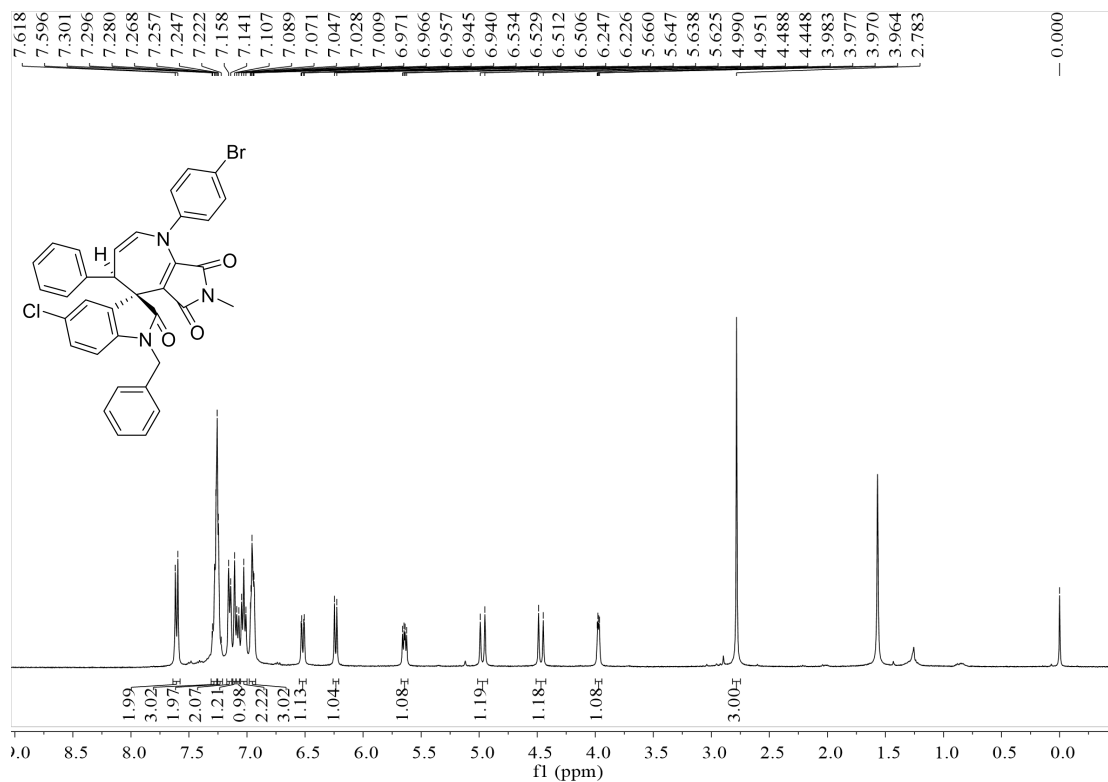


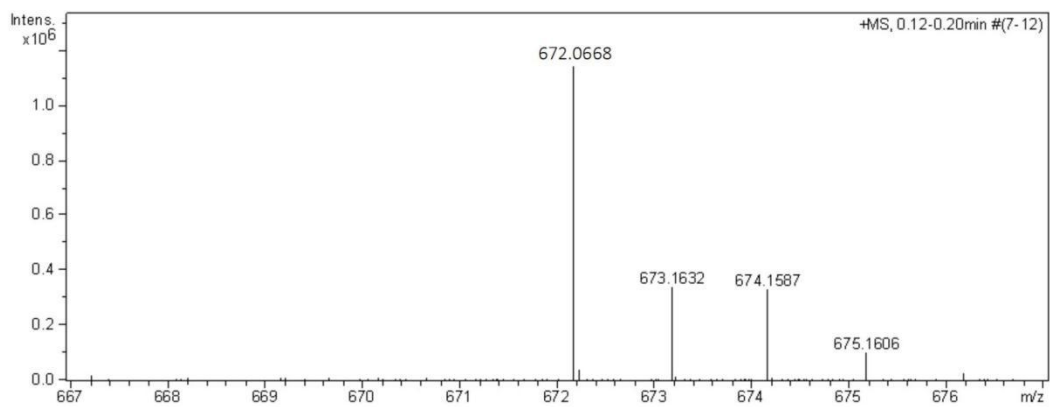
**cis-1-benzyl-5-chloro-1'-(3-chlorophenyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6,8'(7'H)-trione (3n):**





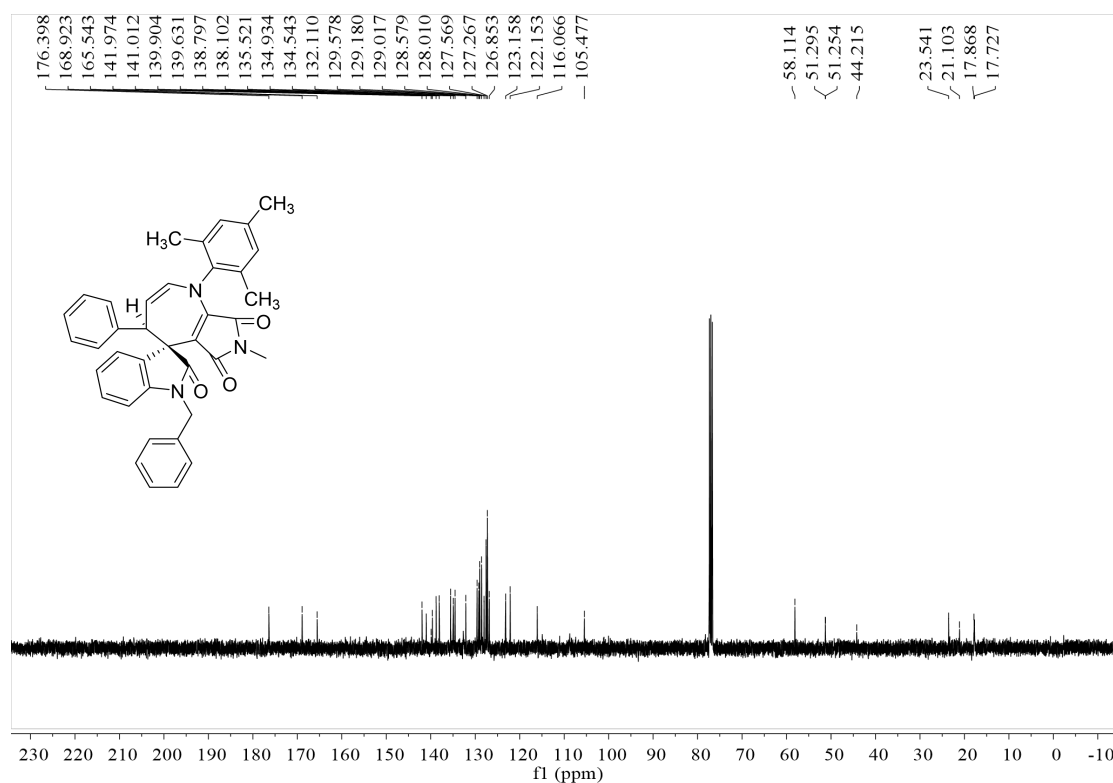
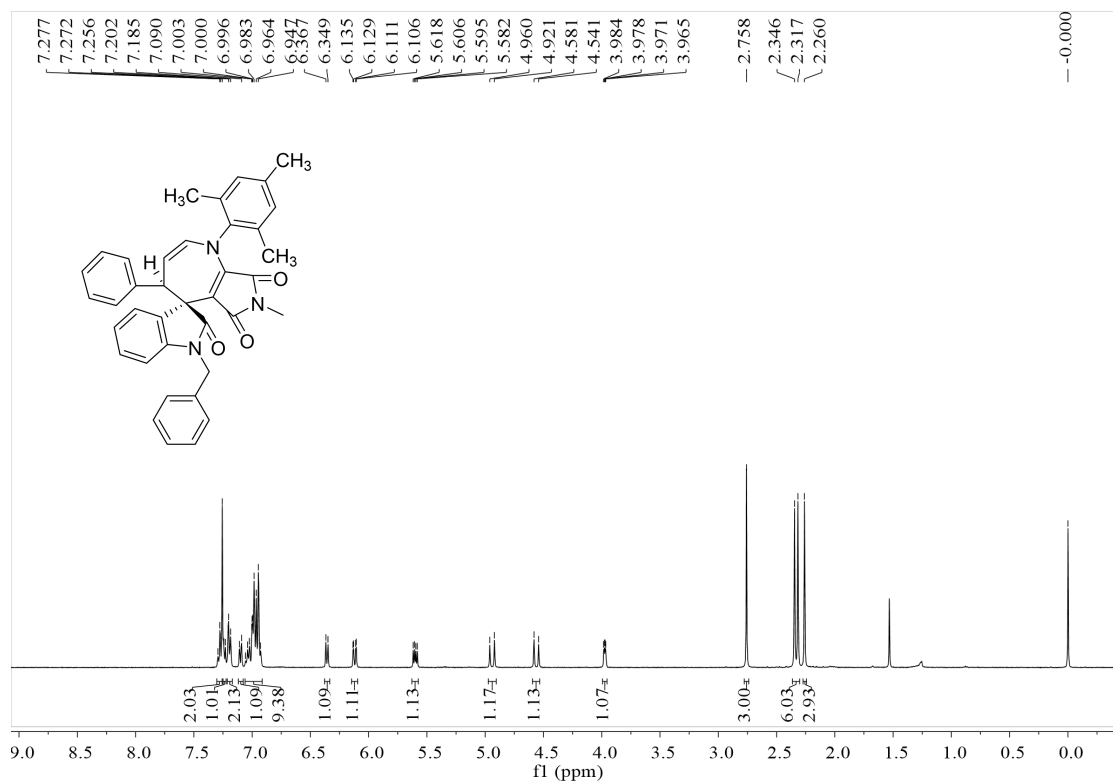
**cis-1-benzyl-1'-(4-bromophenyl)-5-chloro-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6,8'(7'H)-trione (3o):**

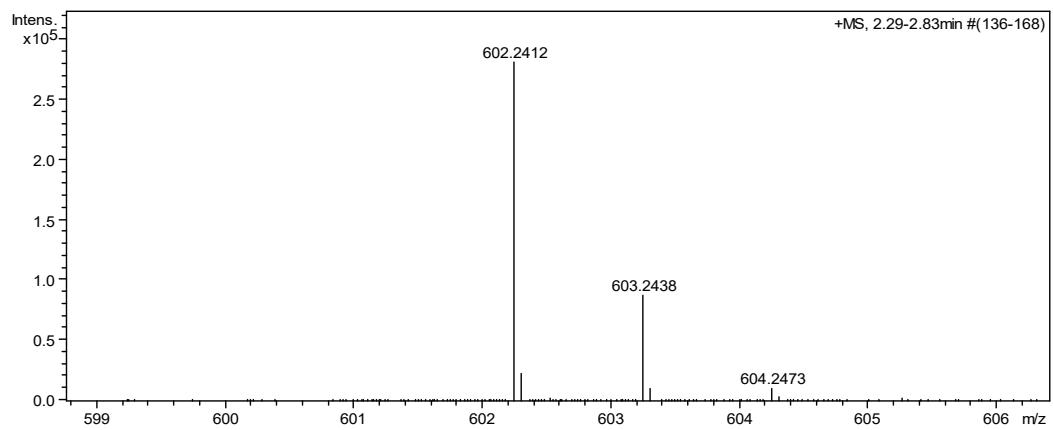




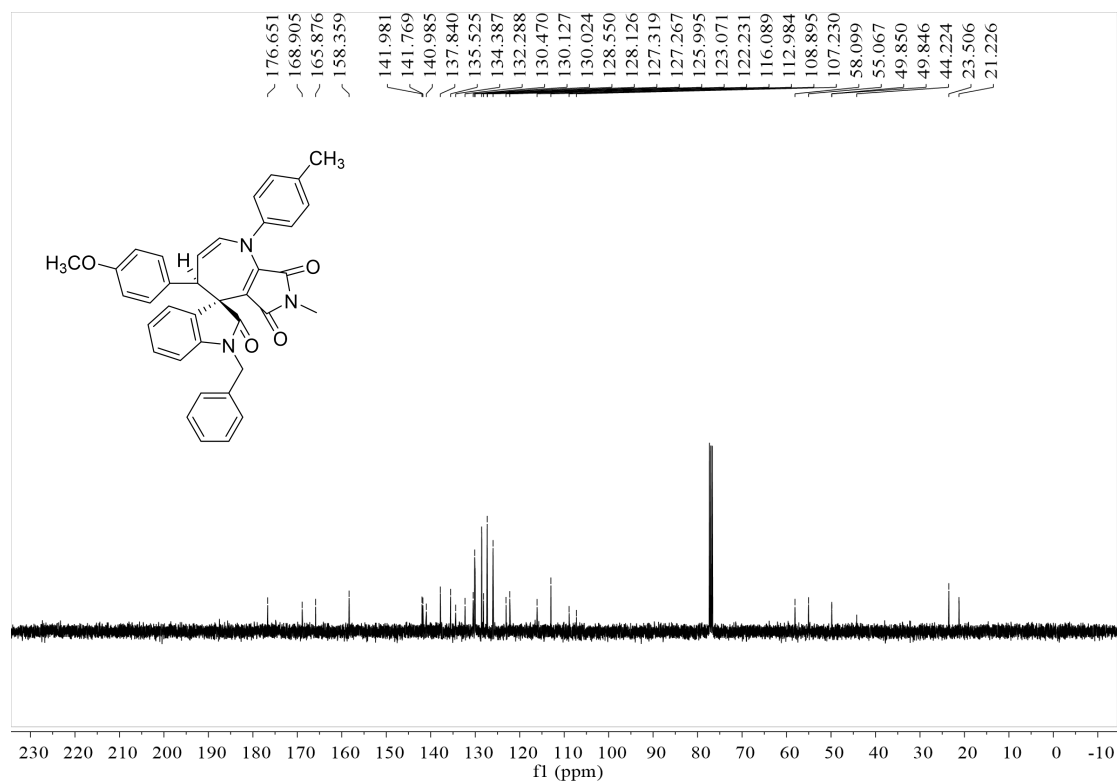
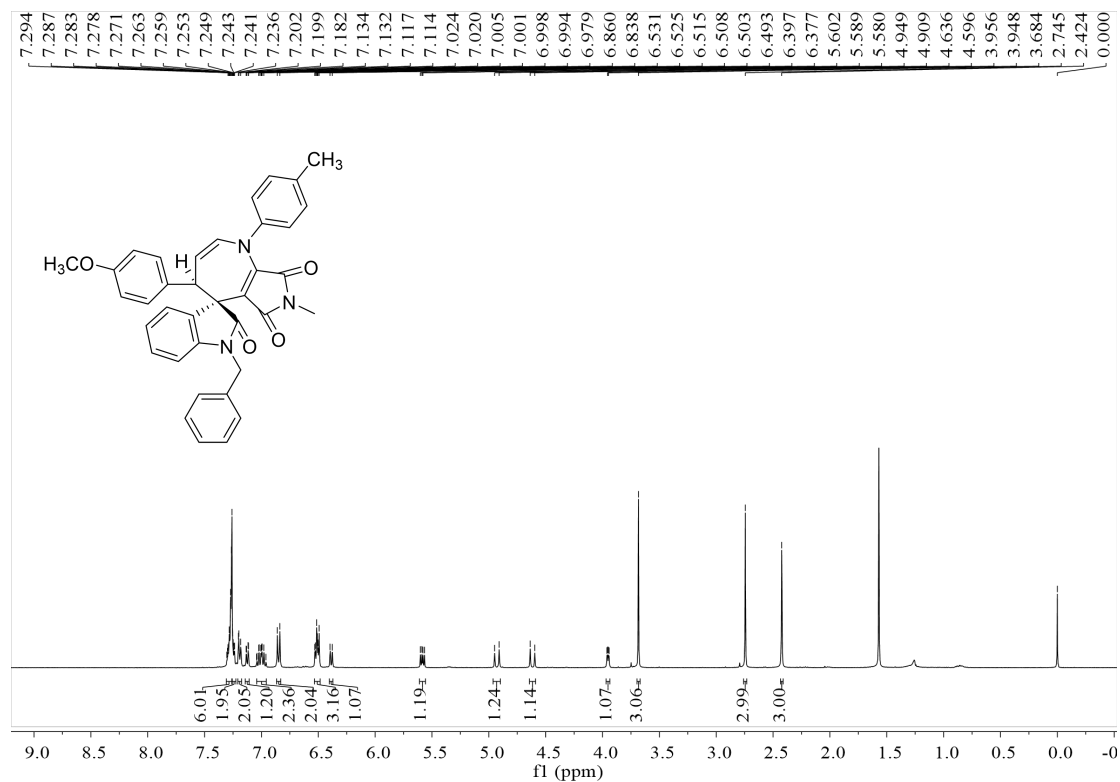


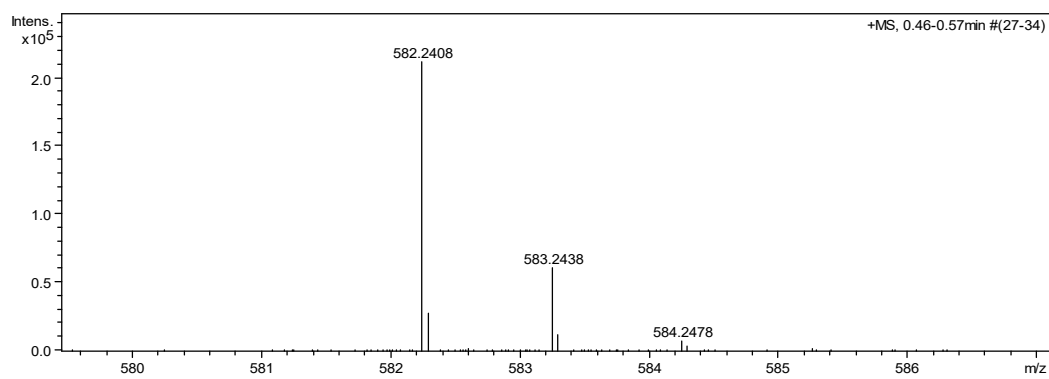
**cis-1-benzyl-1'-mesityl-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (3p):**



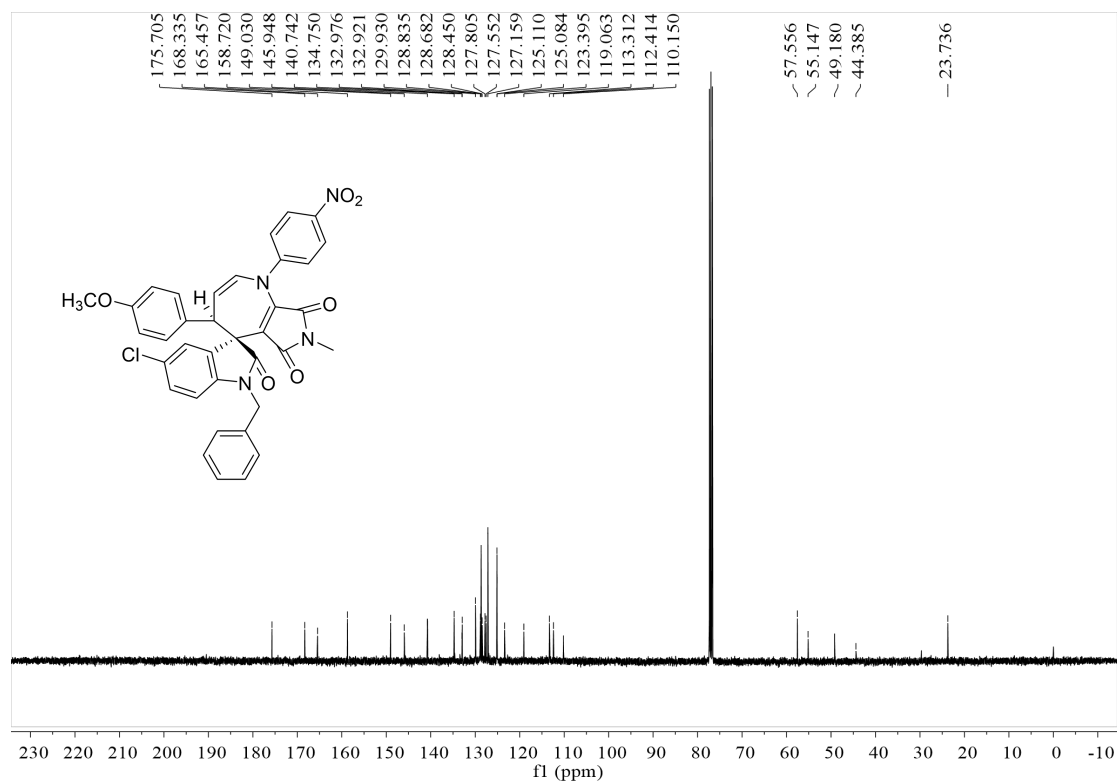
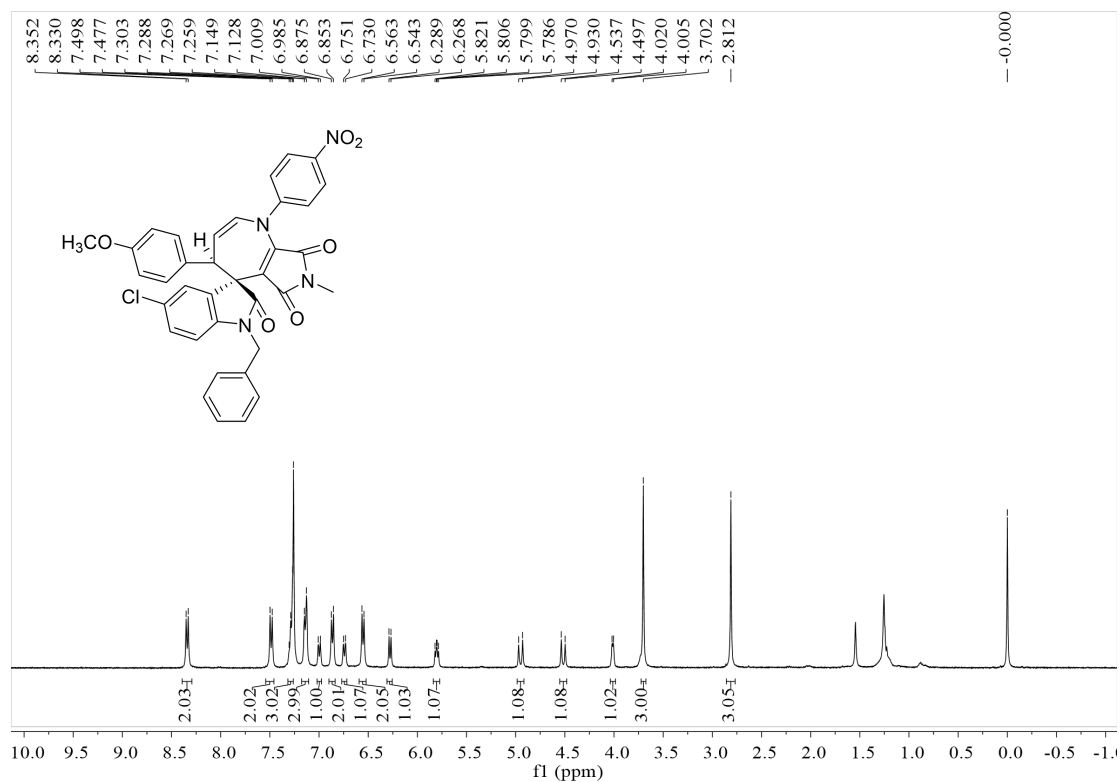


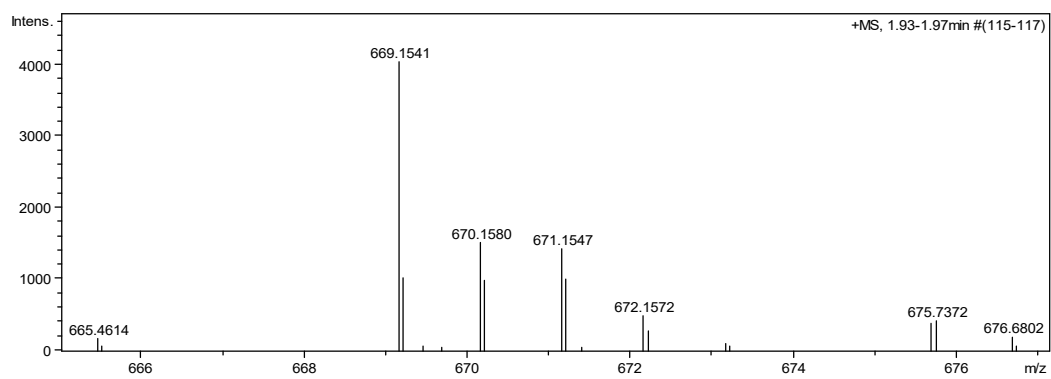
**cis-1-benzyl-4'-(4-methoxyphenyl)-7'-methyl-1'-(p-tolyl)-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (3q):**



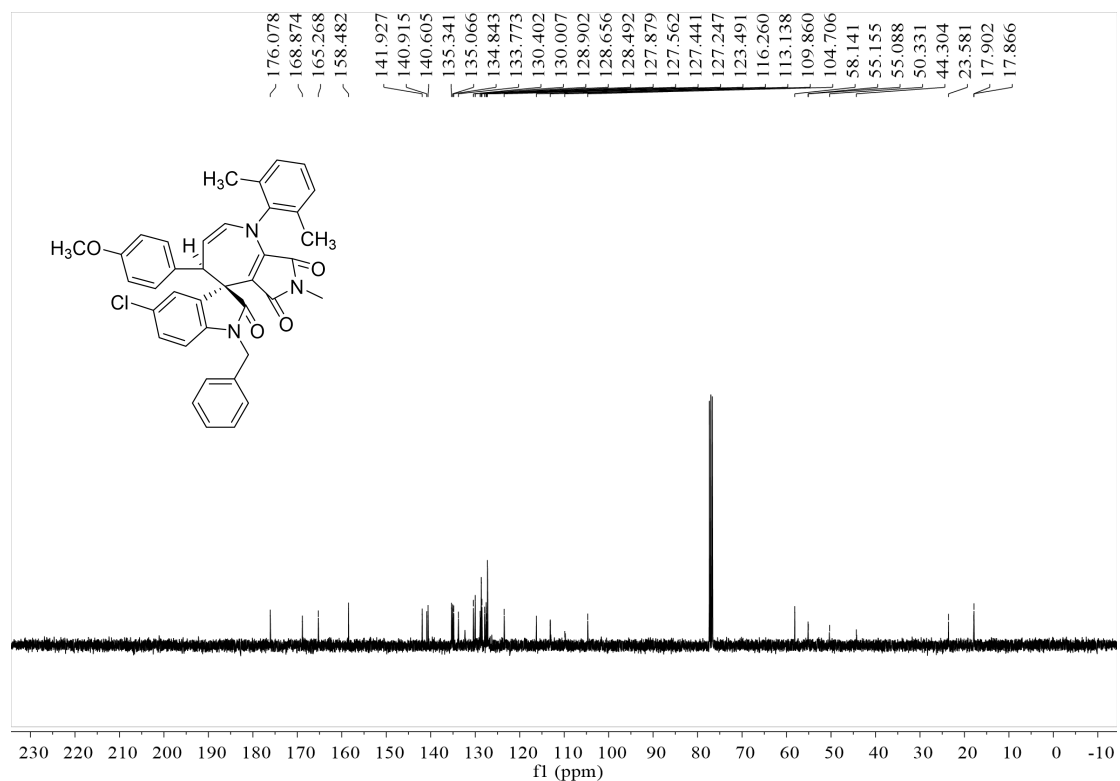
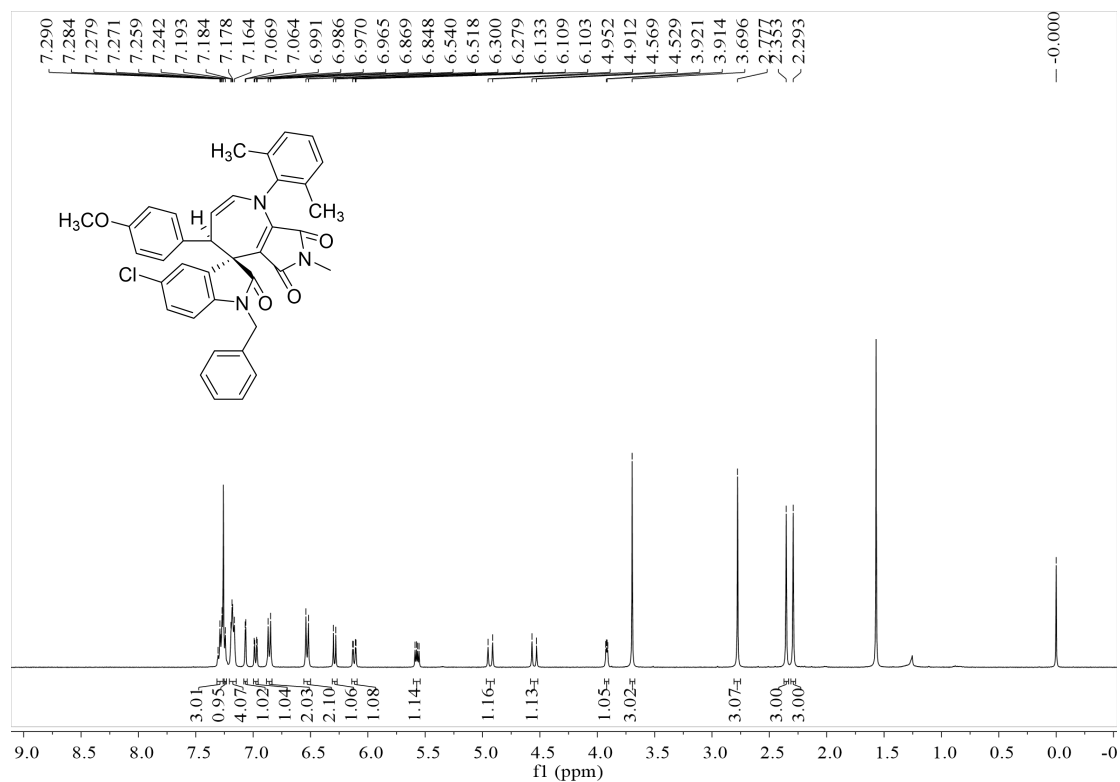


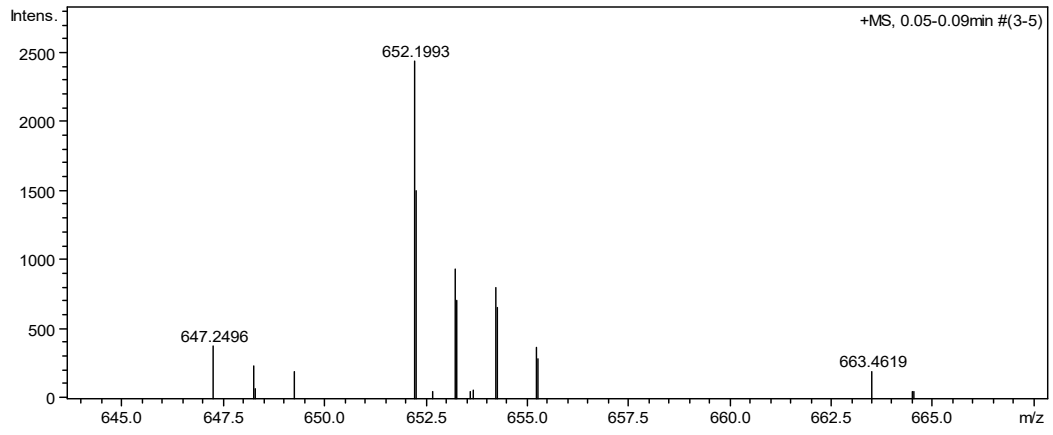
***cis*-1-benzyl-5-chloro-4'-(4-methoxyphenyl)-7'-methyl-1'-(4-nitrophenyl)-1',4'-dihydro-6'*H*-s piro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3r):**





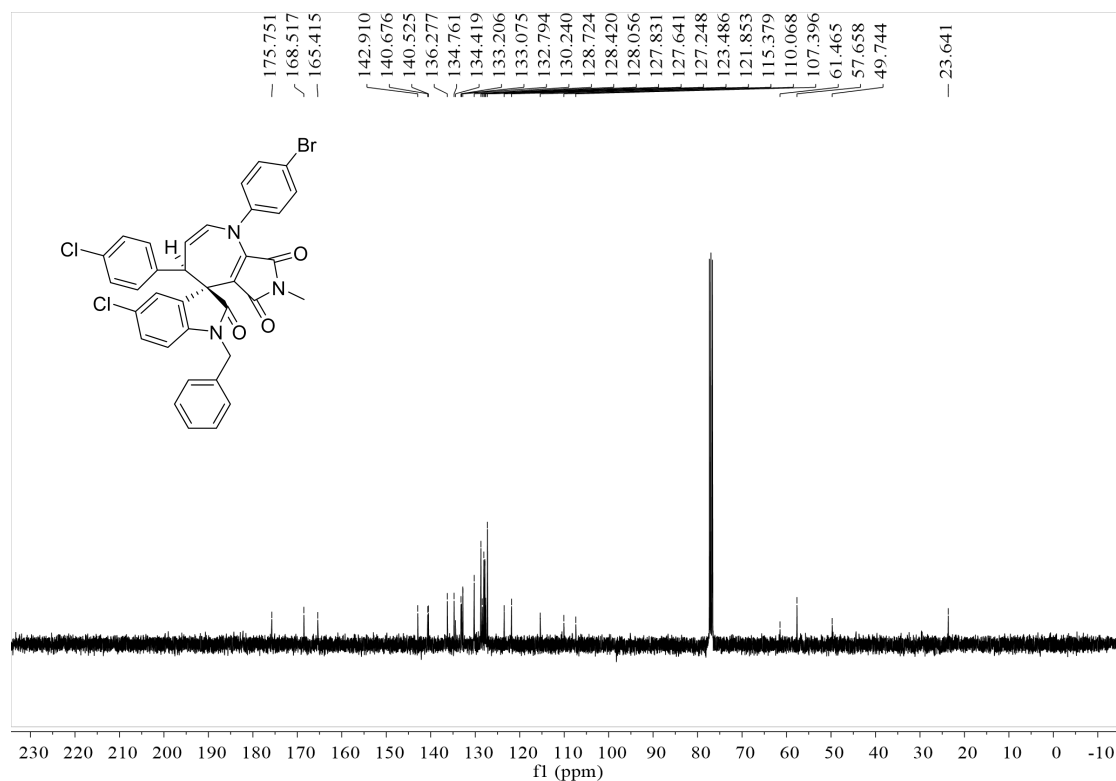
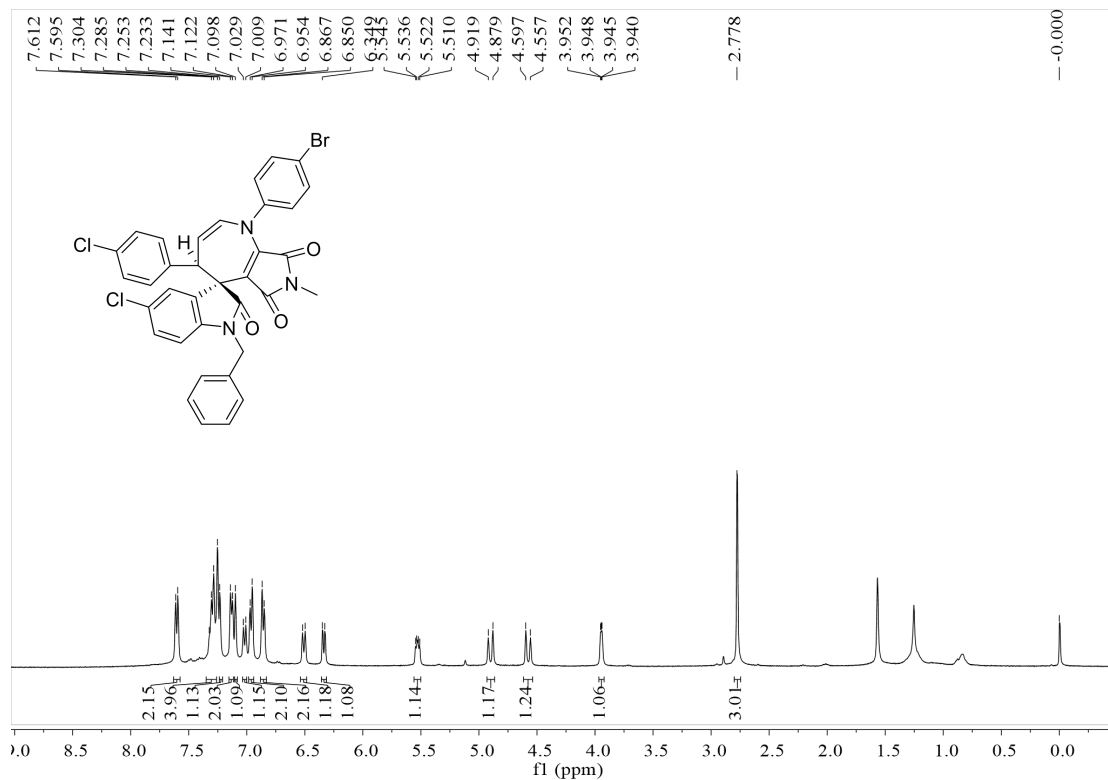
***cis*-1-benzyl-5-chloro-1'-(2,6-dimethylphenyl)-4'-(4-methoxyphenyl)-7'-methyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3s):**

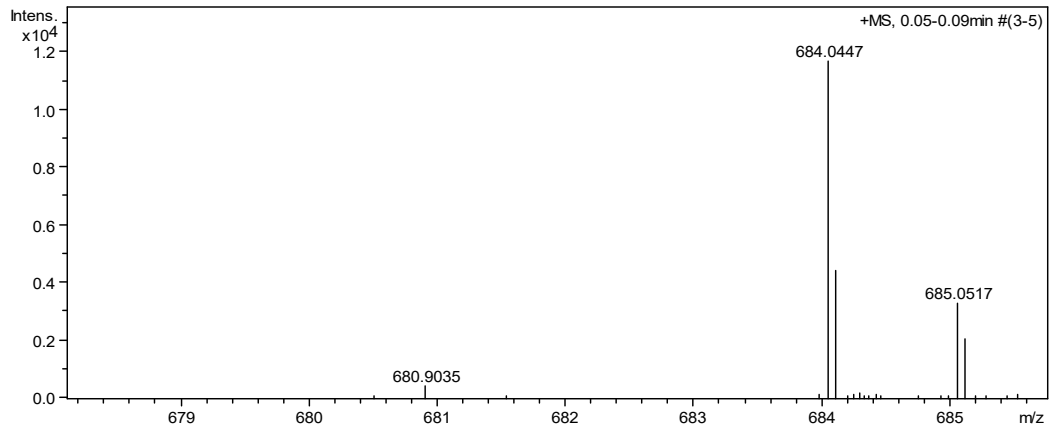




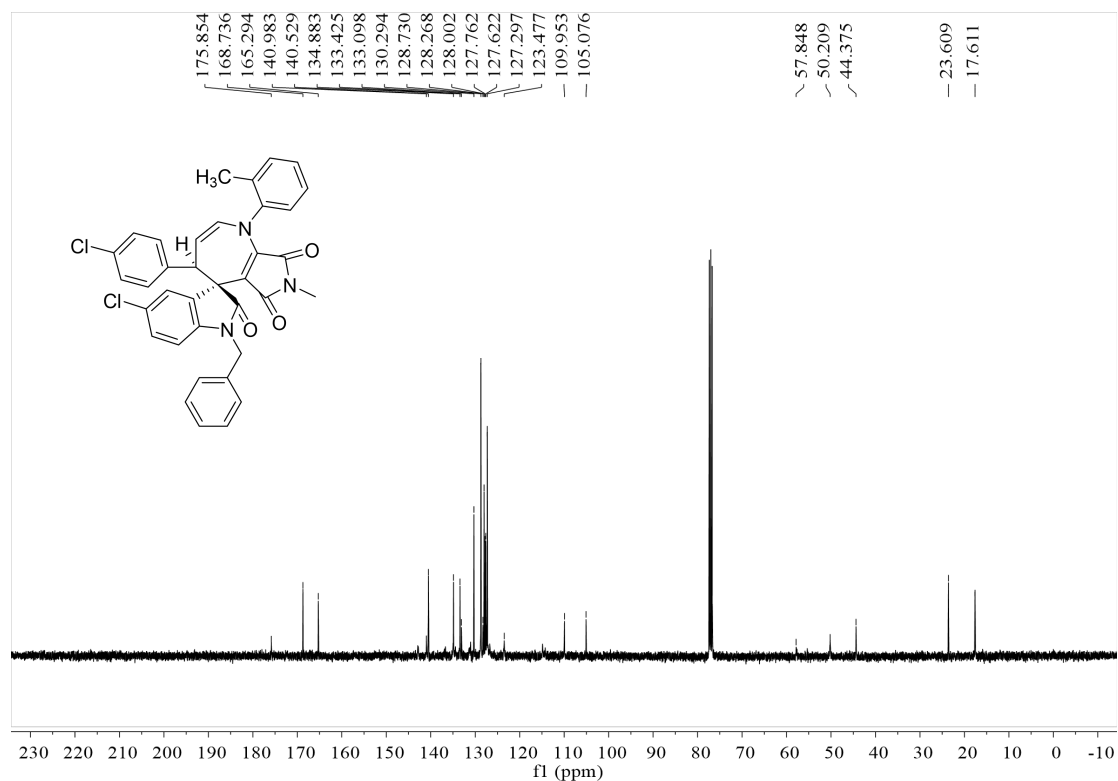
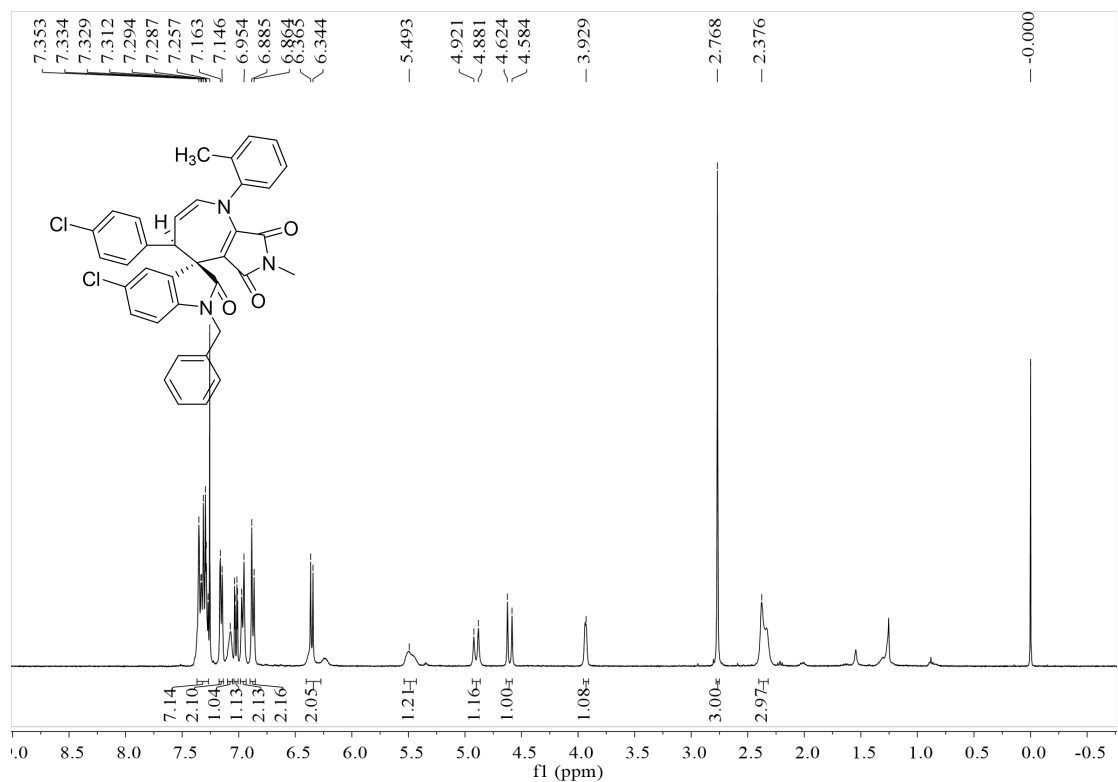


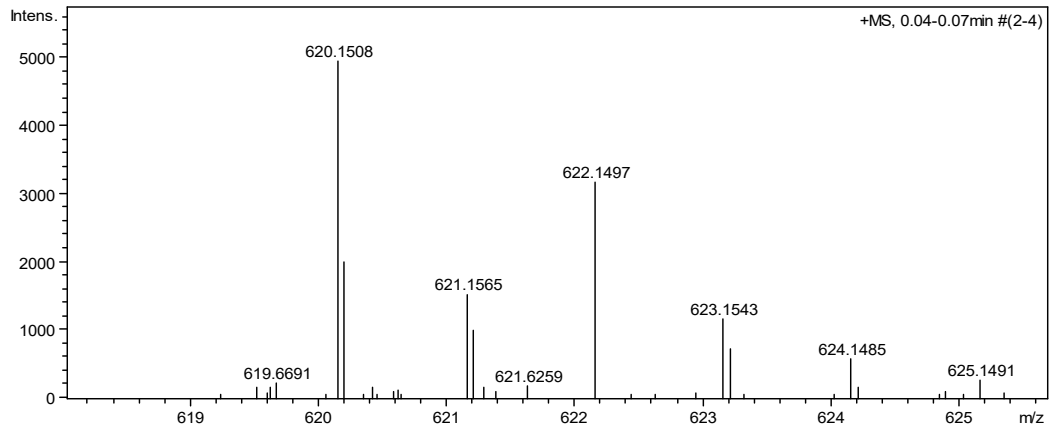
**cis-1-benzyl-1'-(4-bromophenyl)-5-chloro-4'-(4-chlorophenyl)-7'-methyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6,8'(7H)-trione (3t):**



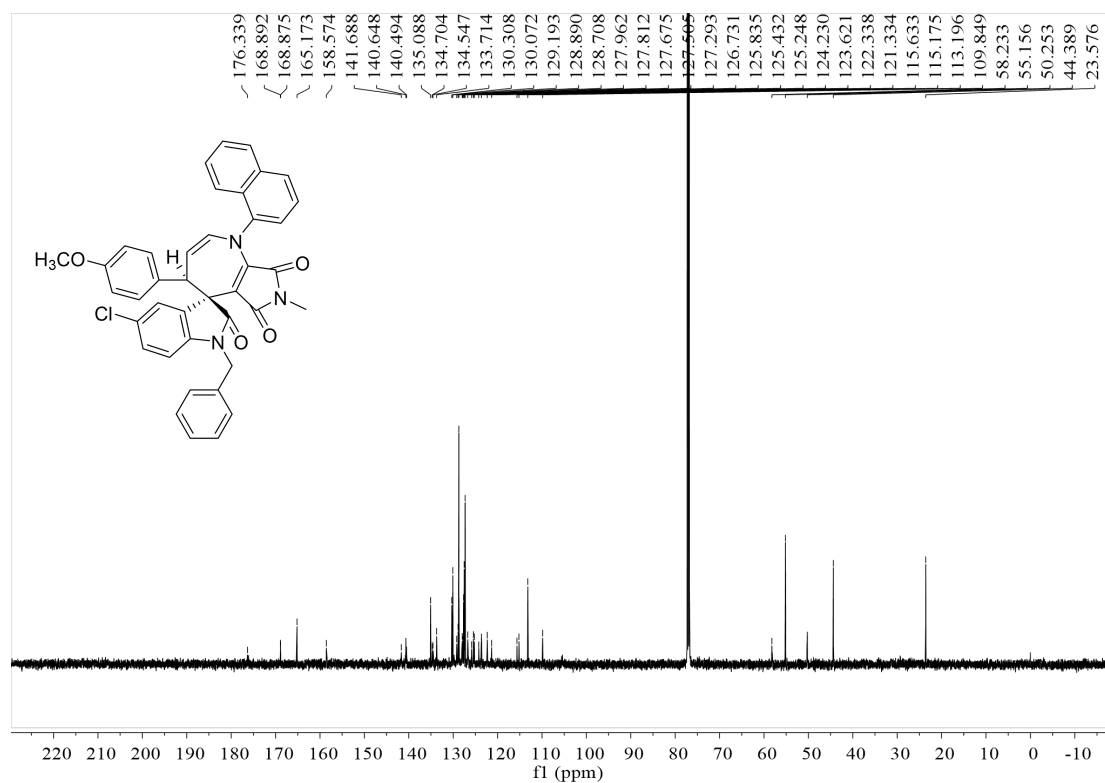
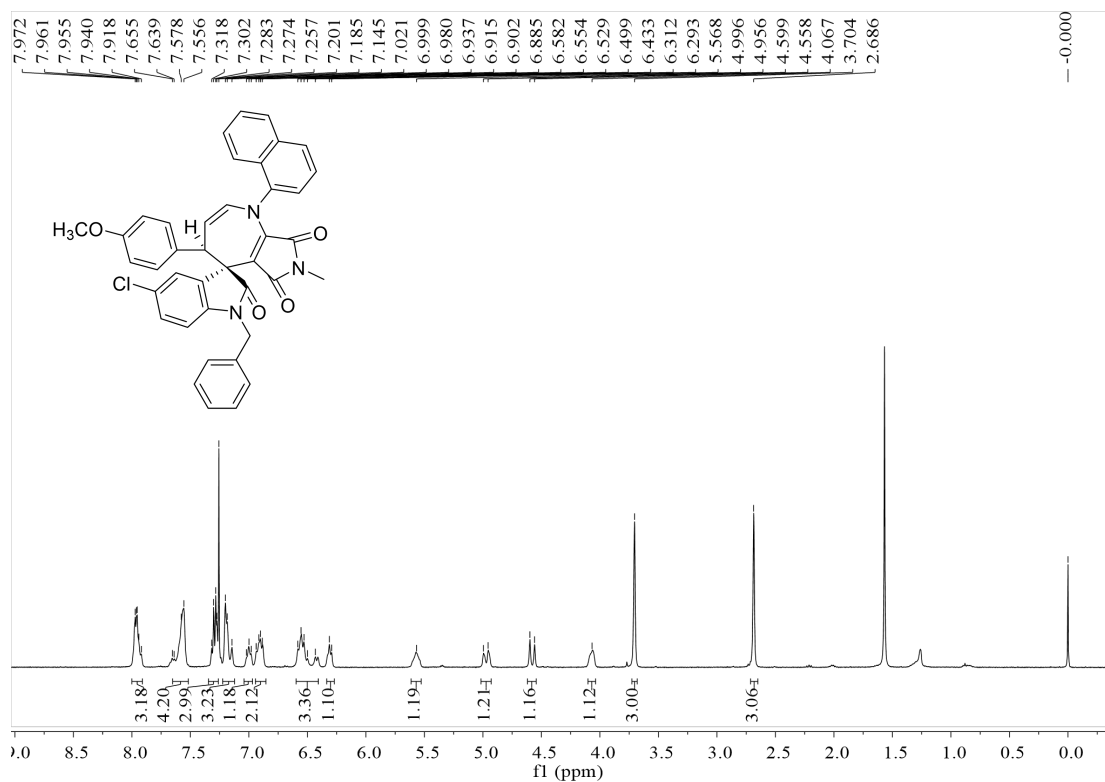


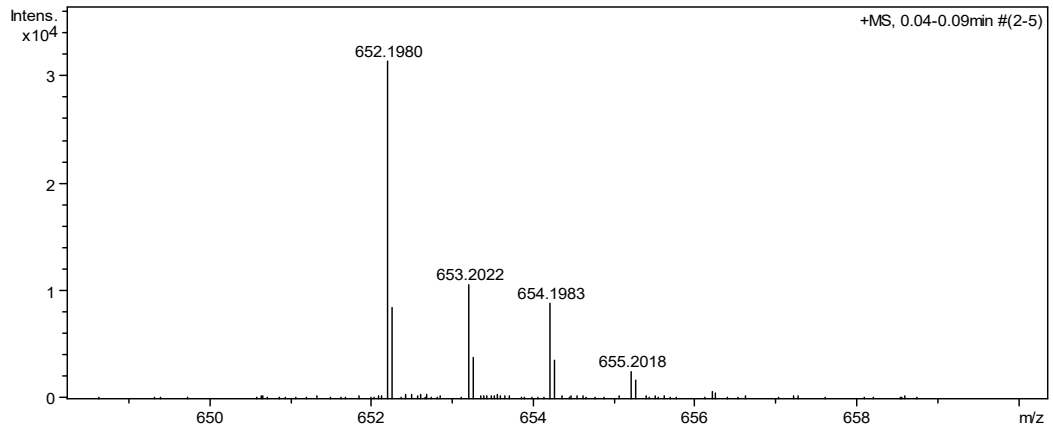
***cis*-1-benzyl-5-chloro-4'-(4-chlorophenyl)-7'-methyl-1'-(*o*-tolyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3u):**



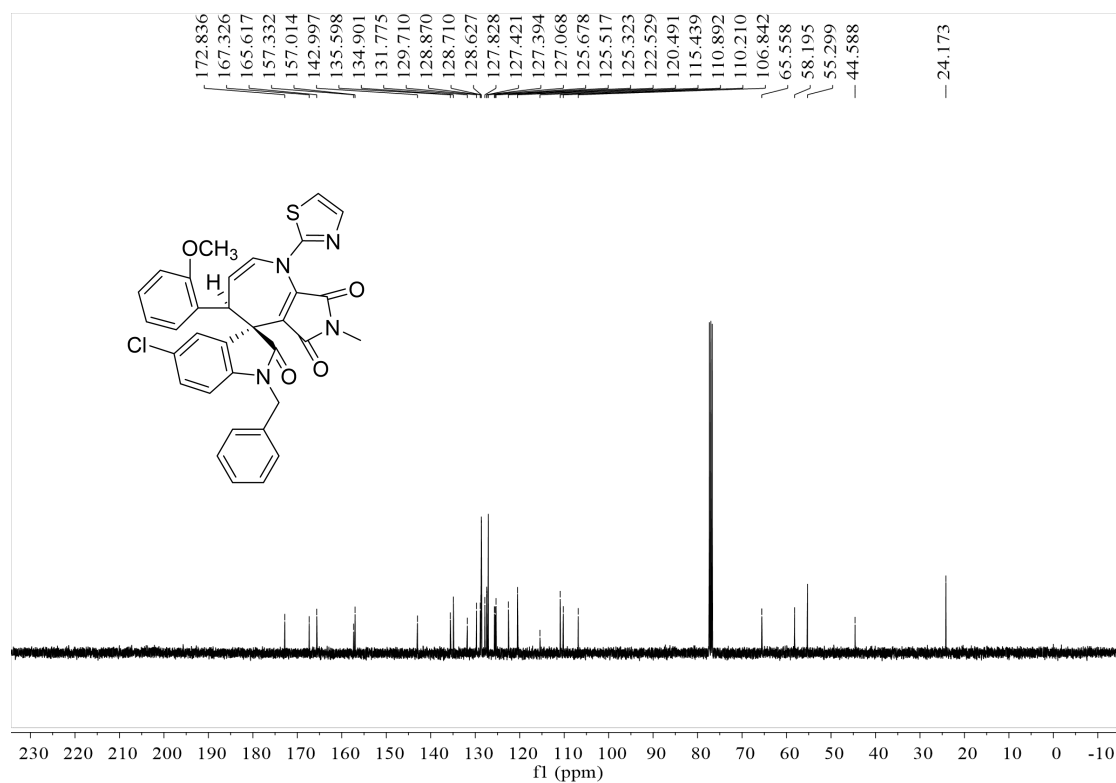
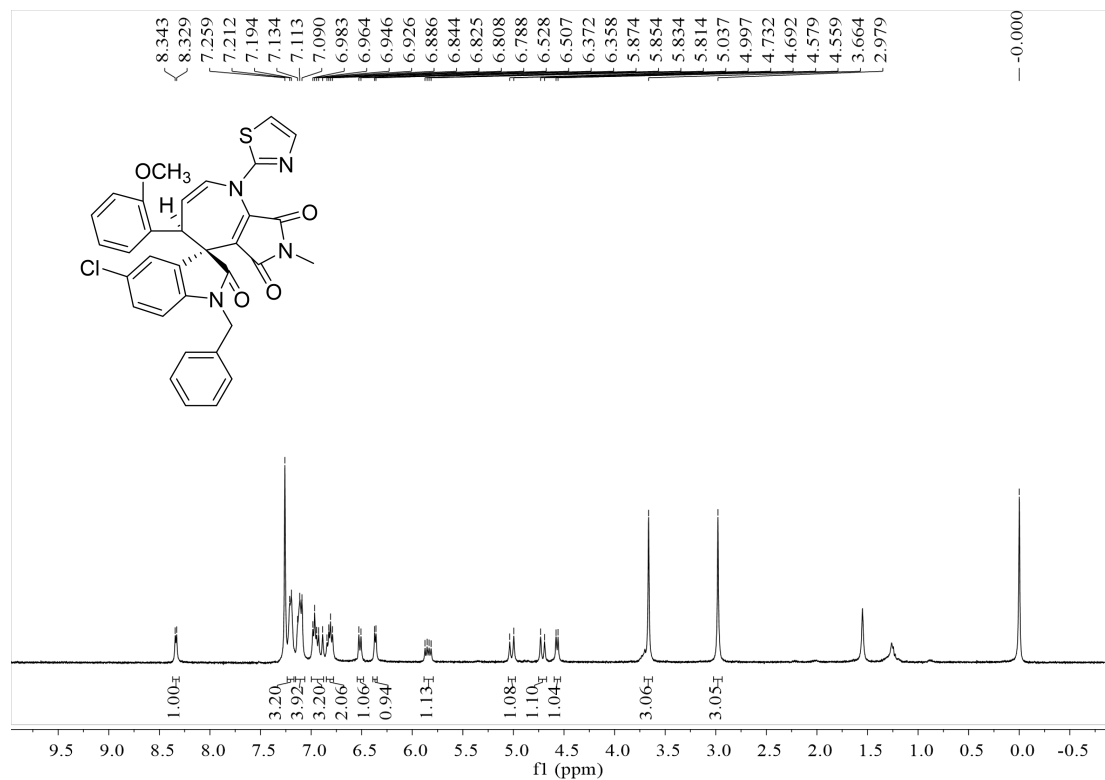


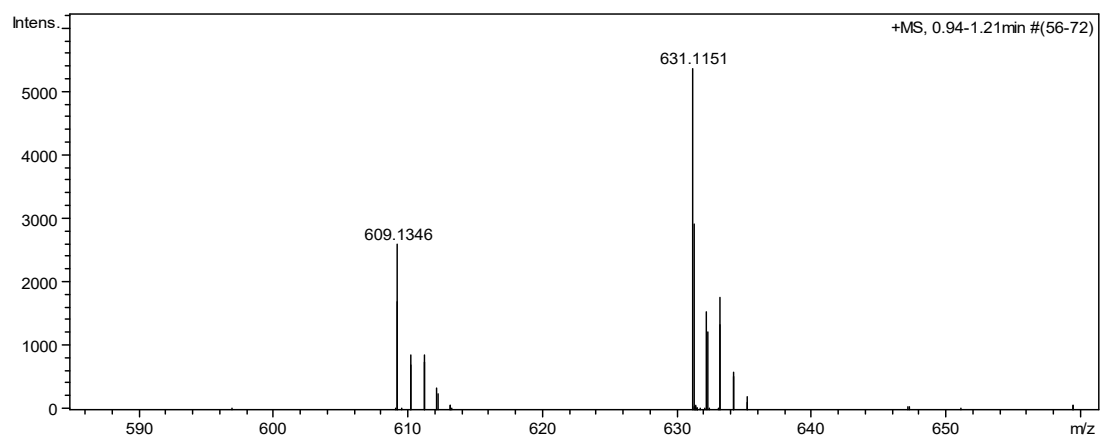
***cis*-1-benzyl-5-chloro-4'-(4-methoxyphenyl)-7'-methyl-1'-(naphthalen-1-yl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3v):**





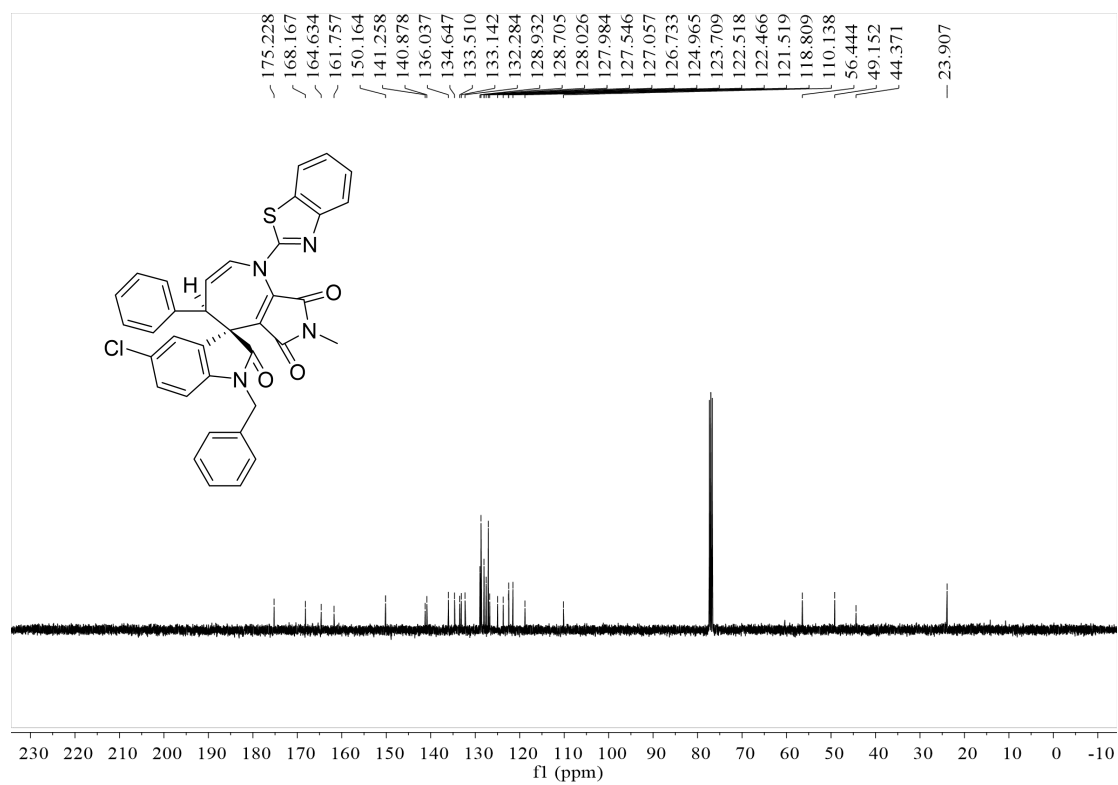
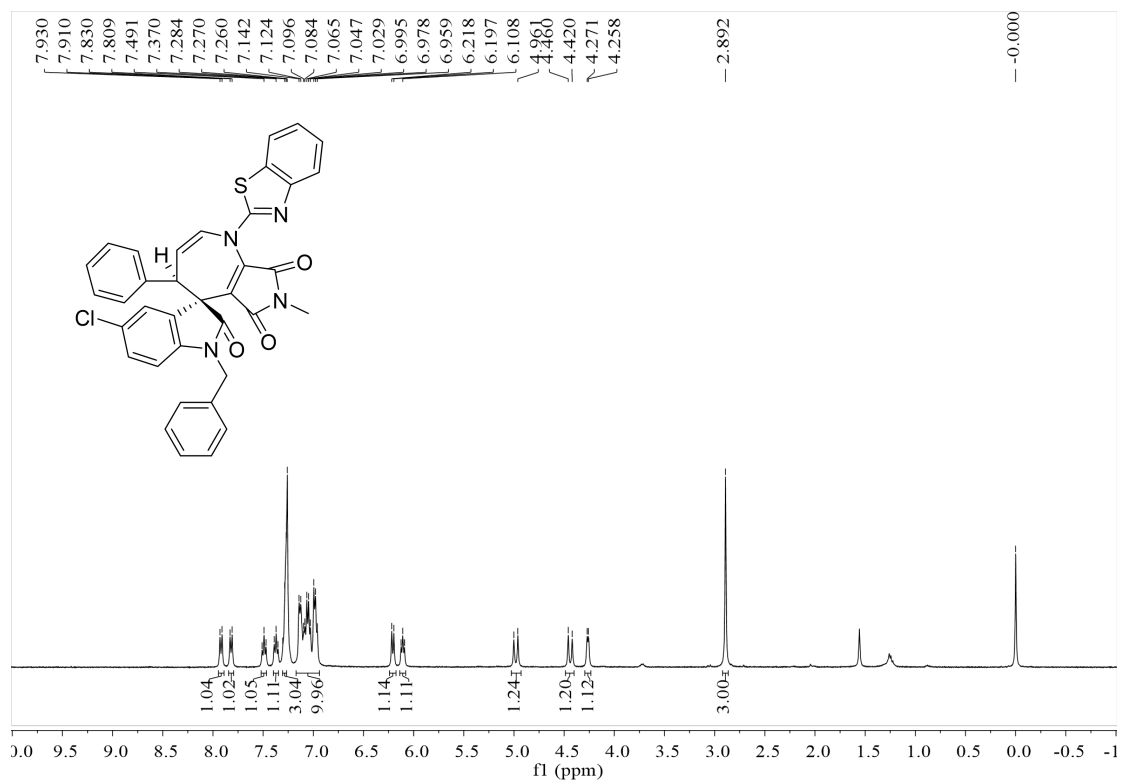
**cis-1-benzyl-5-chloro-4'-(2-methoxyphenyl)-7'-methyl-1'-(thiazol-2-yl)-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (3w):**

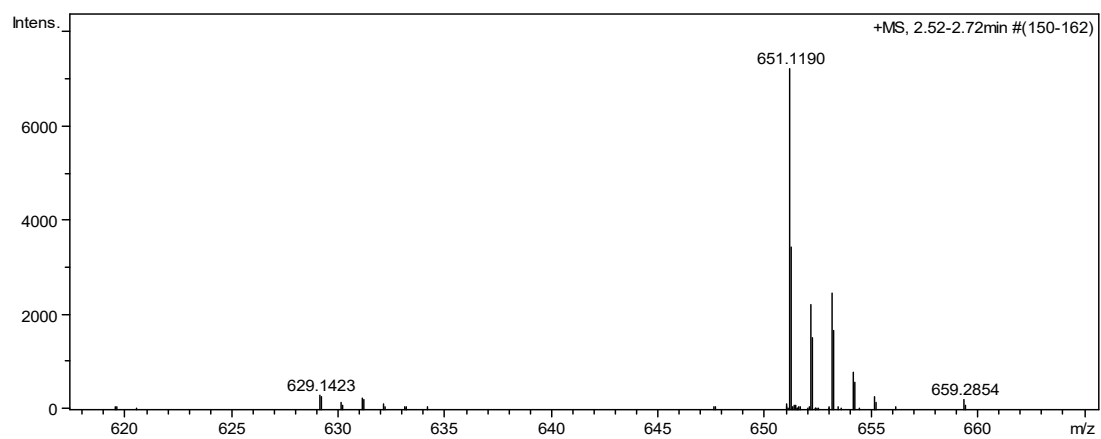




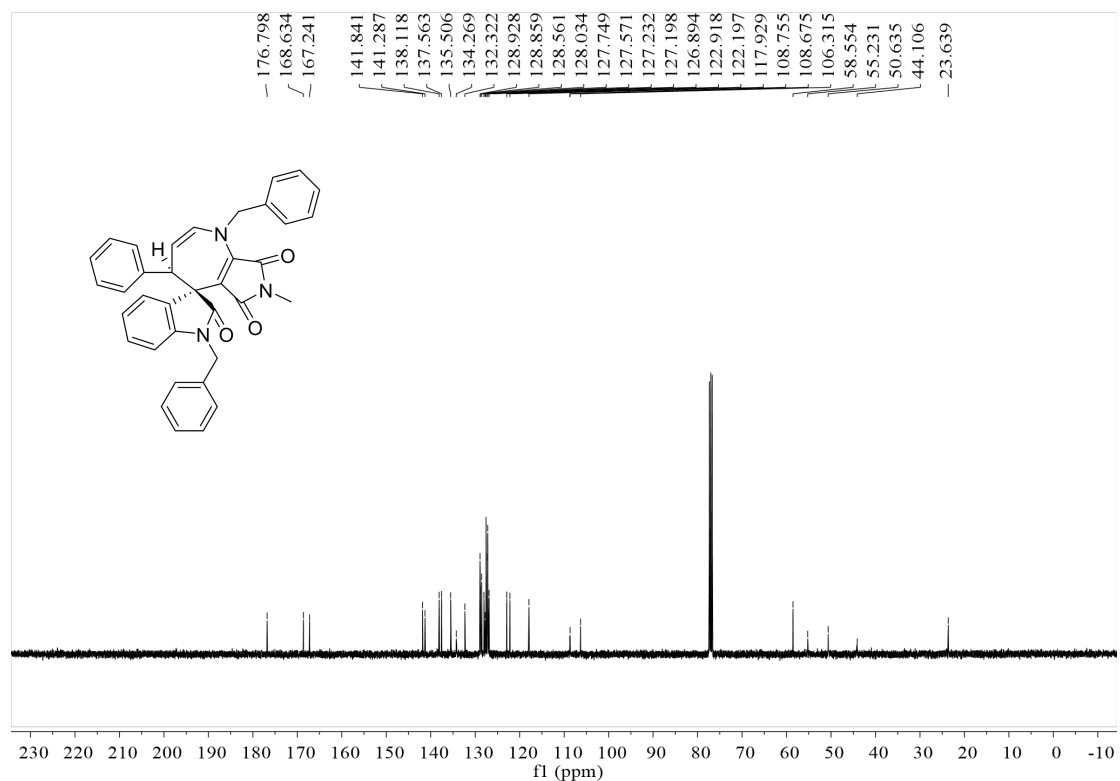
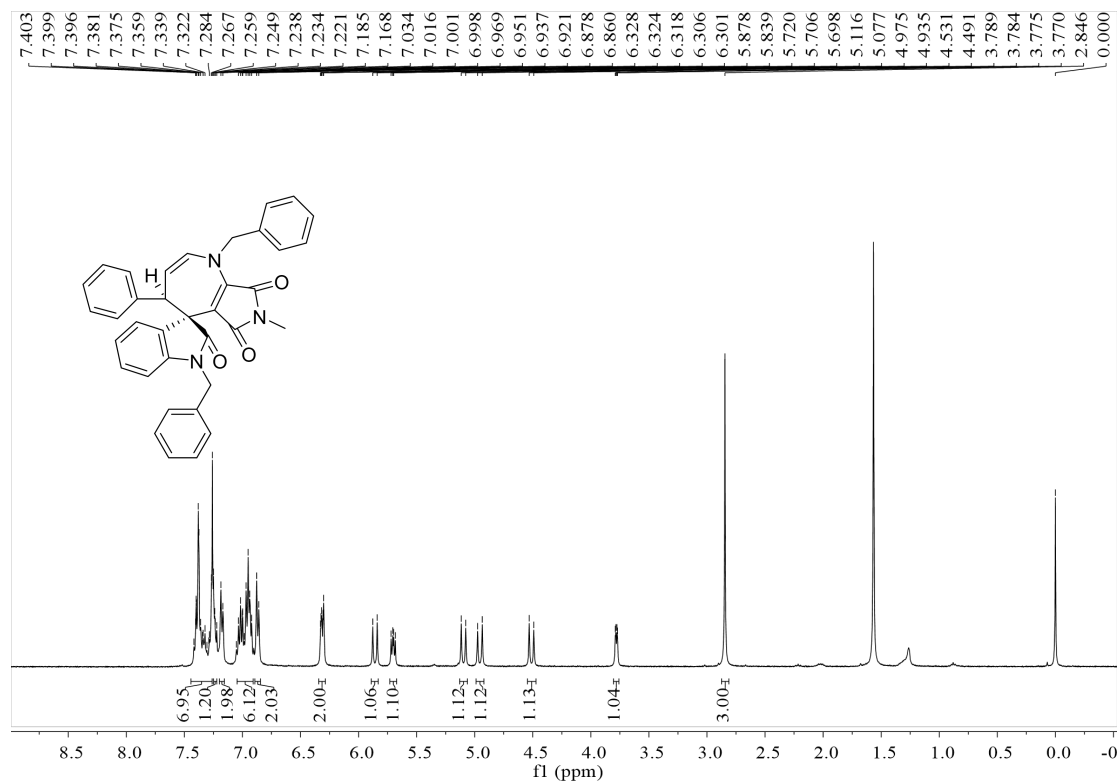


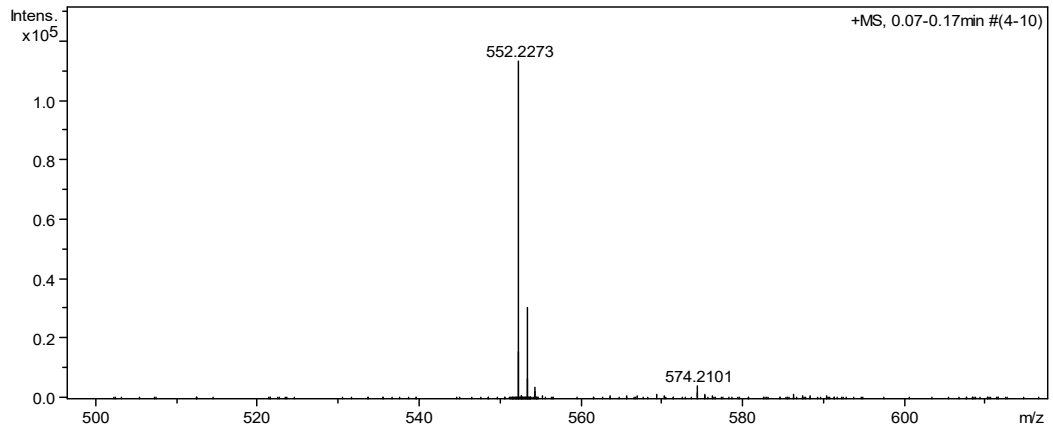
***cis*-1'-(benzo[*d*]thiazol-2-yl)-1-benzyl-5-chloro-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (3x):**



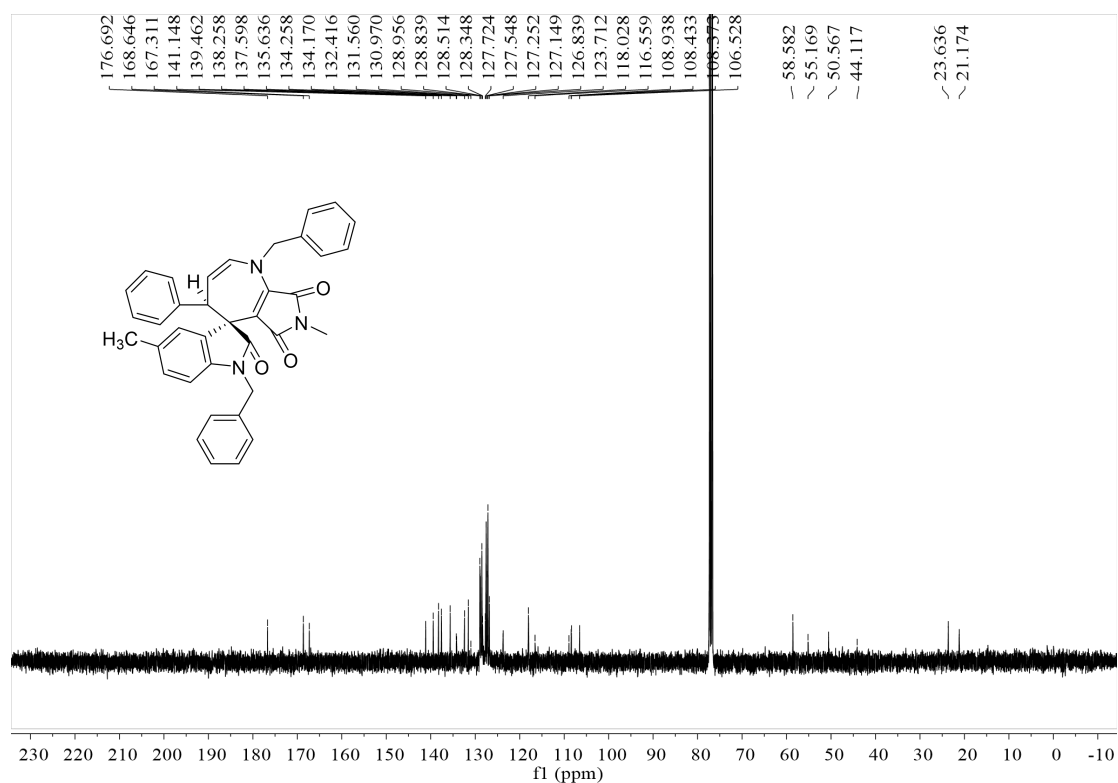
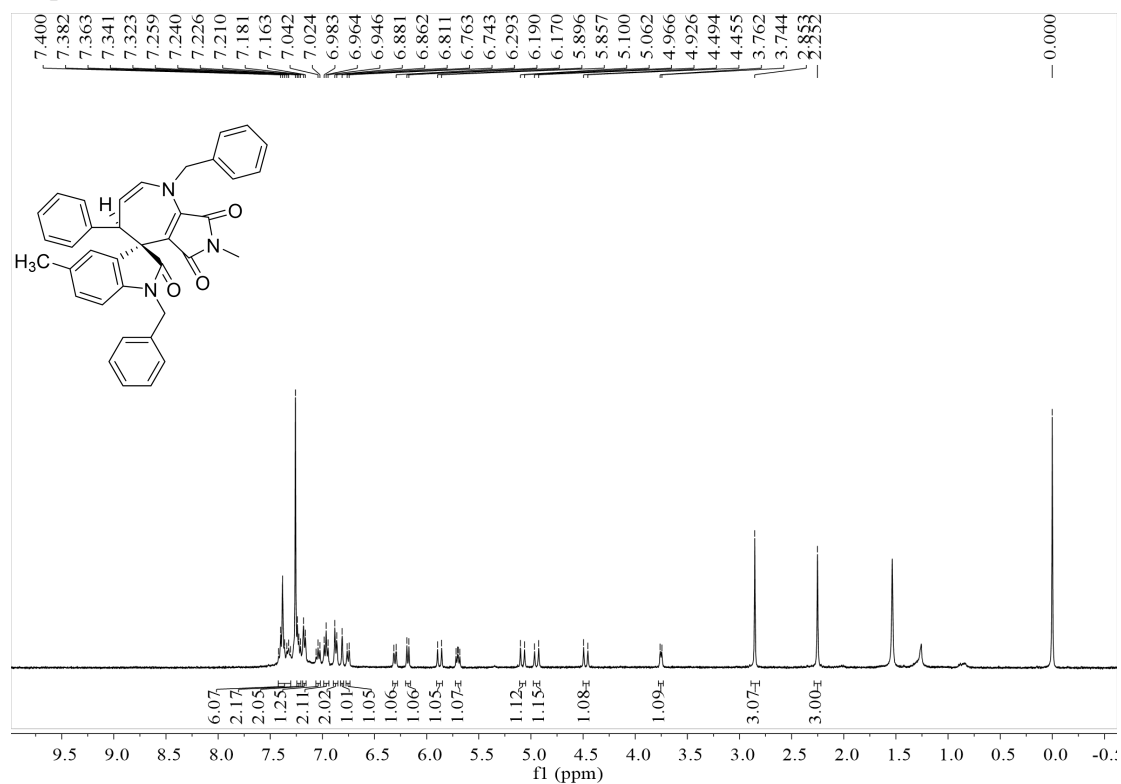


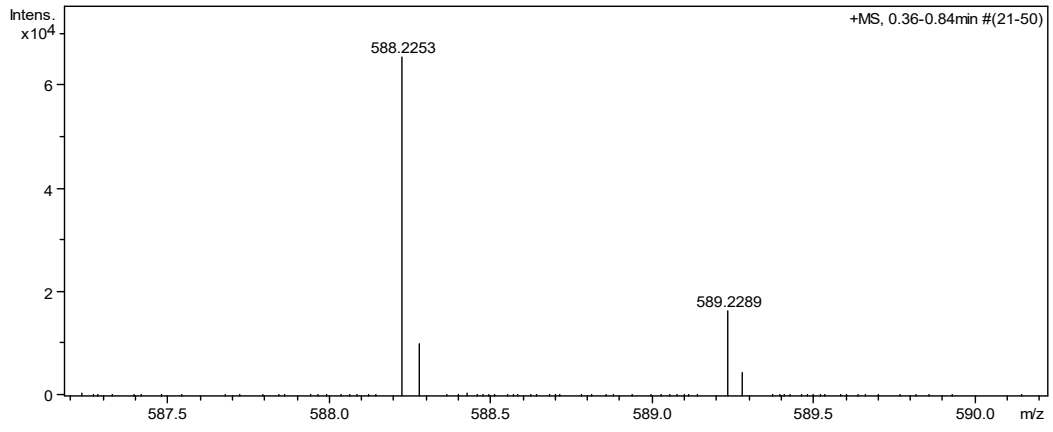
***cis*-1,1'-dibenzyl-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azine]-2,6',8'(7'*H*)-trione (4a):**



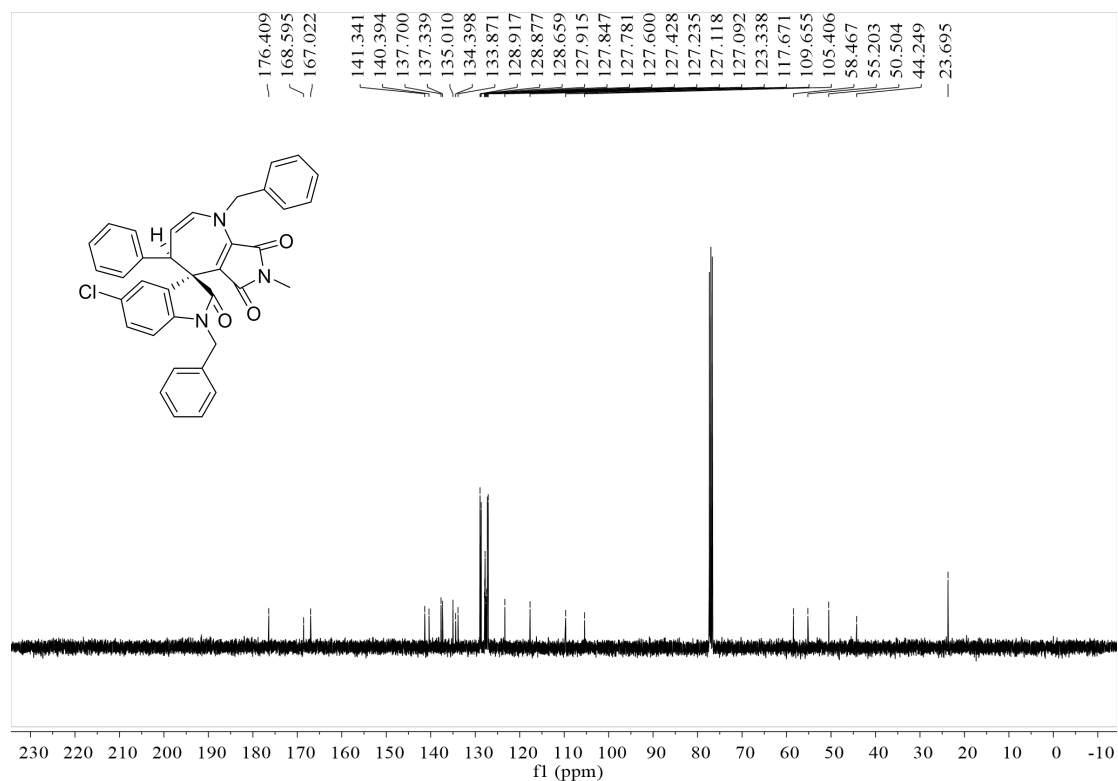
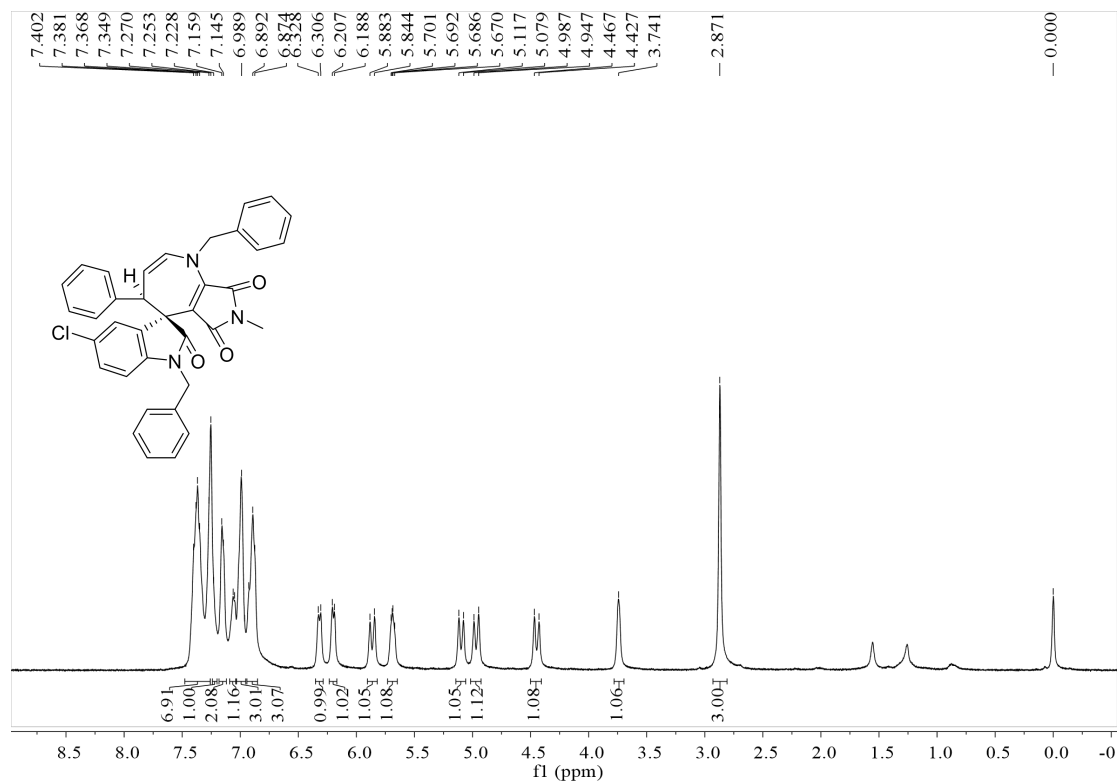


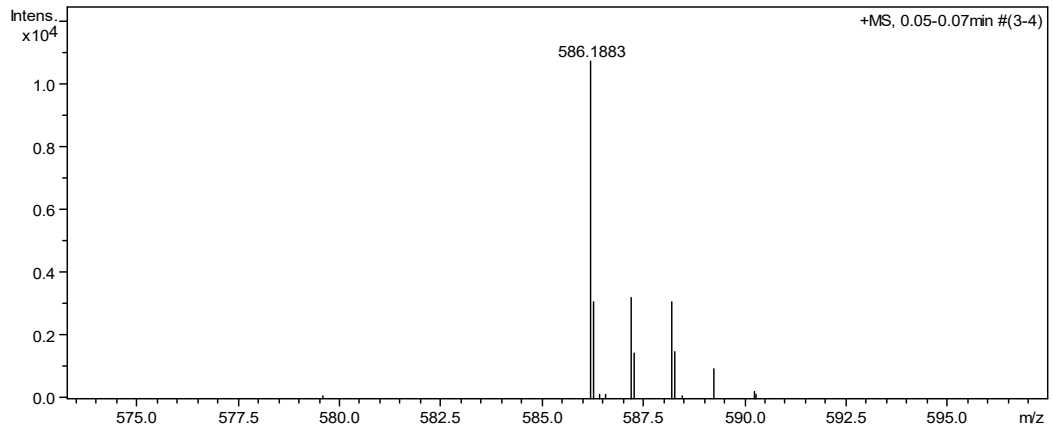
***cis*-1,1'-dibenzyl-5,7'-dimethyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6,8'(7'*H*)-trione (4b):**





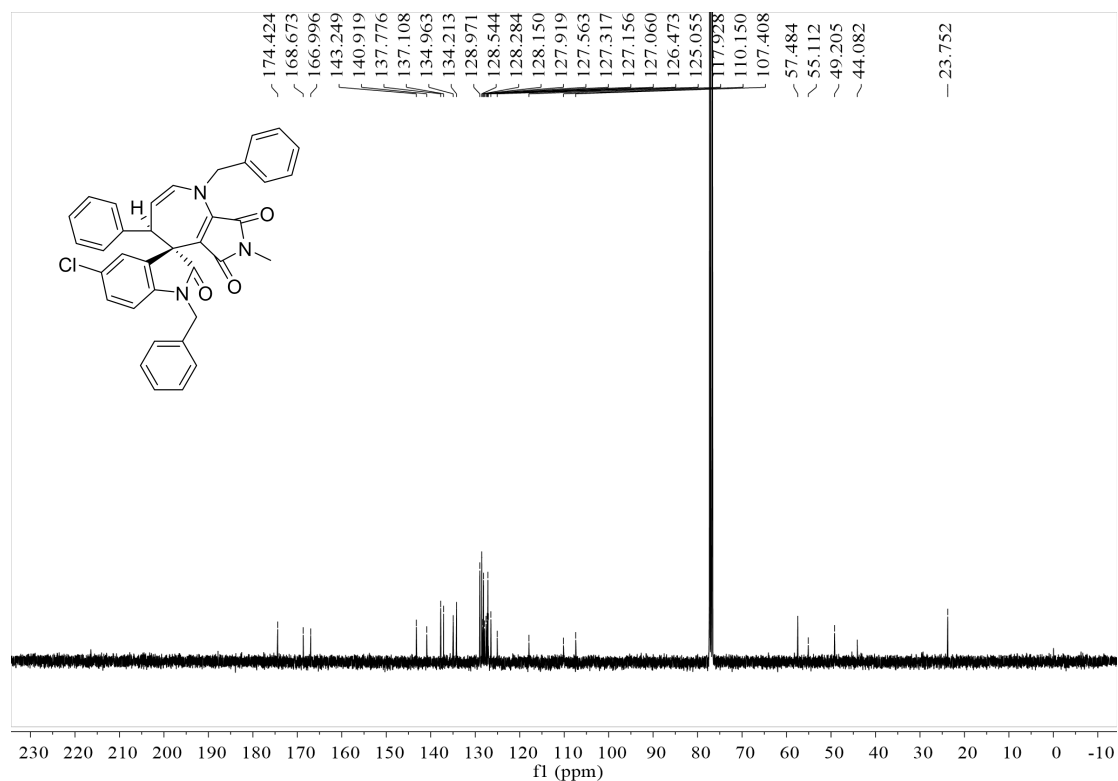
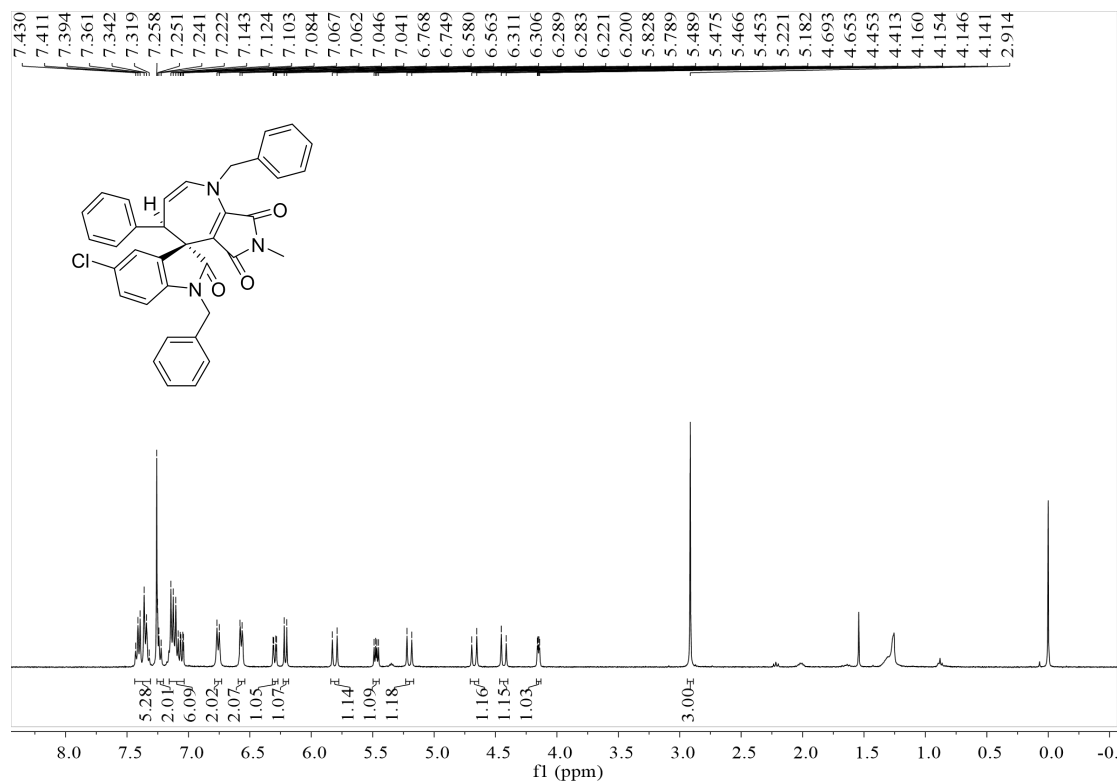
**cis-1,1'-dibenzyl-5-chloro-7'-methyl-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (4c):**

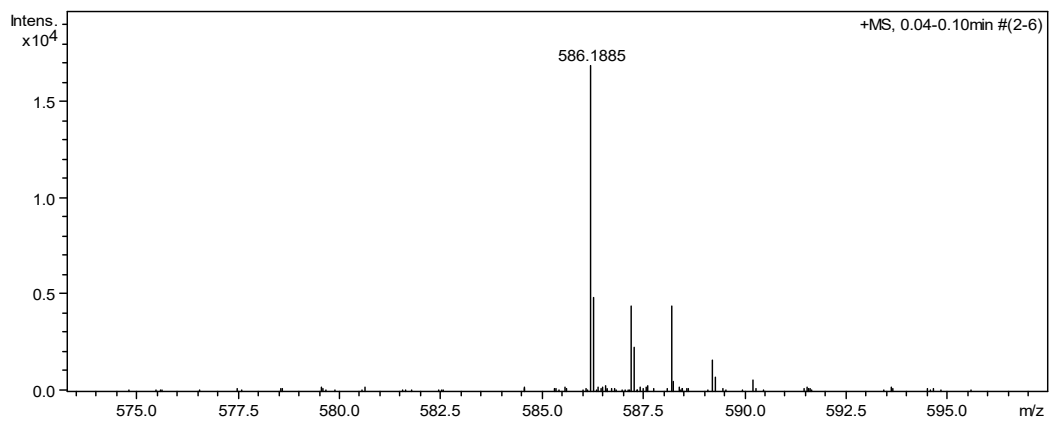




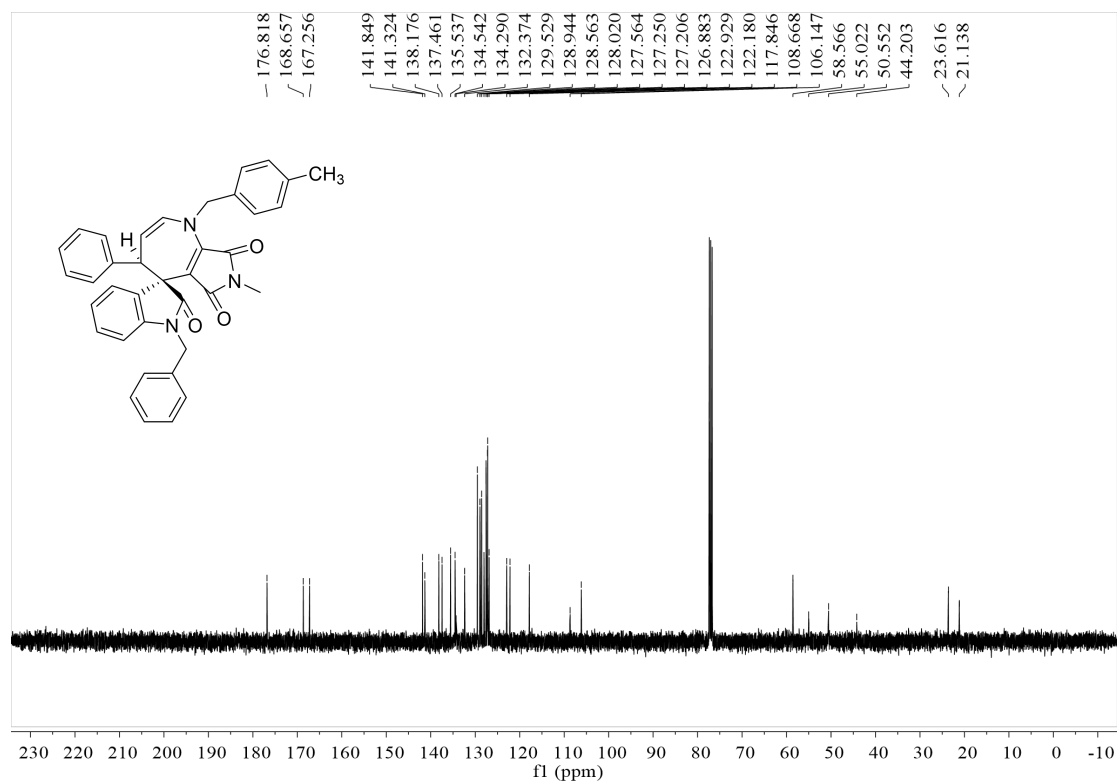
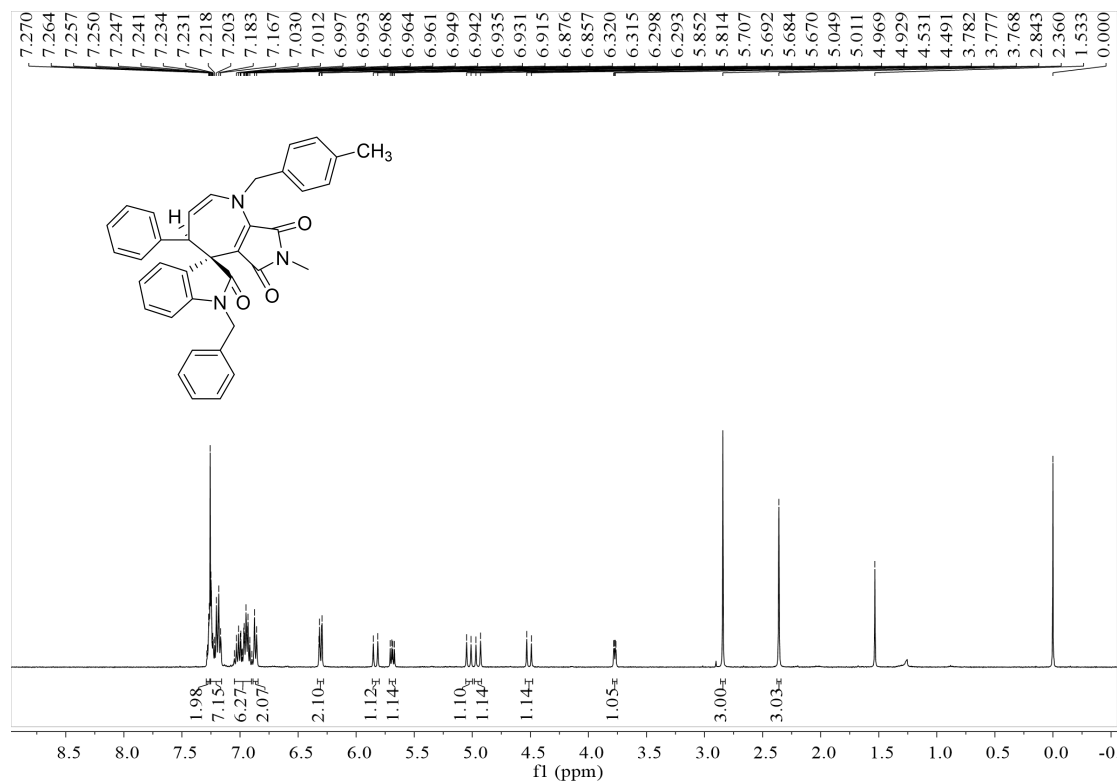


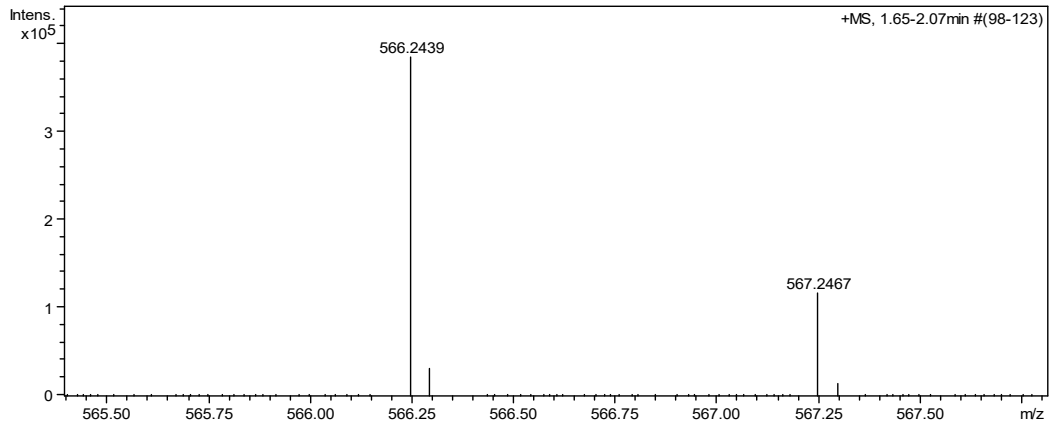
***trans*-1,1'-dibenzyl-5-chloro-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrol  
o[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4c')**



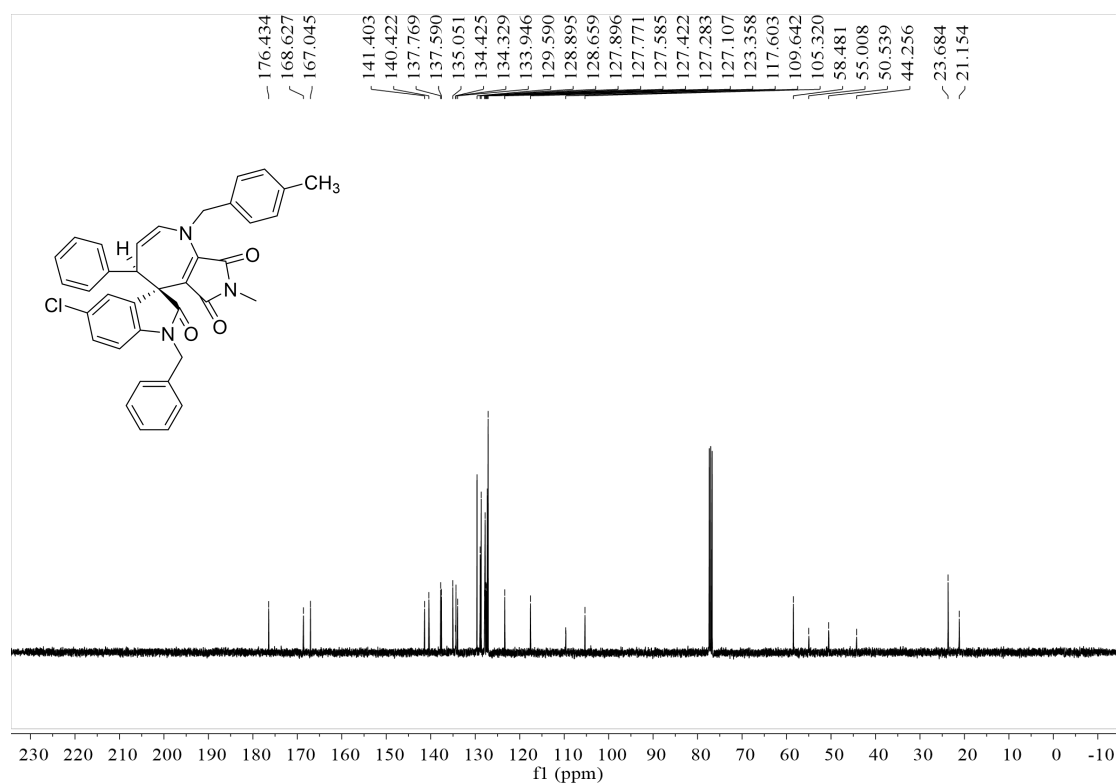
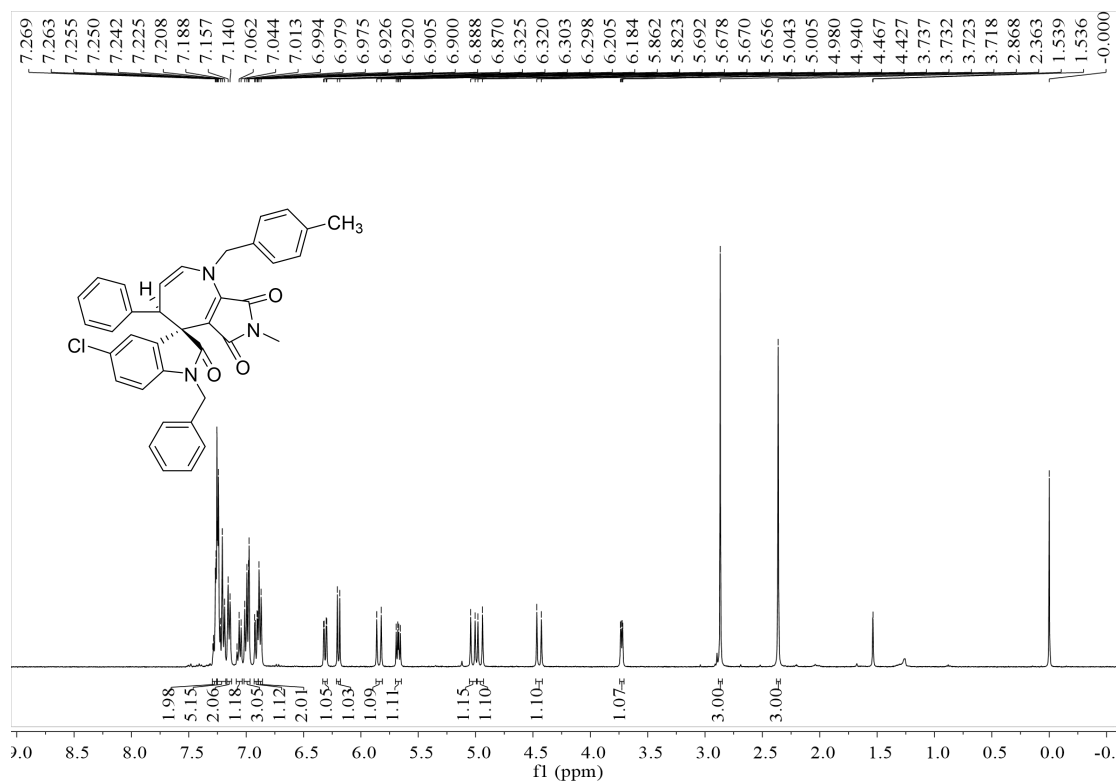


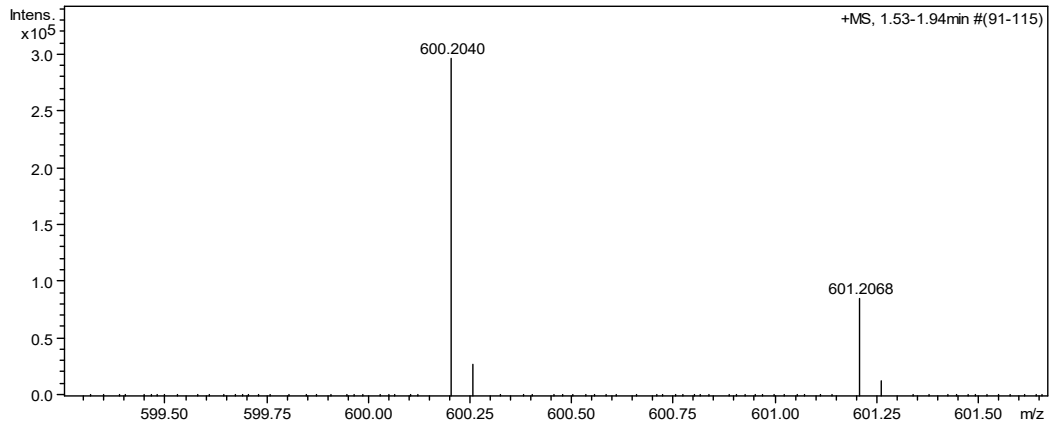
***cis*-1-benzyl-7'-methyl-1'-(4-methylbenzyl)-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-*p*yrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4d):**



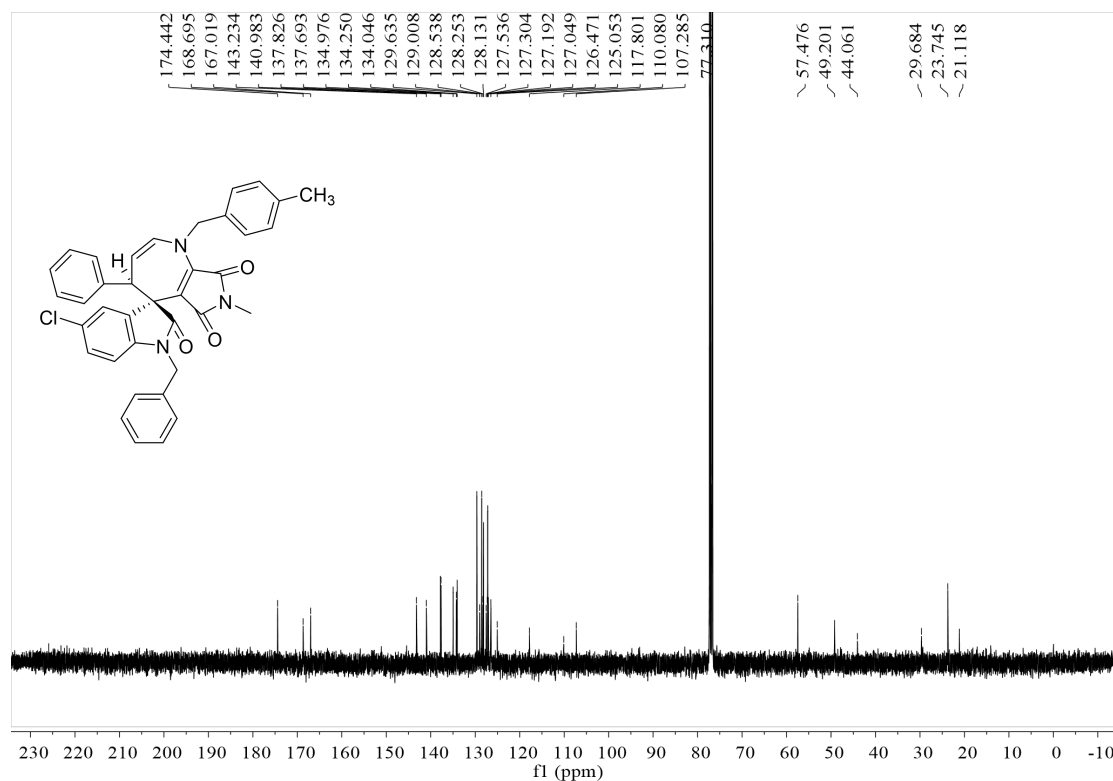
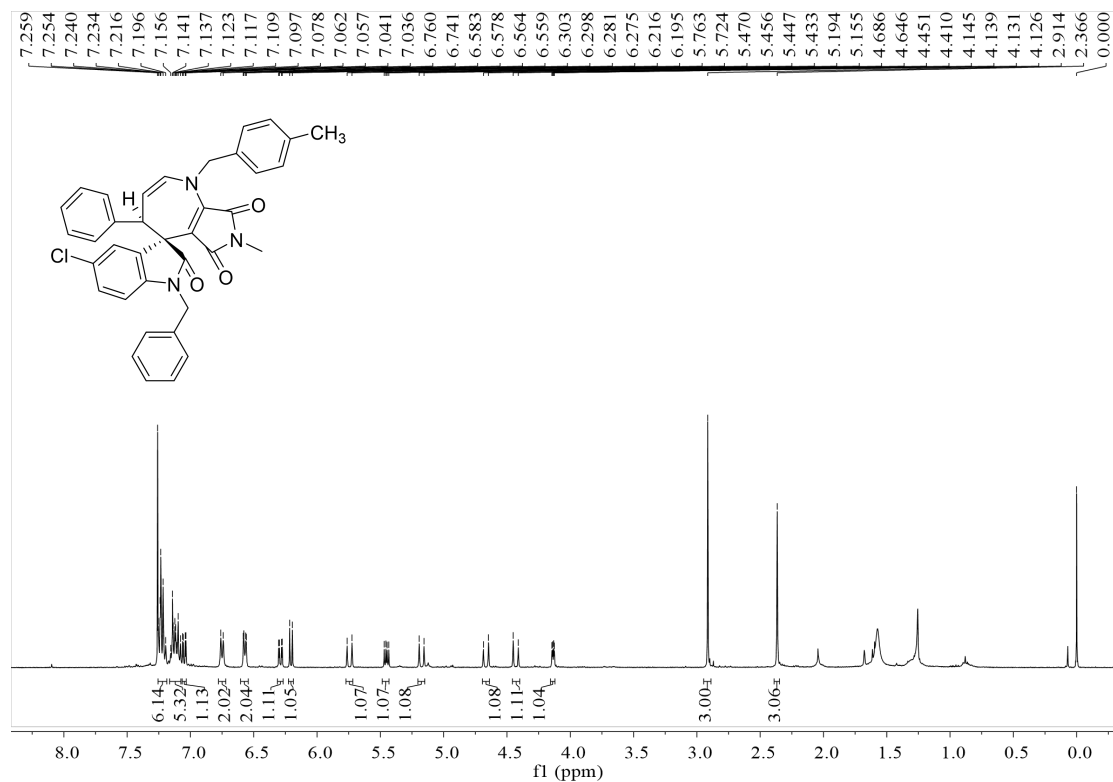


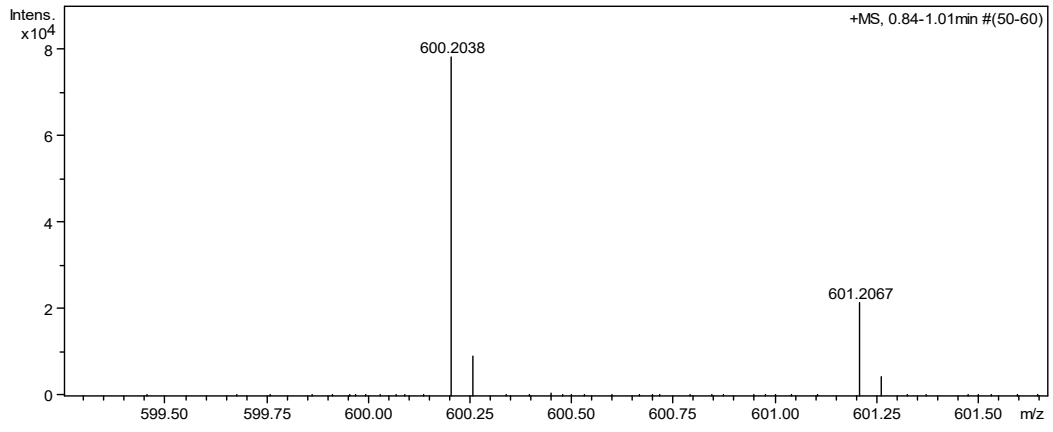
**cis-1-benzyl-5-chloro-7'-methyl-1'-(4-methylbenzyl)-4'-phenyl-1',4'-dihydro-6'H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (4e):**





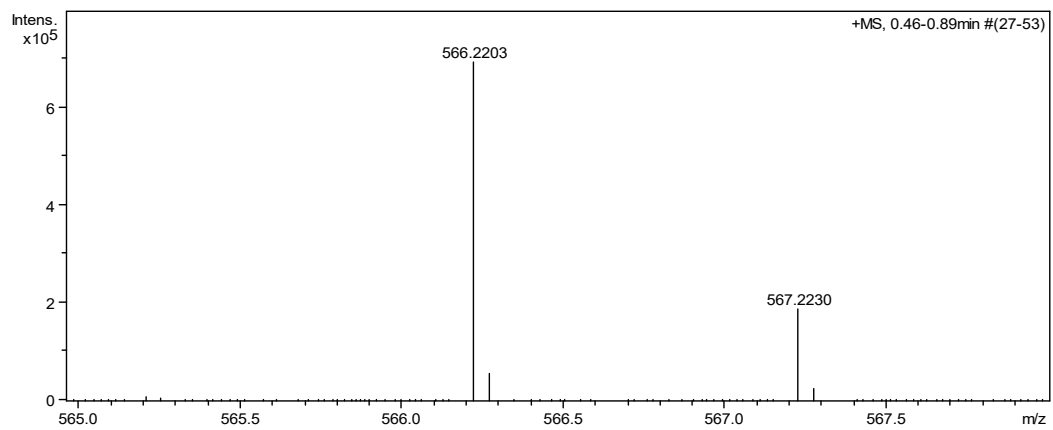
***trans*-1-benzyl-5-chloro-7'-methyl-1'-(4-methylbenzyl)-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6,8'(7'*H*)-trione (4e')**



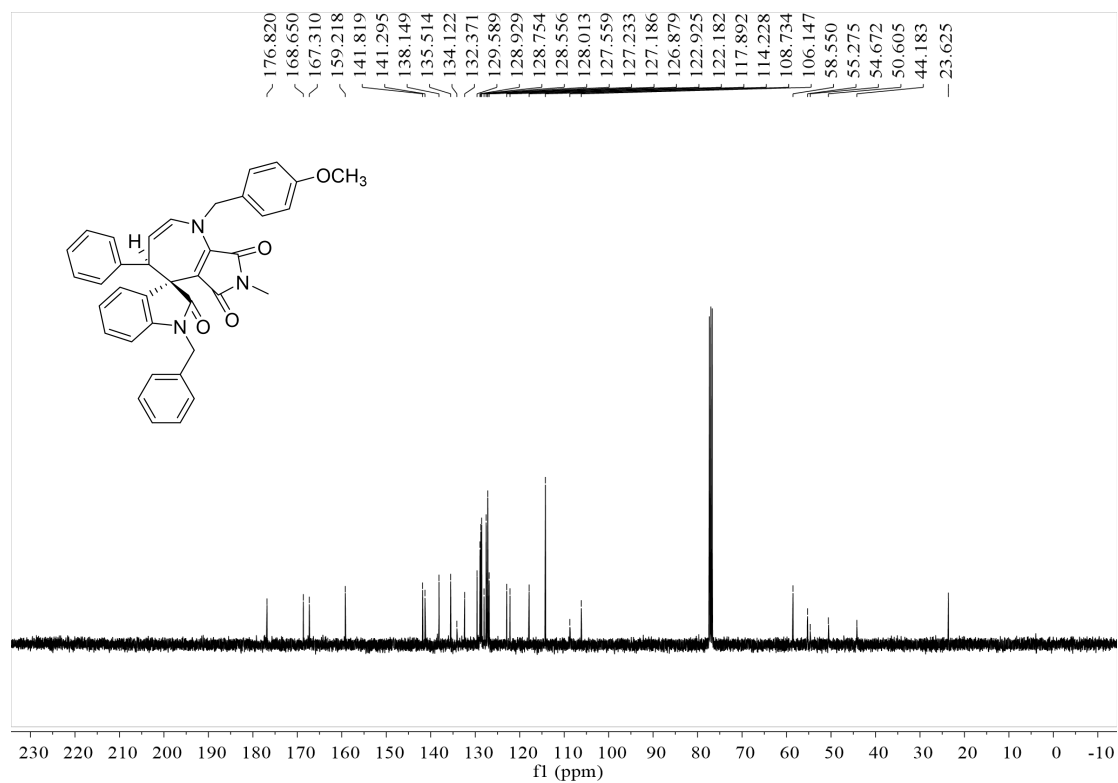
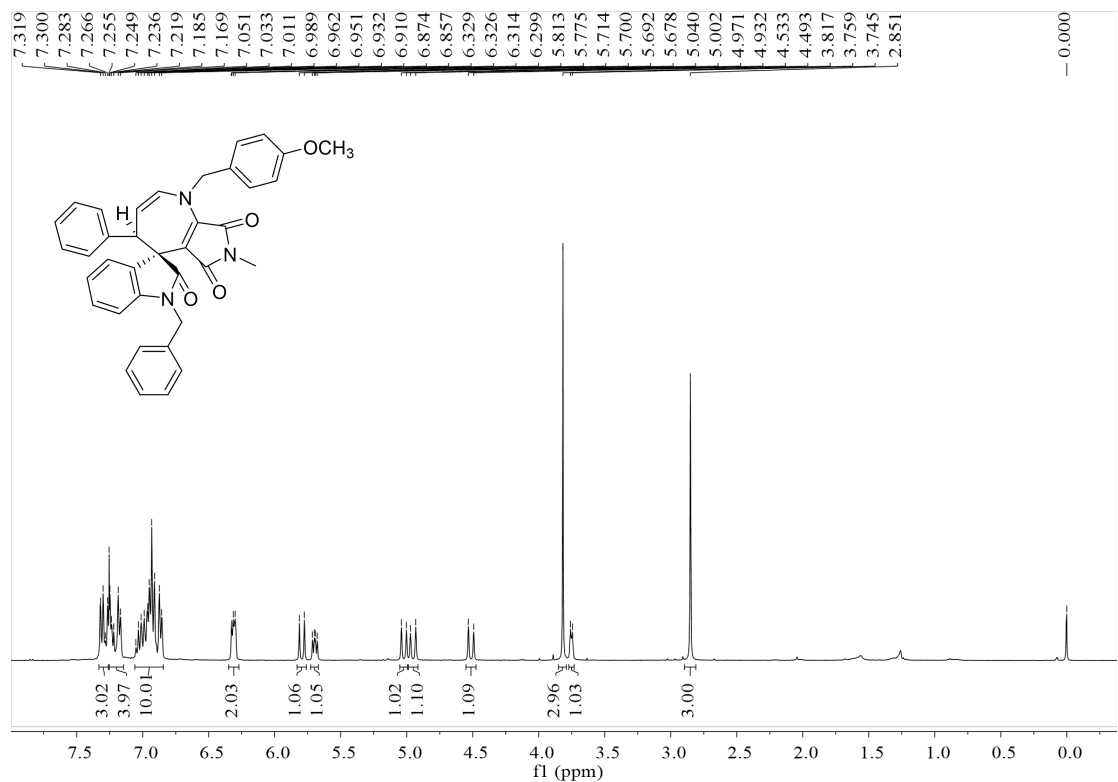


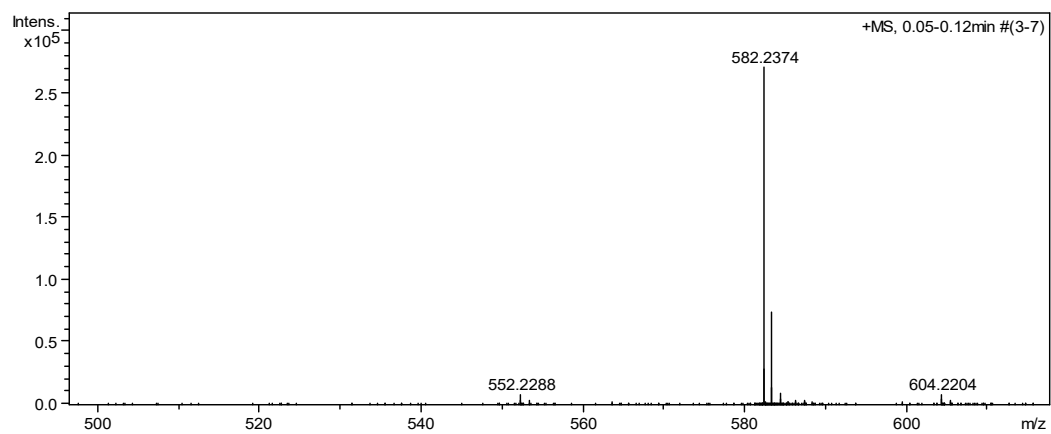




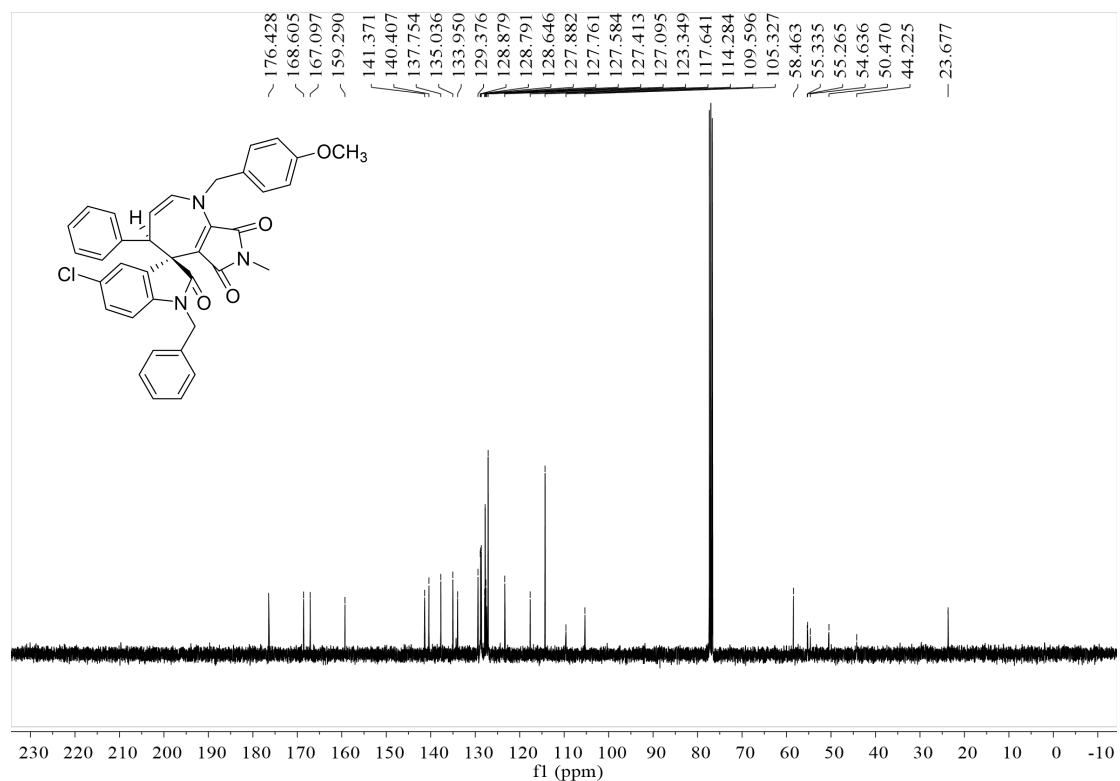
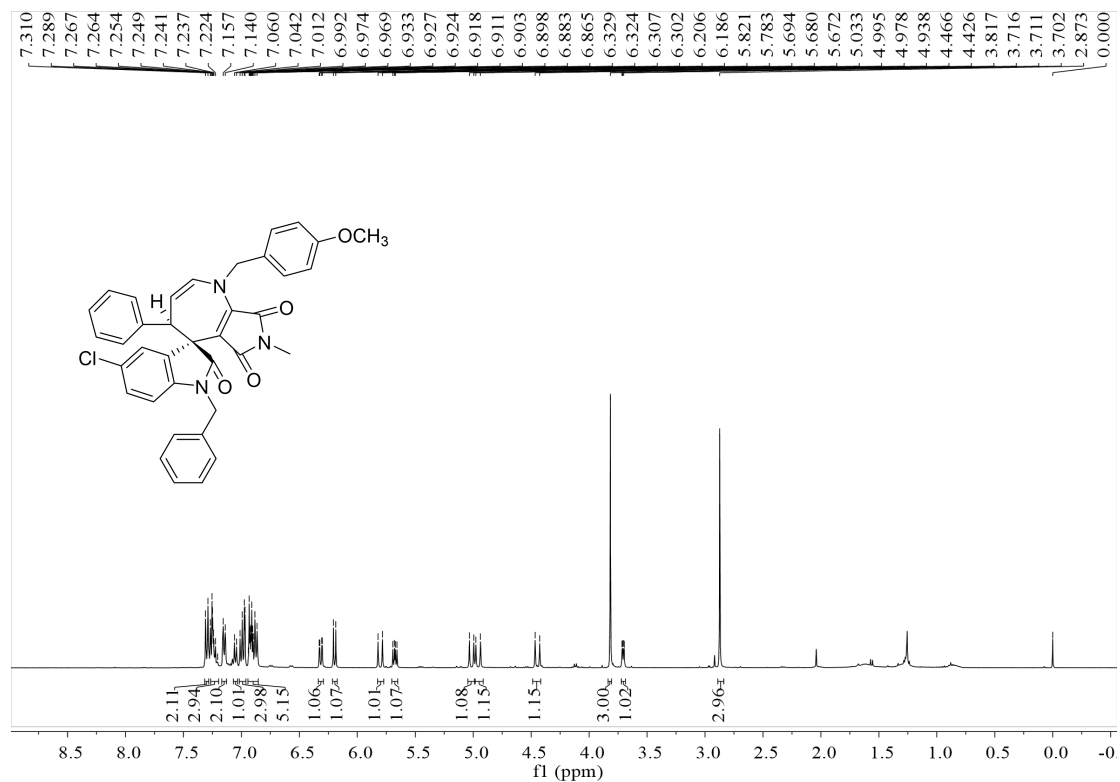


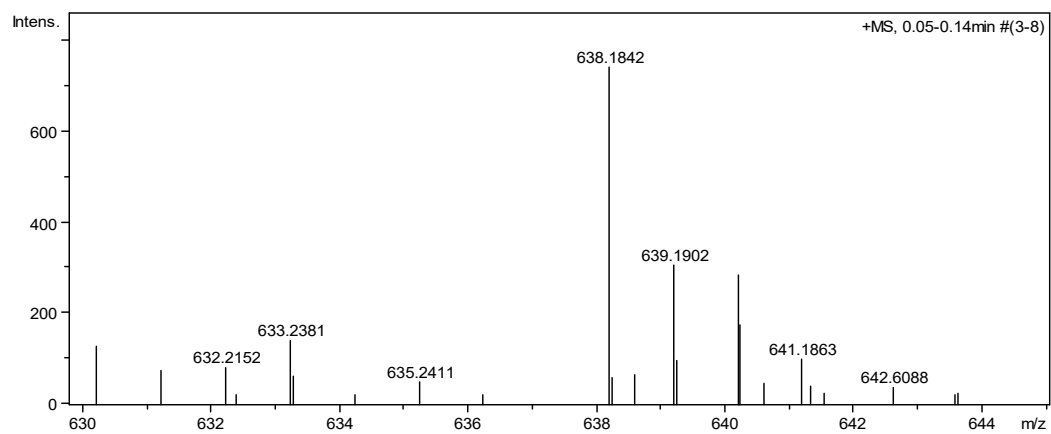
***cis*-1-benzyl-1'-(4-methoxybenzyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4g):**



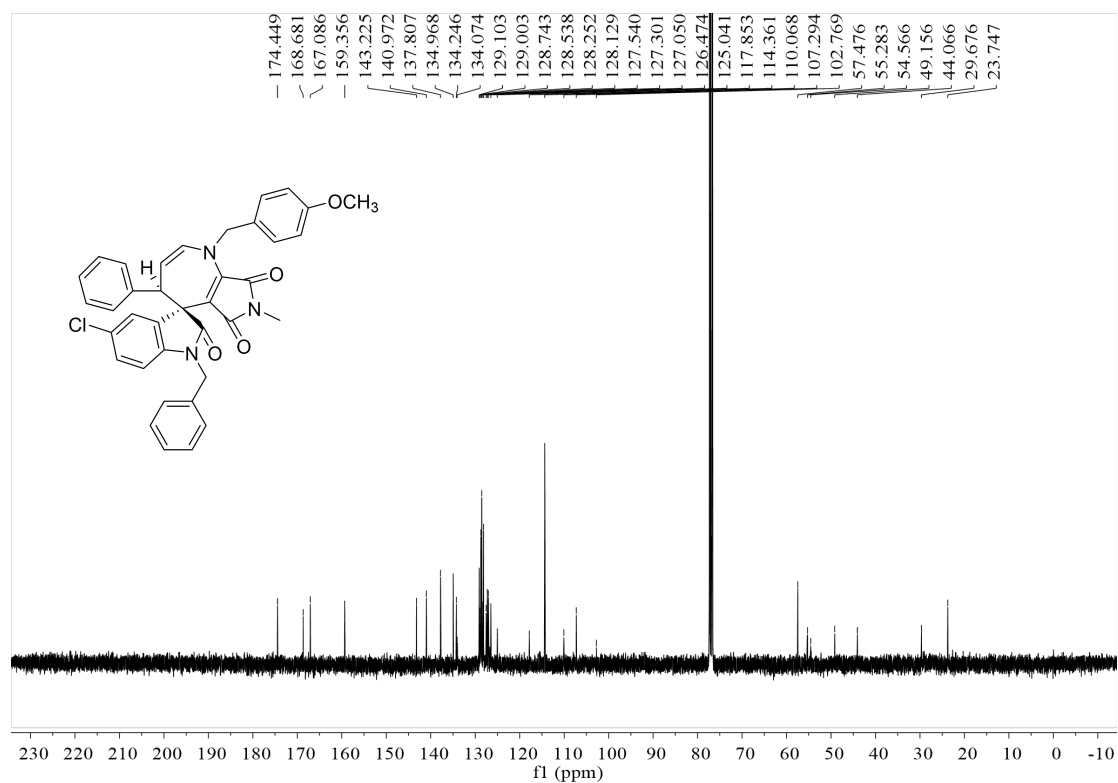
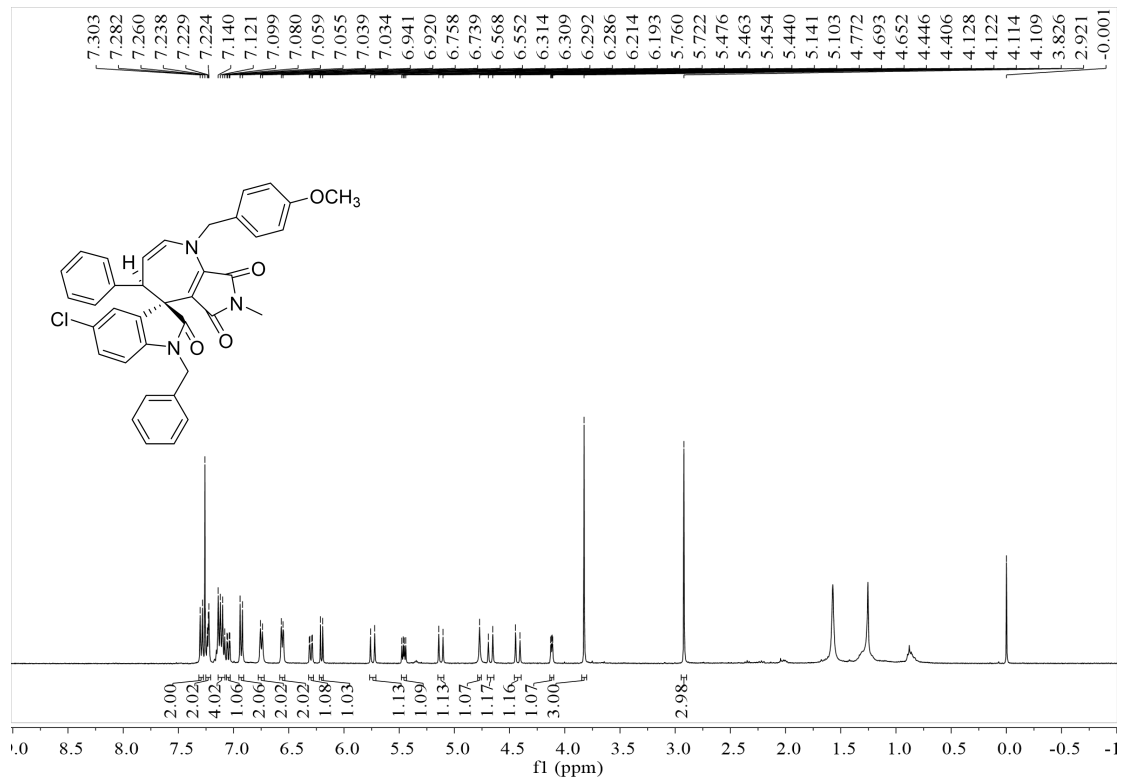


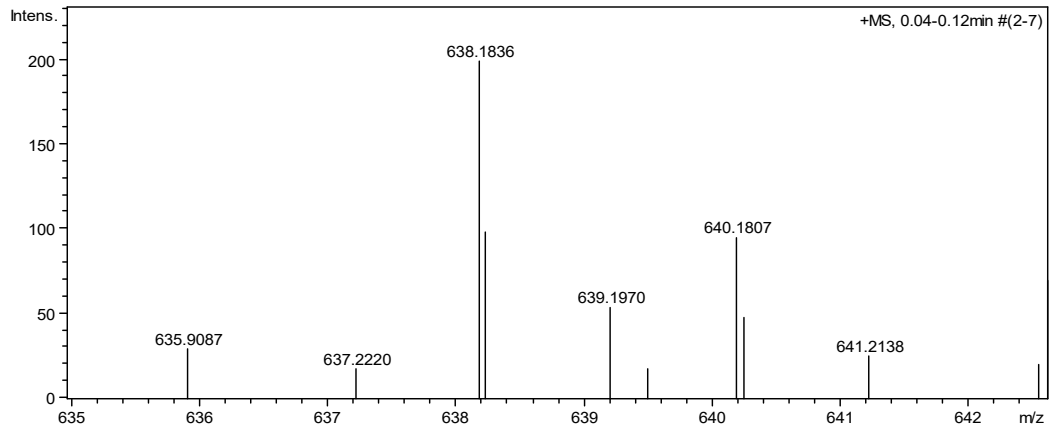
***cis*-1-benzyl-5-chloro-1'-(4-methoxybenzyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4h):**





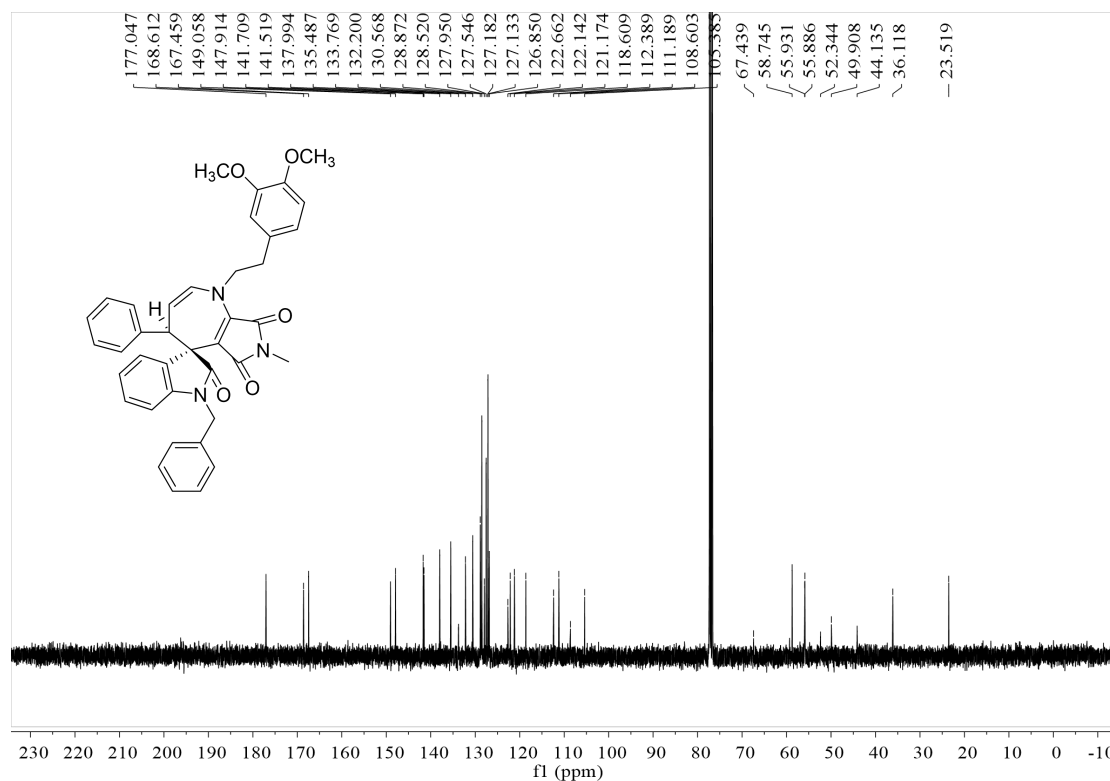
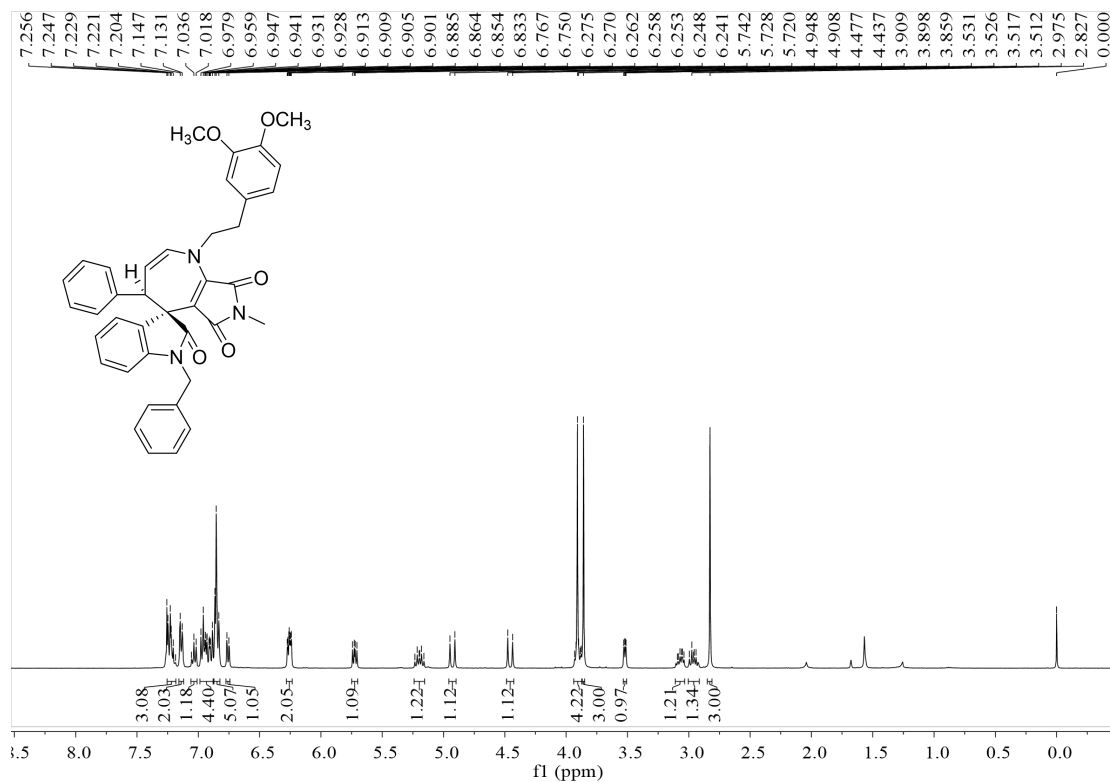
***trans*-1-benzyl-5-chloro-1'-(4-methoxybenzyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[*indoline*-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4h')**

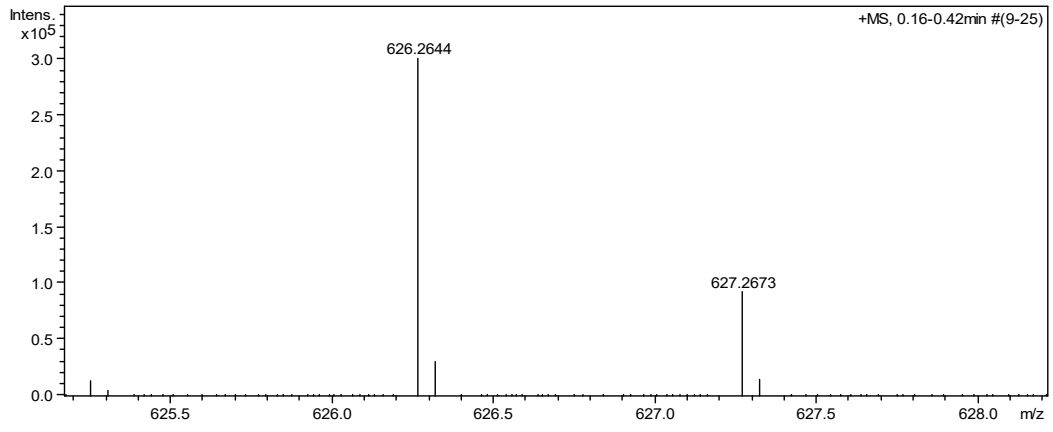




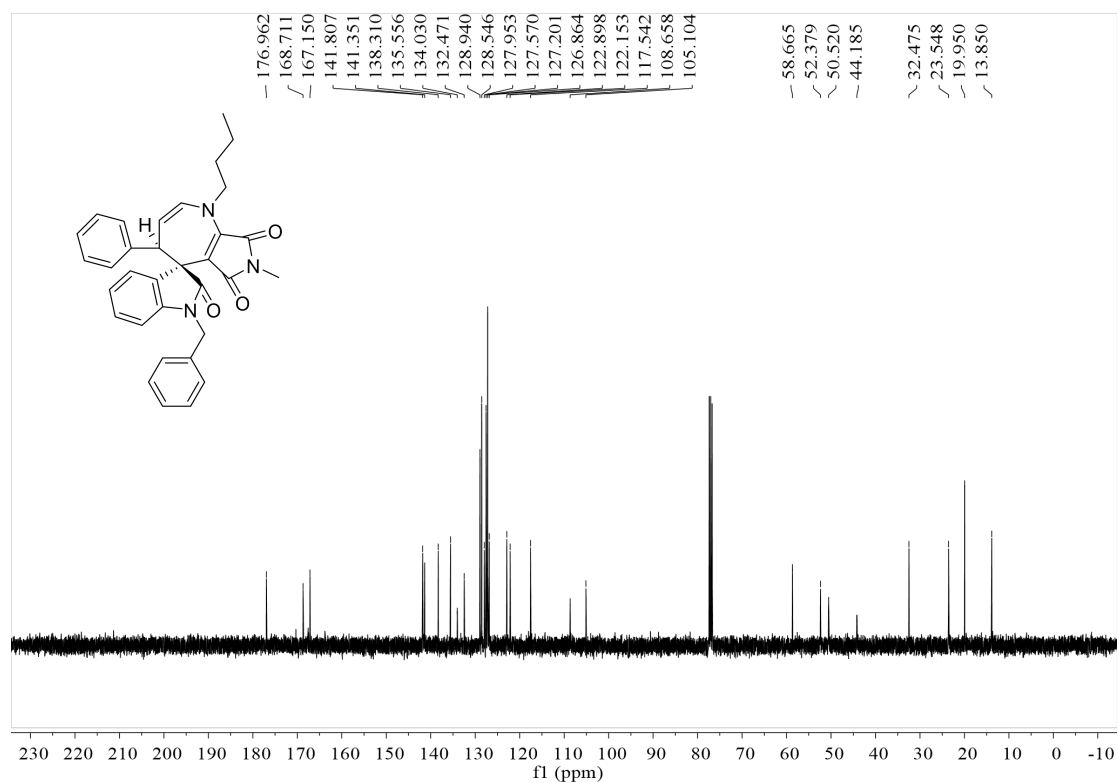
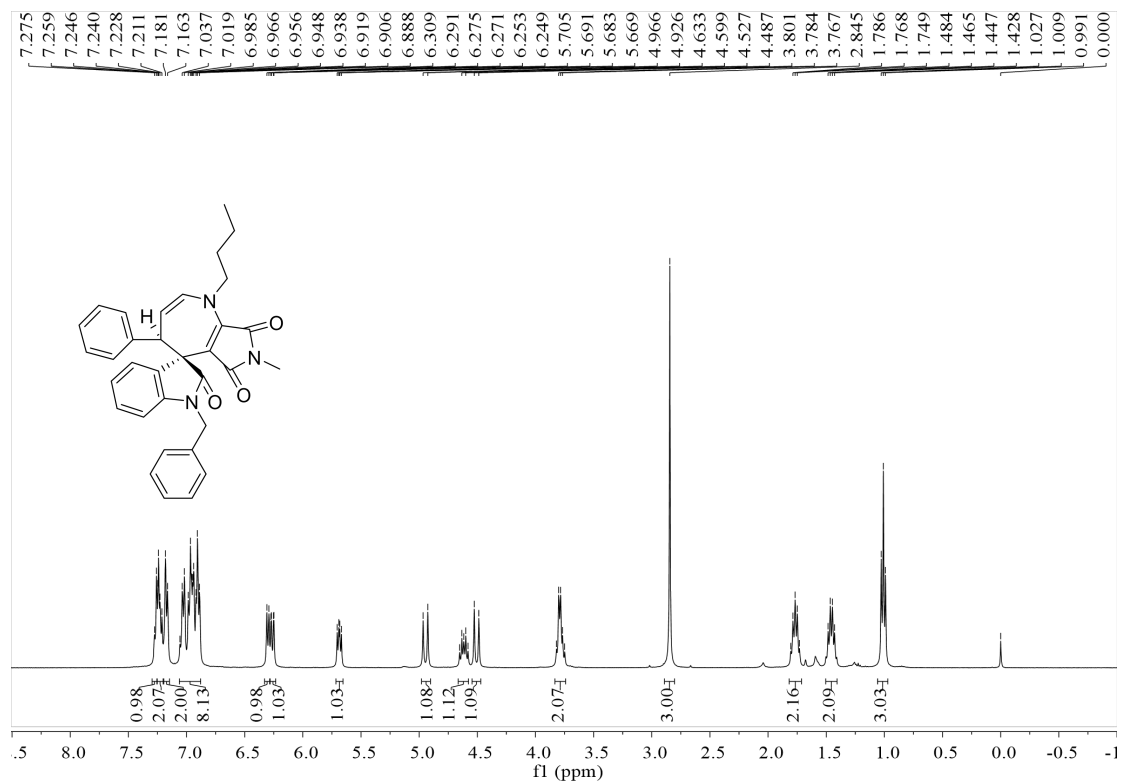


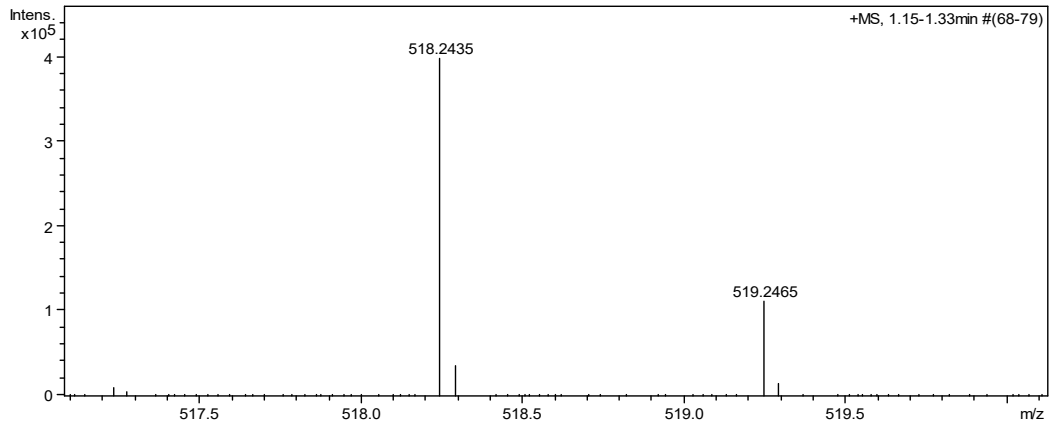
***cis*-1-benzyl-1'-(3,4-dimethoxyphenethyl)-7'-methyl-4'-phenyl-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6,8'(7'*H*)-trione (4i):**



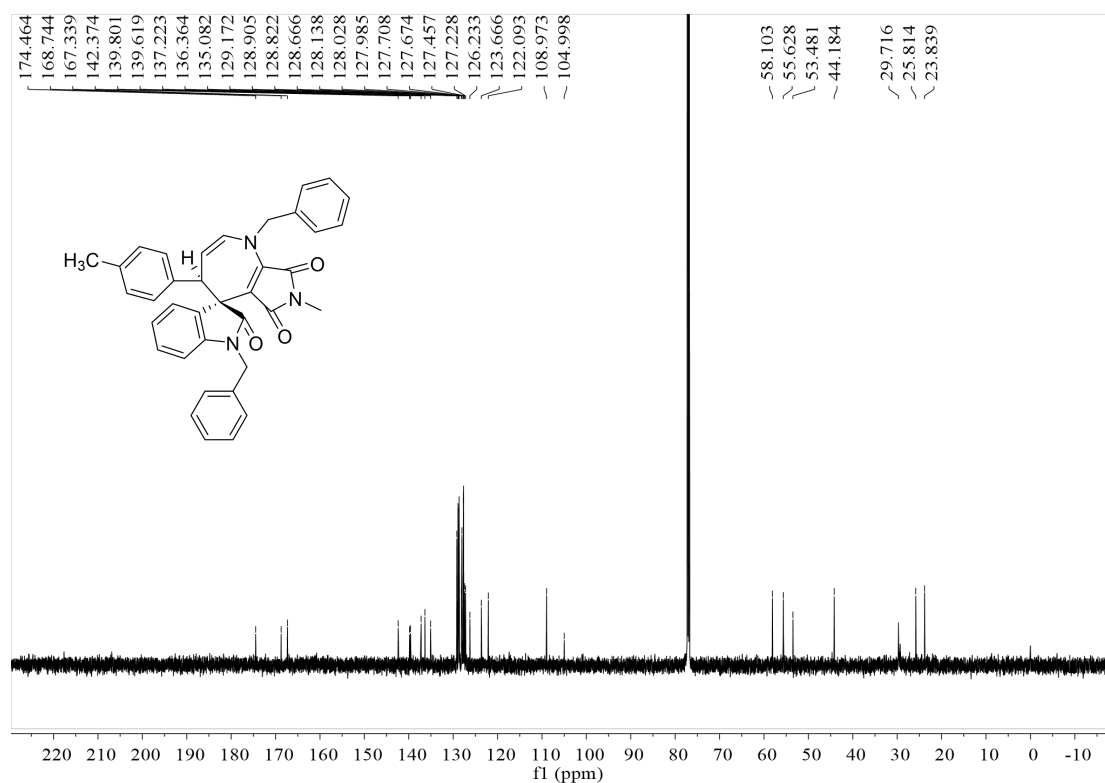
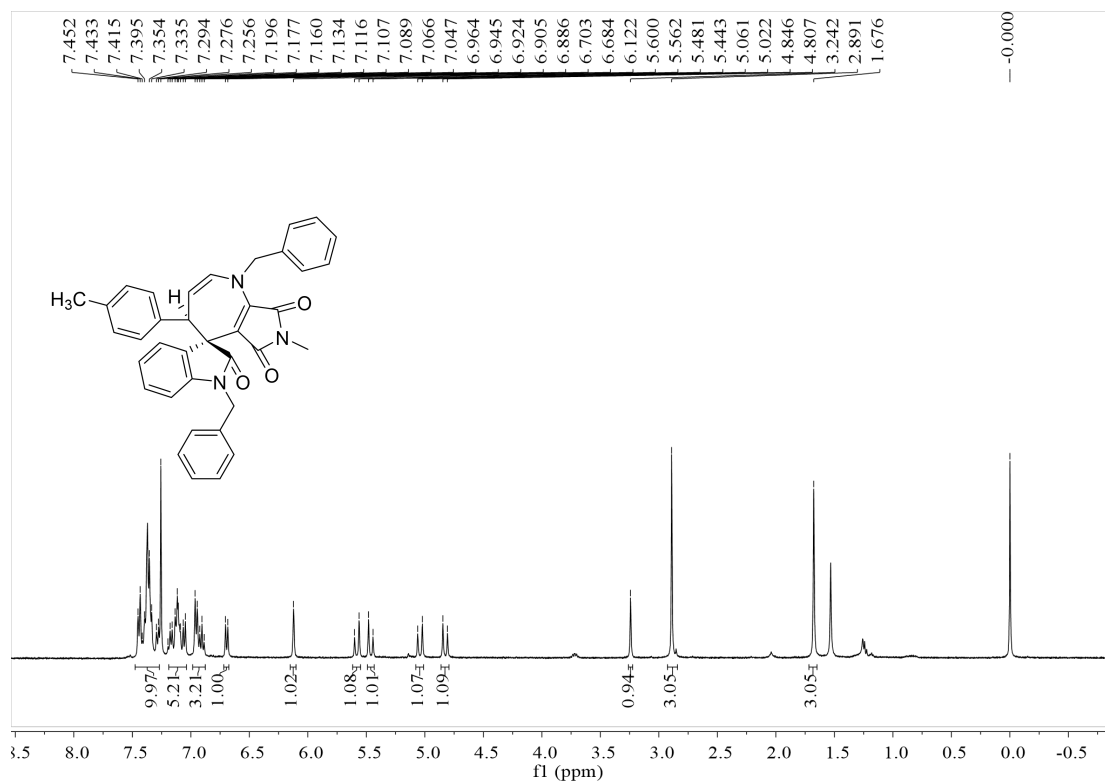


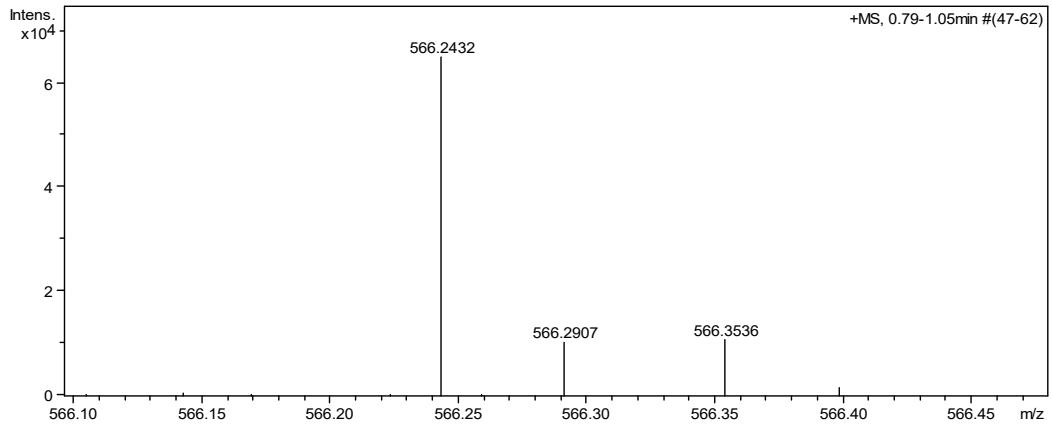
**cis-1-benzyl-1'-butyl-7'-methyl-4'-phenyl-1',4'-dihydro-6'-H-spiro[indoline-3,5'-pyrrolo[3,4-b]azepine]-2,6',8'(7'H)-trione (4j):**



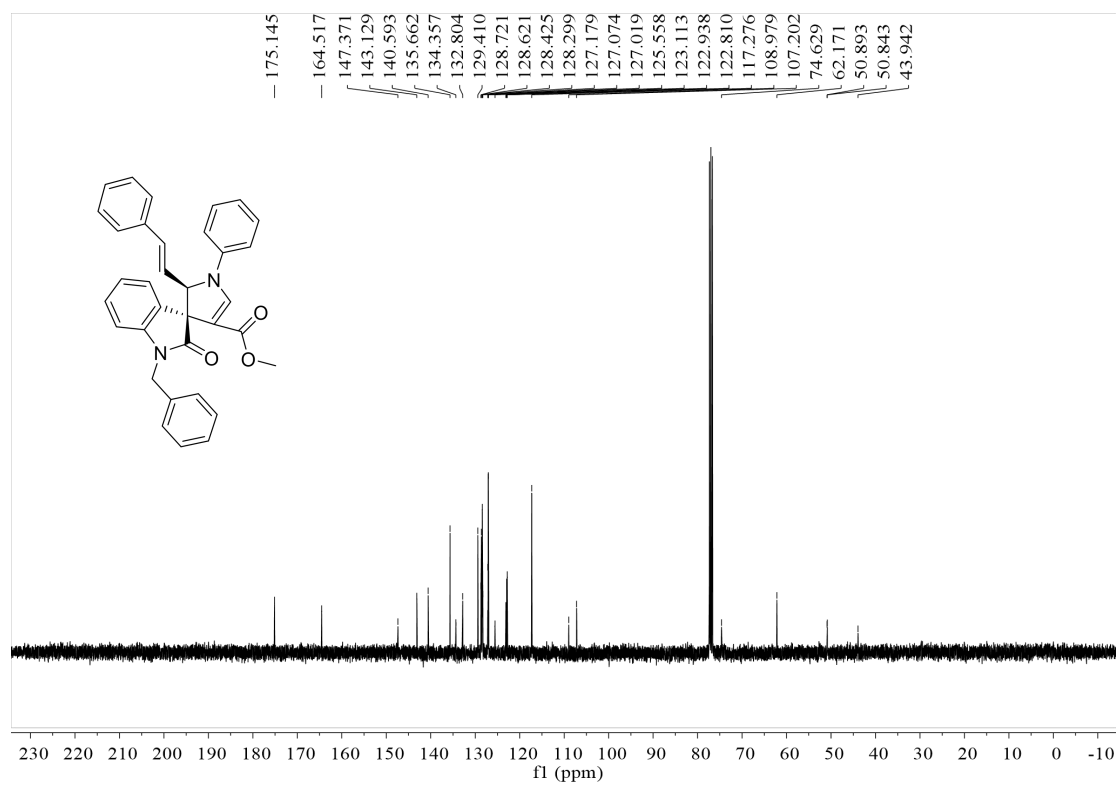
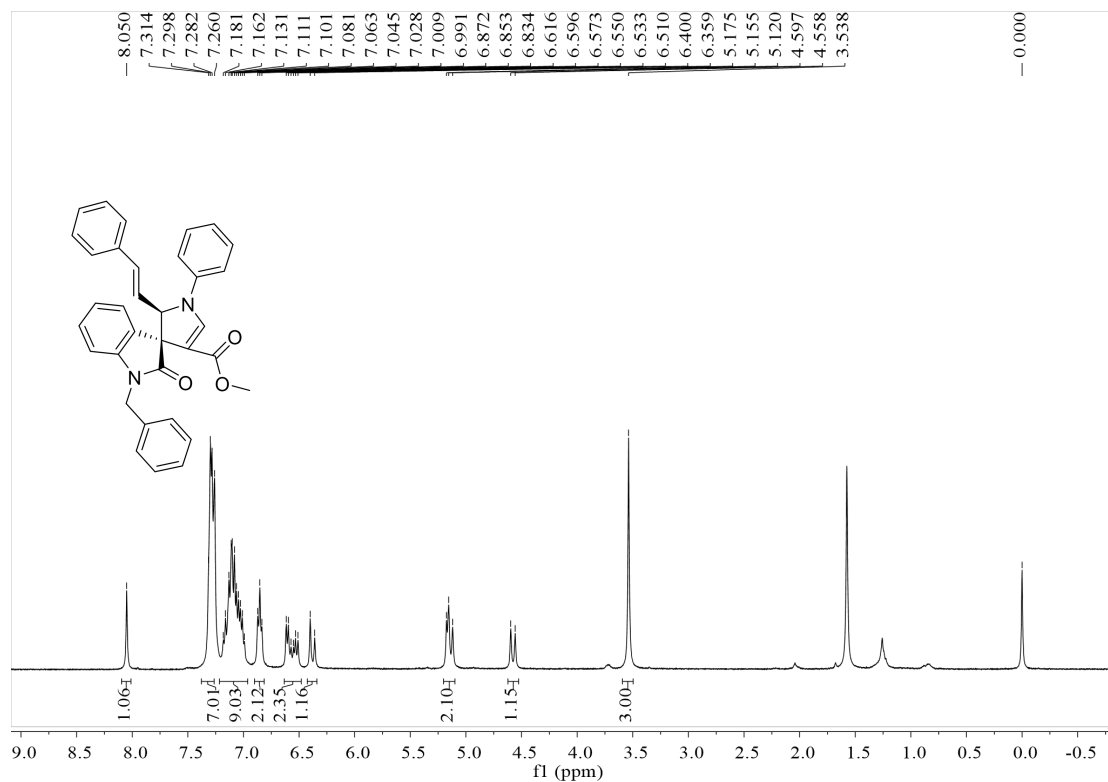


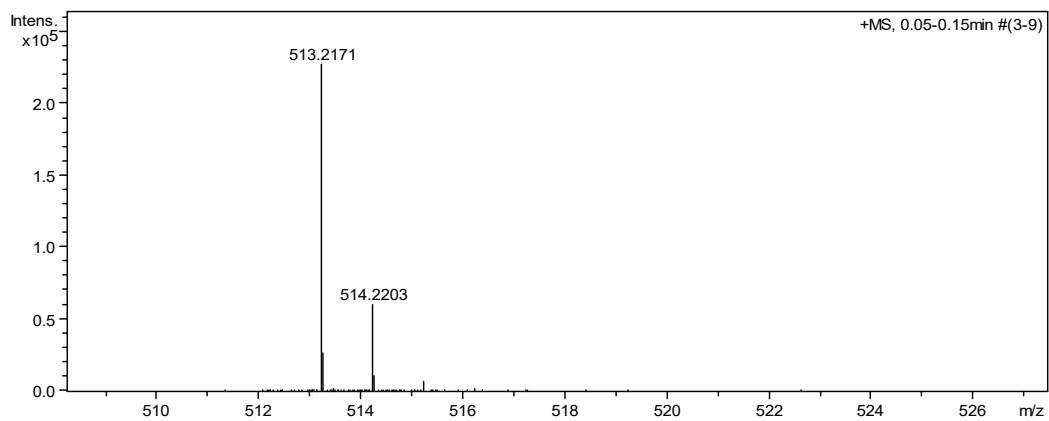
***cis*-1,1'-dibenzyl-7'-methyl-4'-(*p*-tolyl)-1',4'-dihydro-6'*H*-spiro[indoline-3,5'-pyrrolo[3,4-*b*]azepine]-2,6',8'(7'*H*)-trione (4k):**





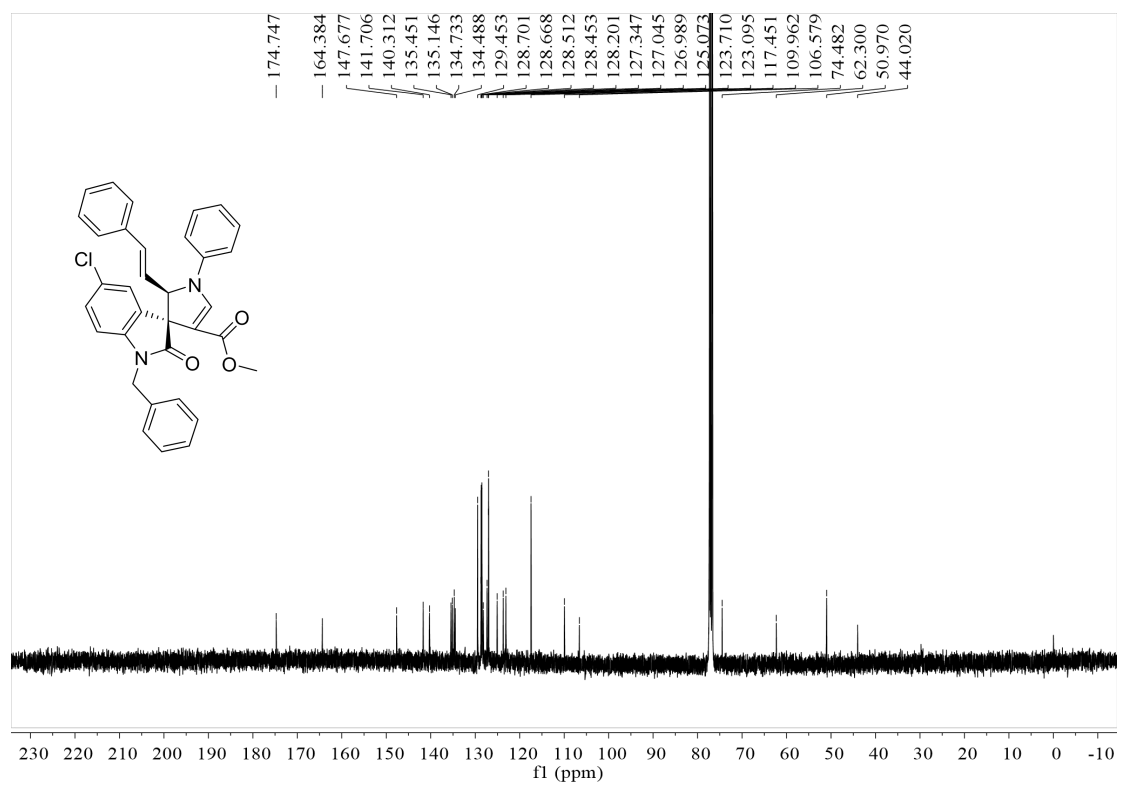
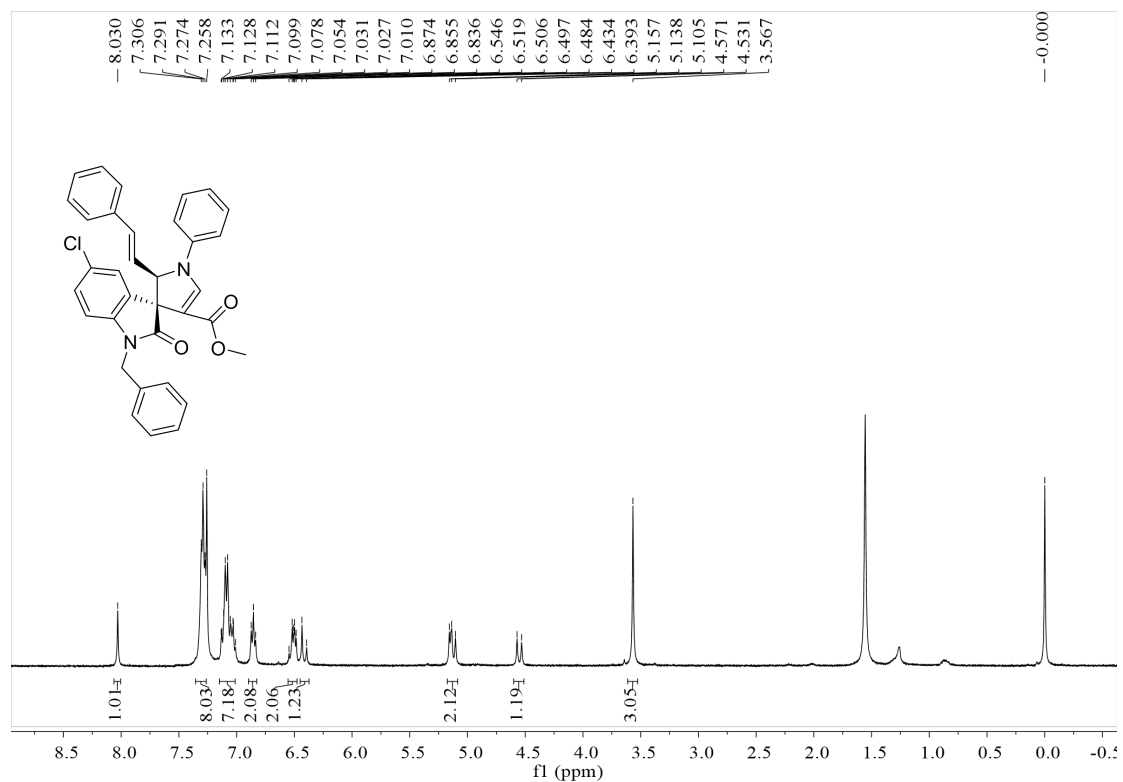
**Methyl-*cis*-1-benzyl-2-oxo-1'-phenyl-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6a):**

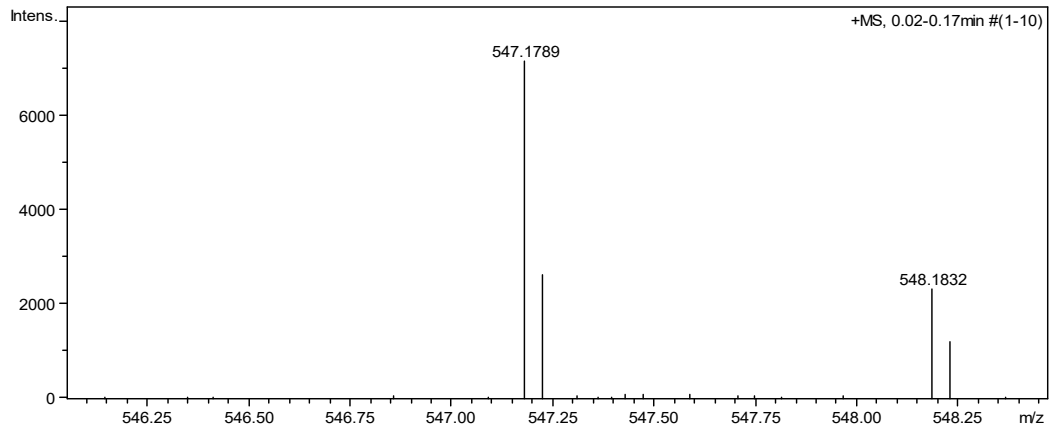




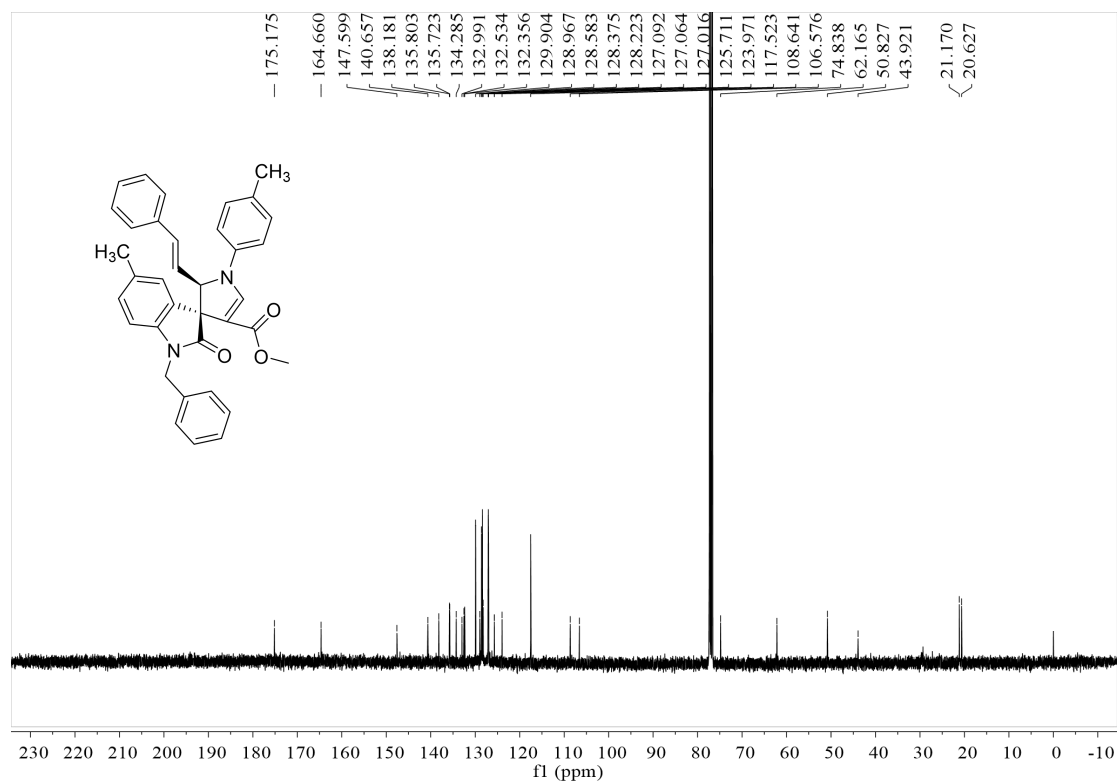
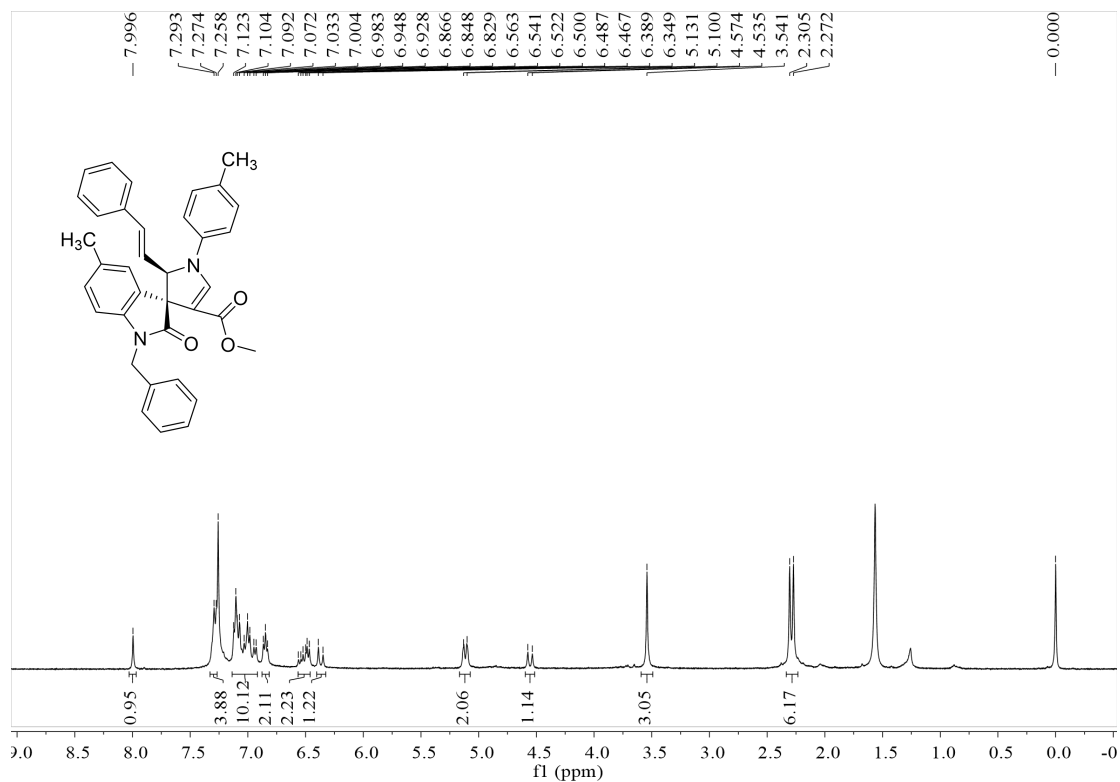


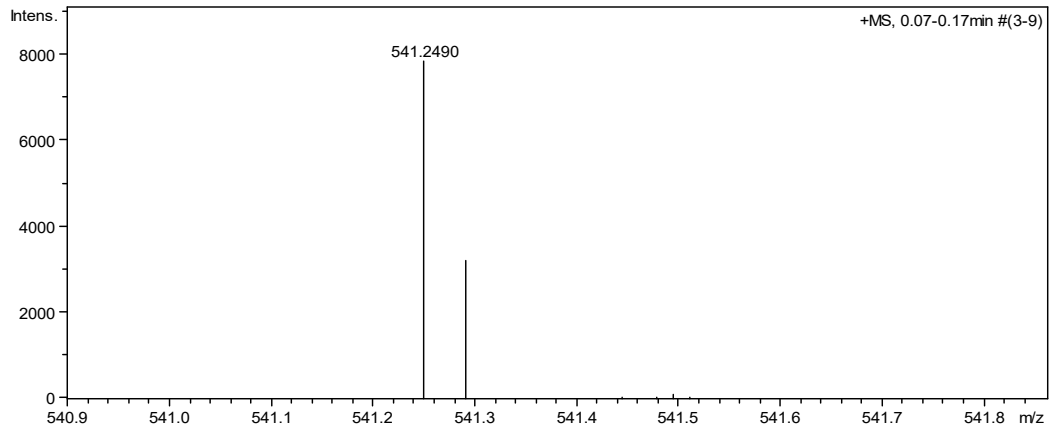
**Methyl-*cis*-1-benzyl-5-chloro-2-oxo-1'-phenyl-2'-((*E*)-styryl)-1,2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6b):**



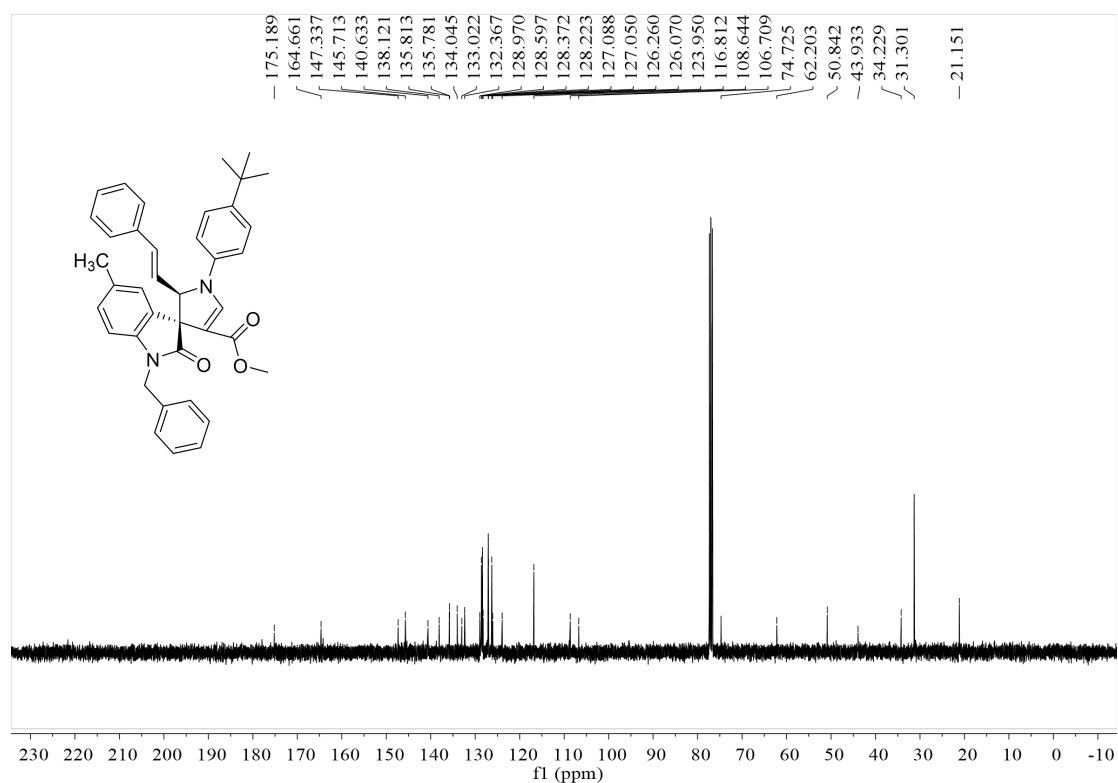
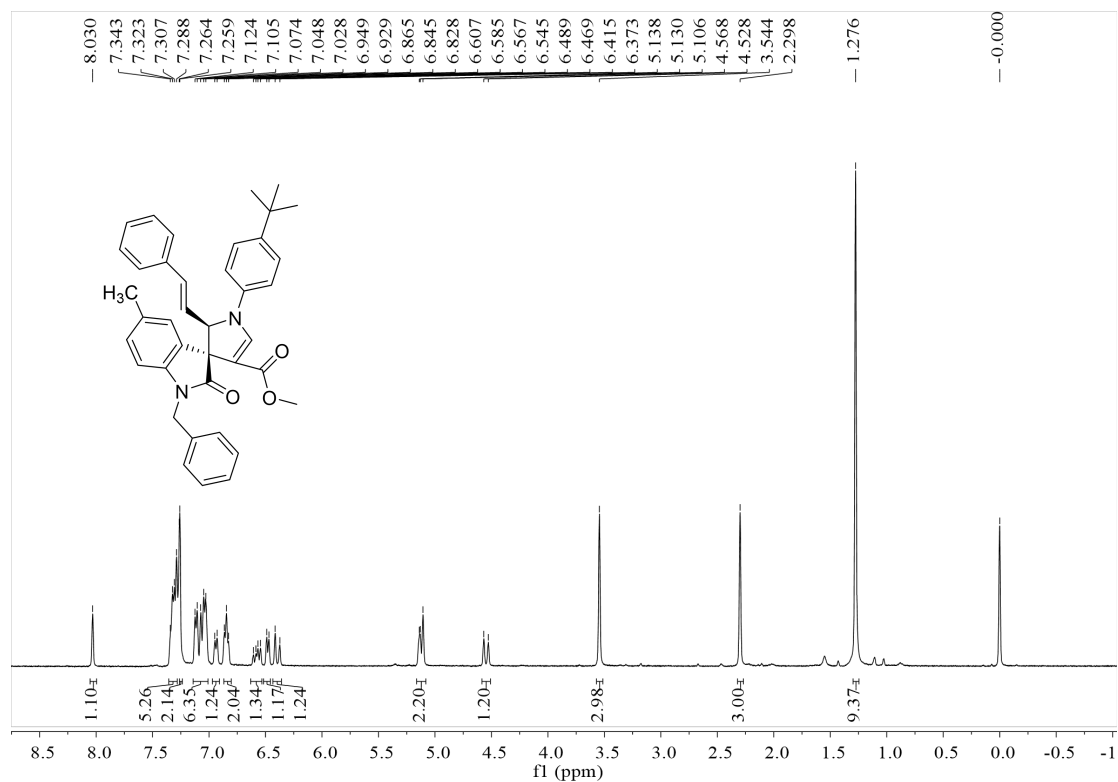


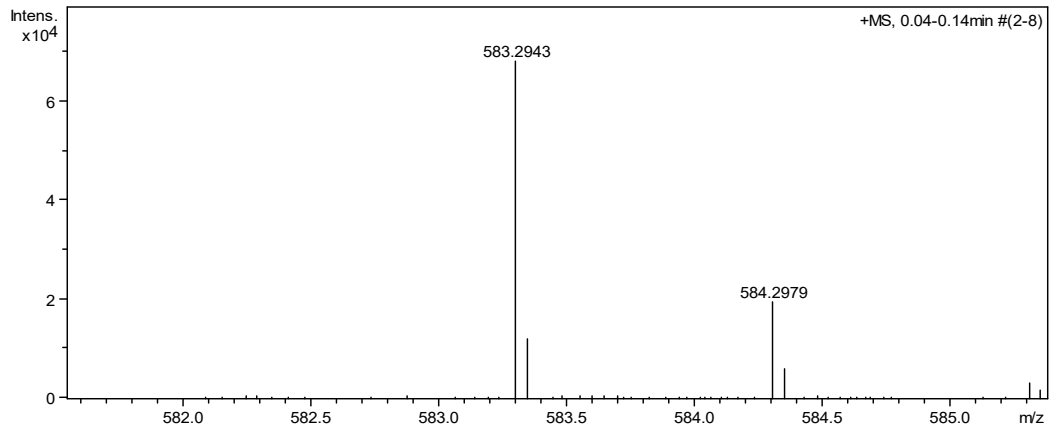
**Methyl-*cis*-1-benzyl-5-methyl-2-oxo-2'-((*E*)-styryl)-1'-(*p*-tolyl)-1',2'-dihydrospiro[indoline-3, 3'-pyrrole]-4'-carboxylate (6c):**



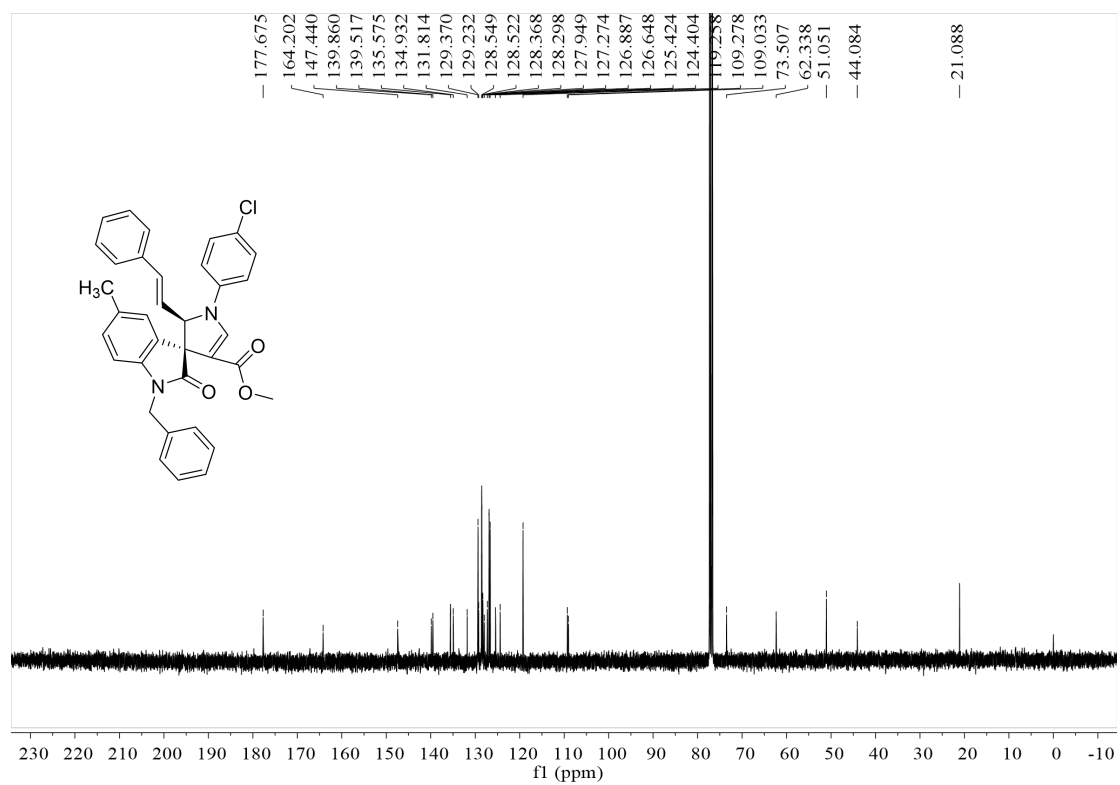
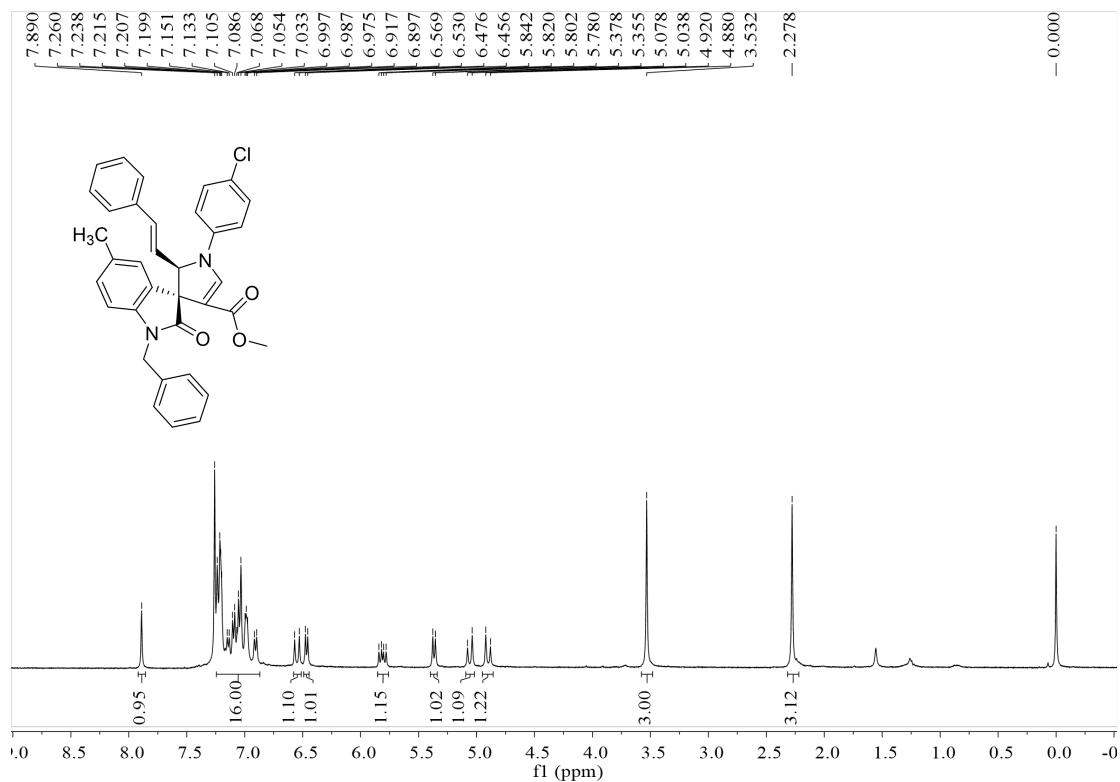


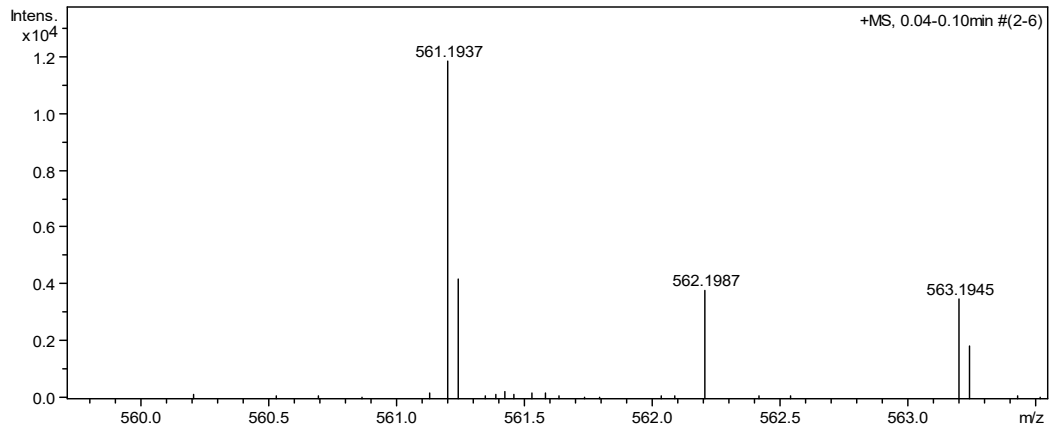
**Methyl-*cis*-1-benzyl-1'-(4-(*tert*-butyl)phenyl)-5-methyl-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6d):**





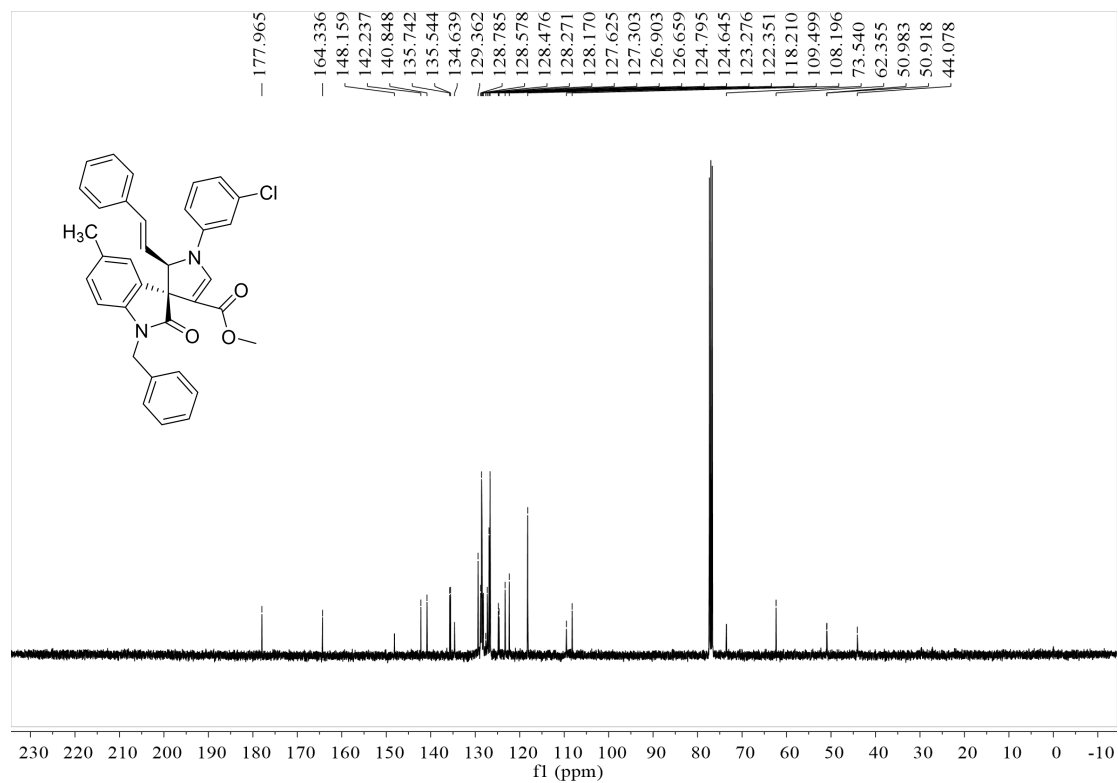
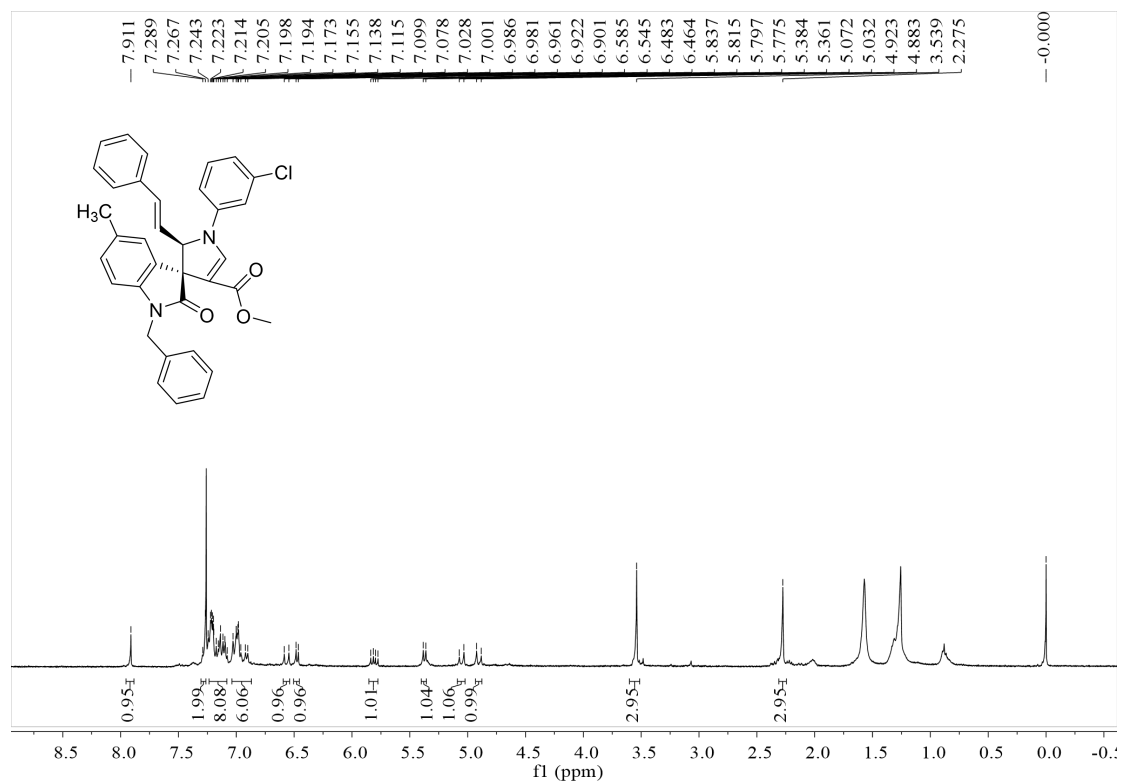
**Methyl-*cis*-1-benzyl-1'-(4-chlorophenyl)-5-methyl-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[in  
doline-3,3'-pyrr (6e):**

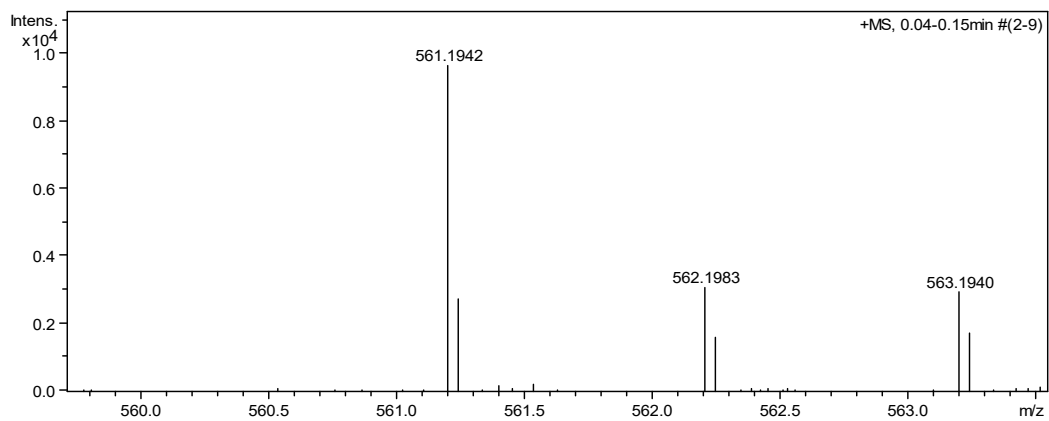




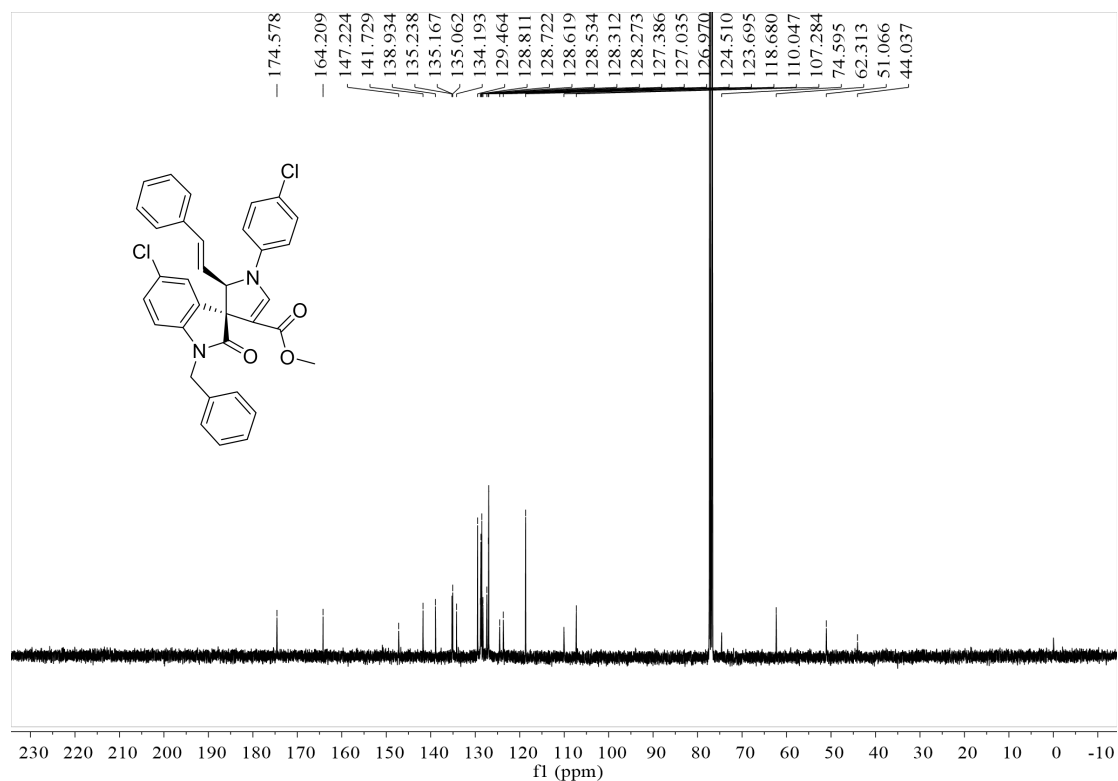
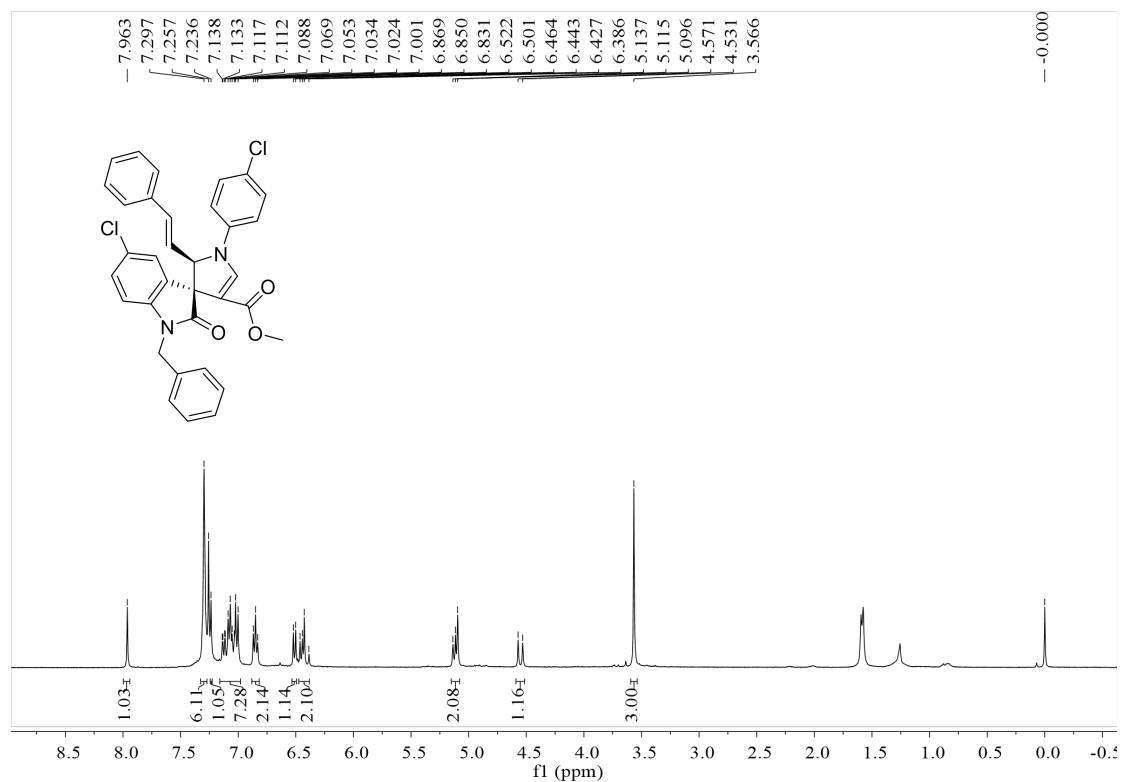


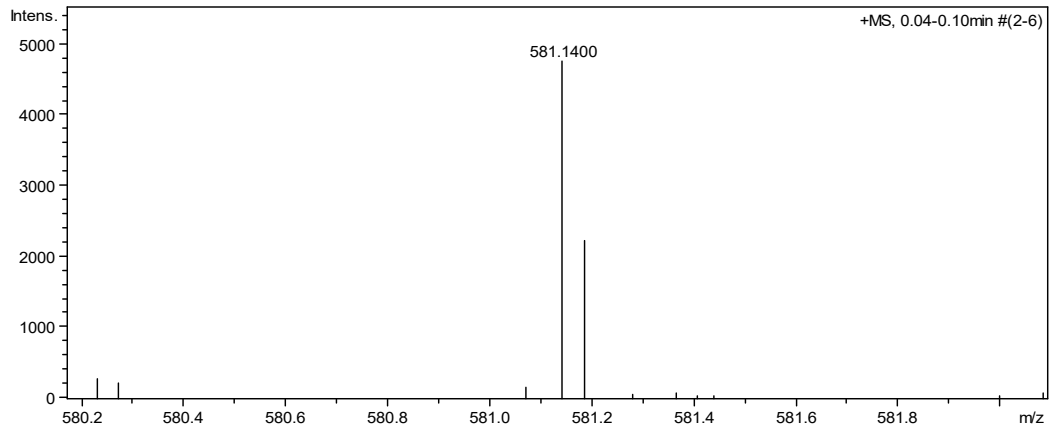
**Methyl-*cis*-1-benzyl-1'-(3-chlorophenyl)-5-methyl-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylateole]-4'-carboxylate (6f):**



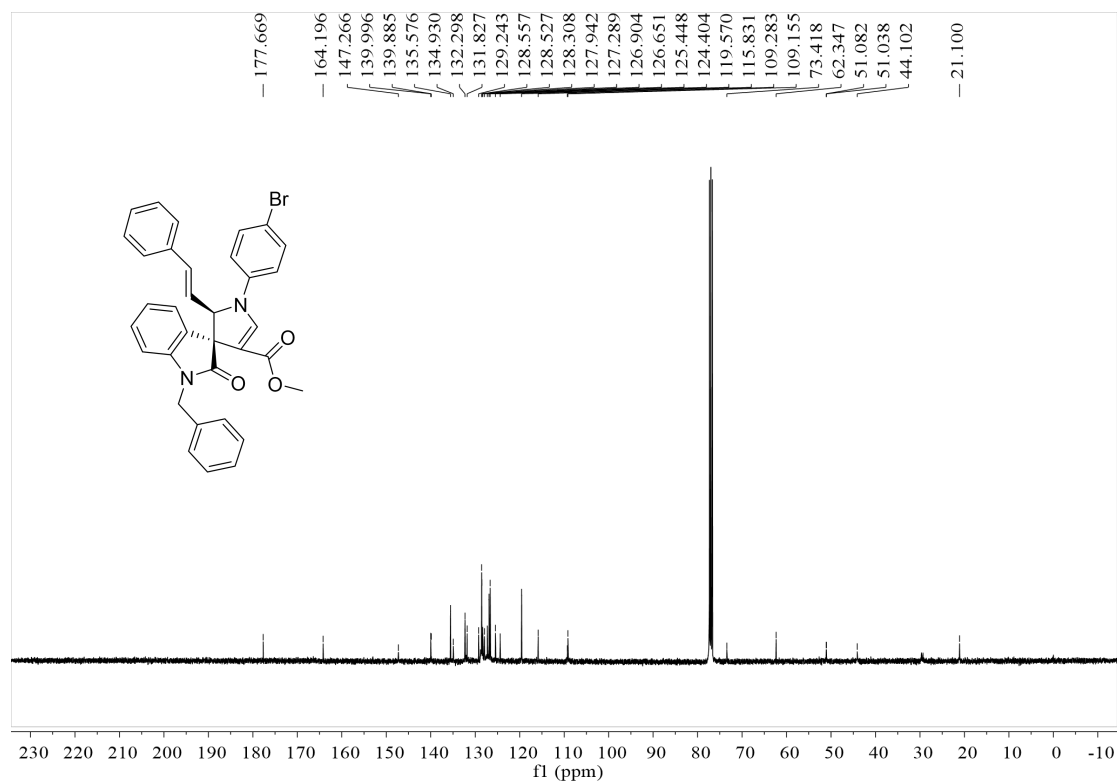
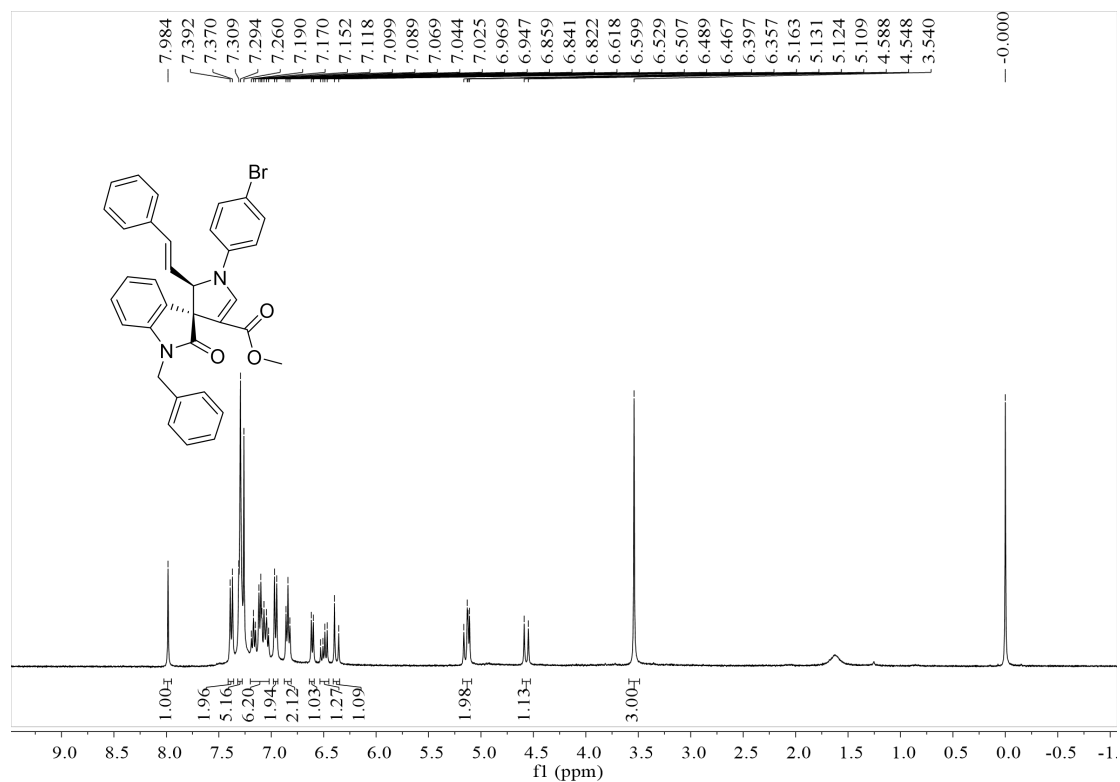


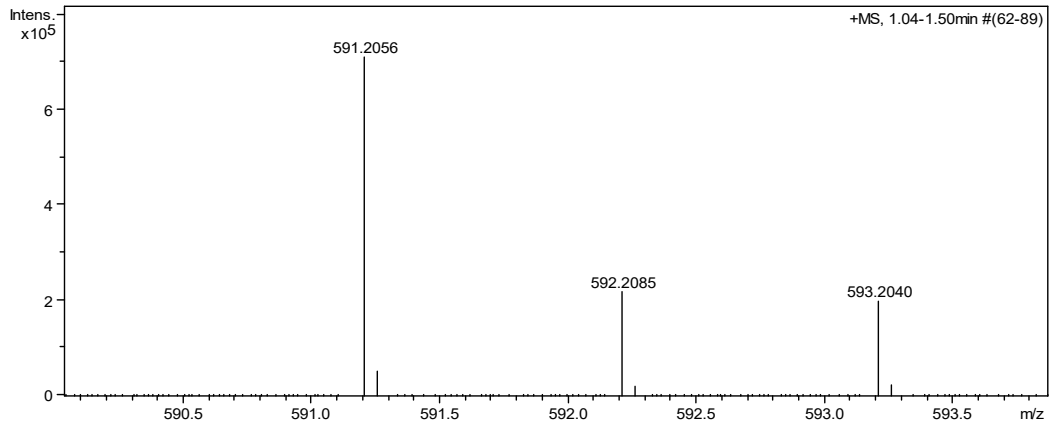
**Methyl-*cis*-1-benzyl-5-chloro-1'-(4-chlorophenyl)-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6g):**



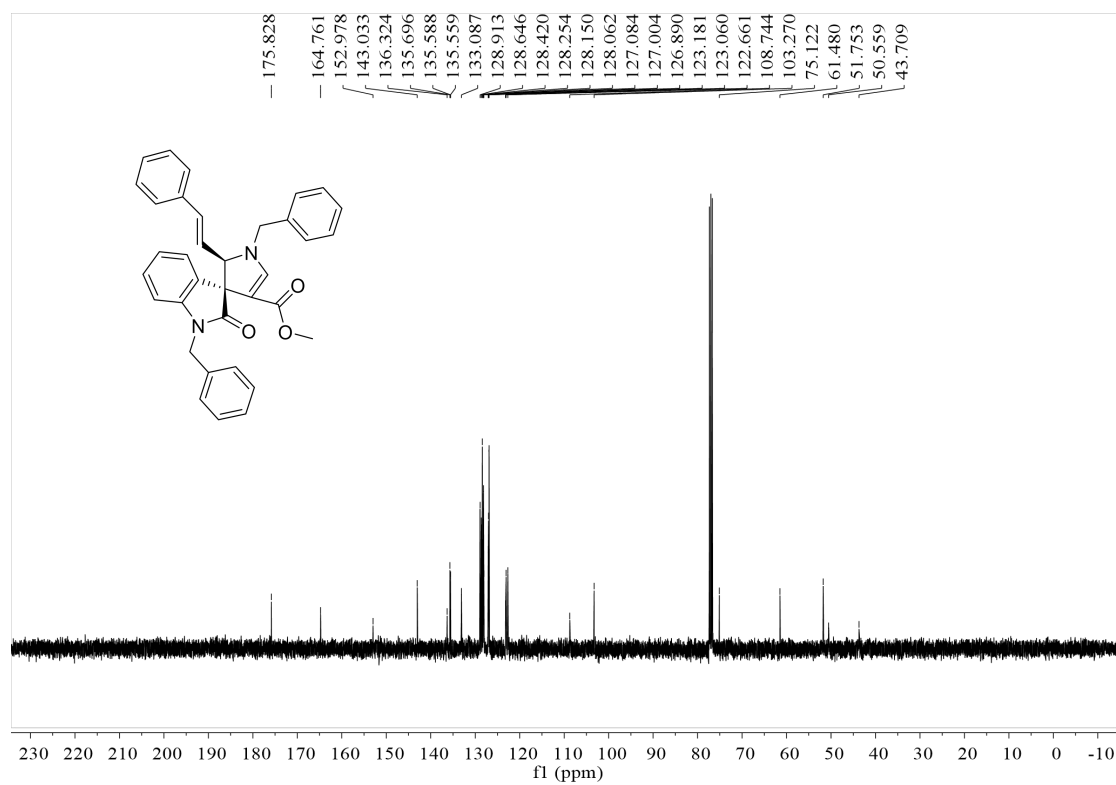
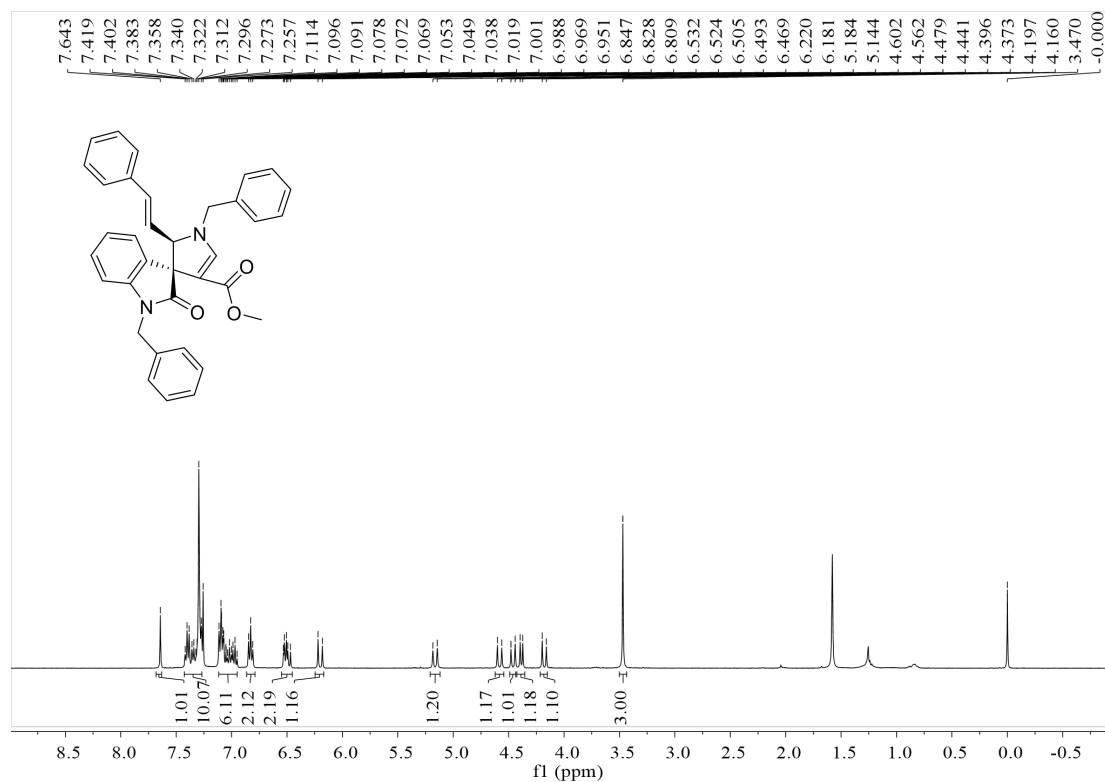


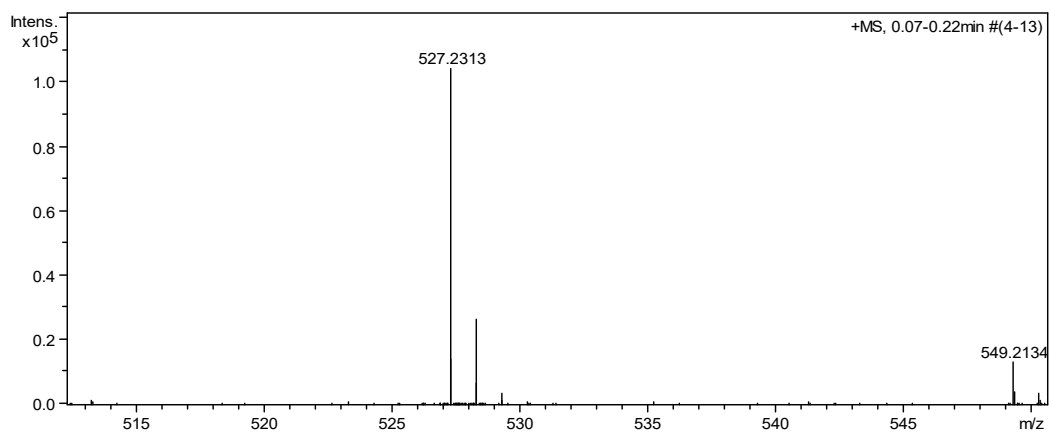
**Methyl-*cis*-1-benzyl-1'-(4-bromophenyl)-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6h):**





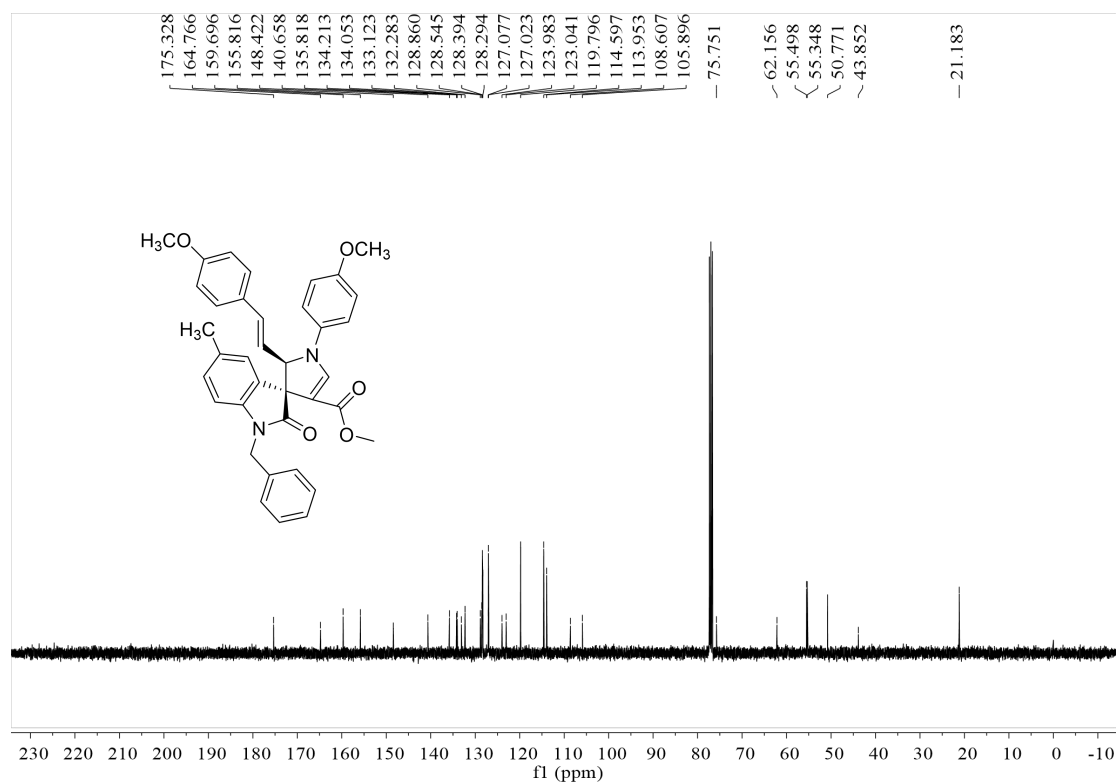
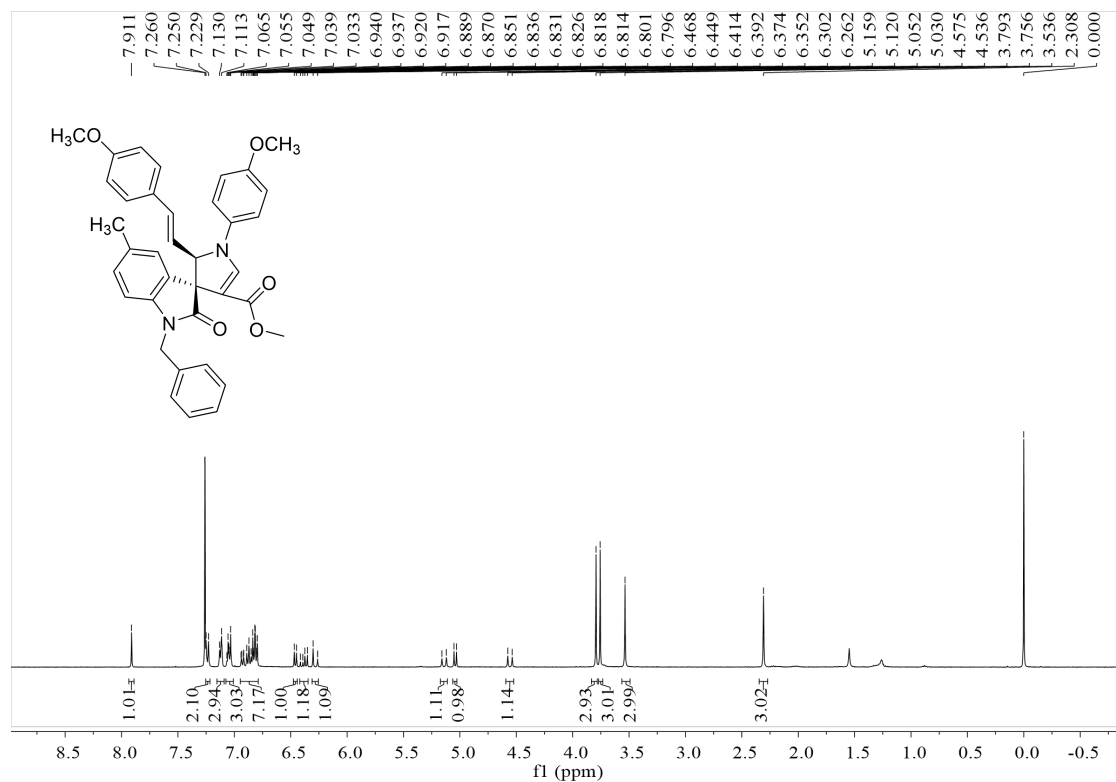
**Methyl-*cis*-1,1'-dibenzyl-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6i):**

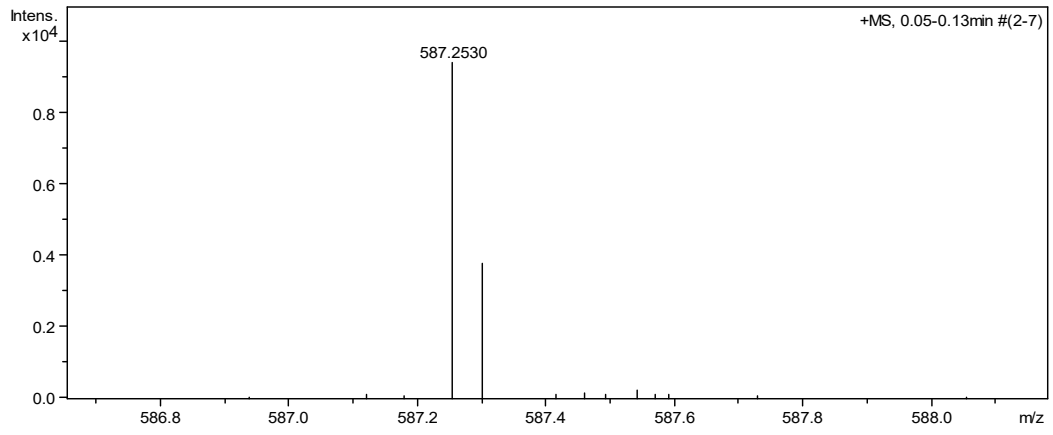




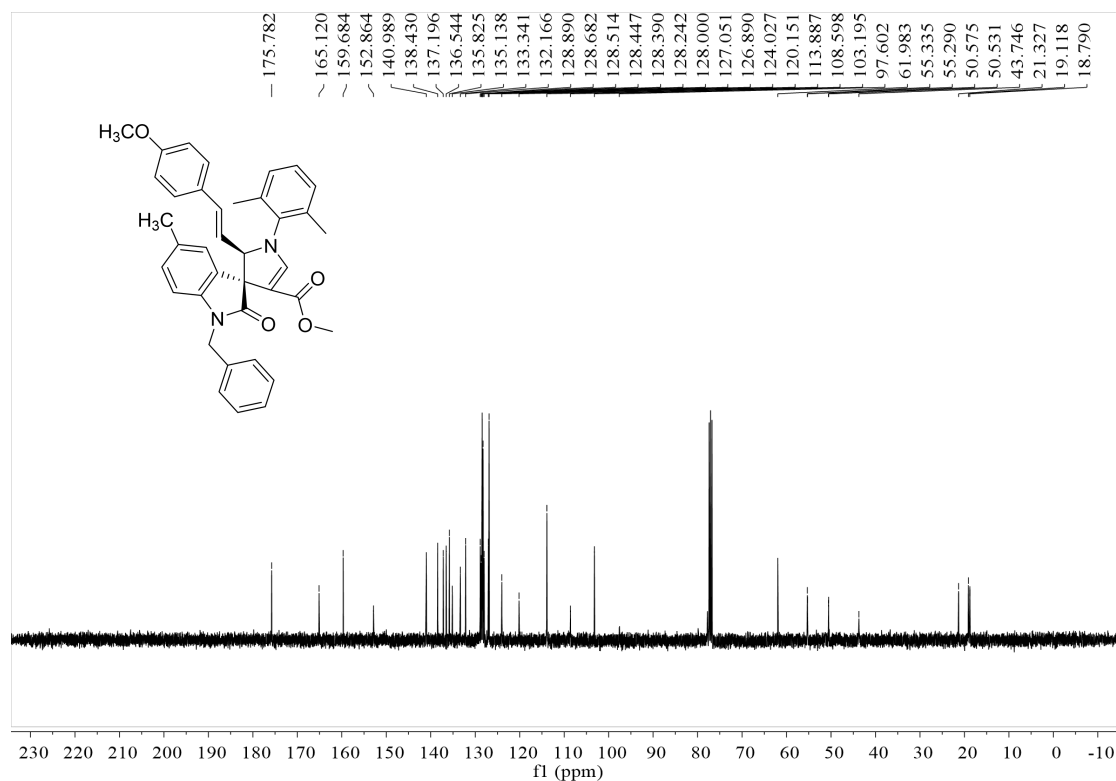
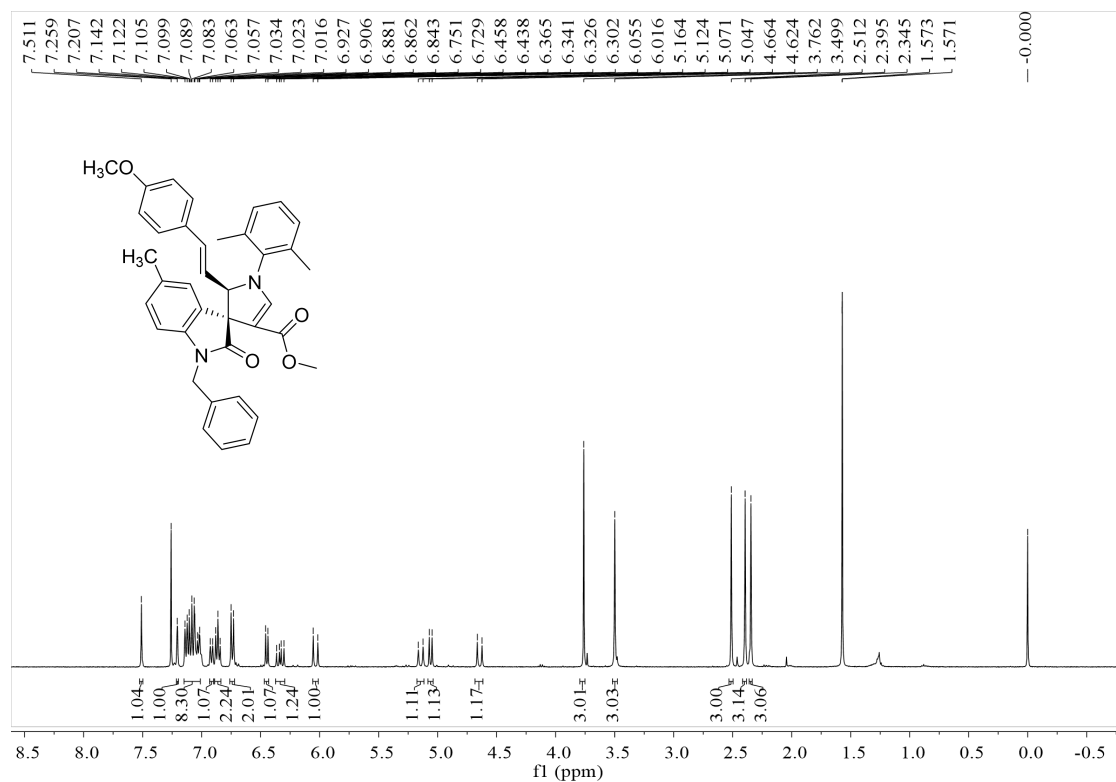


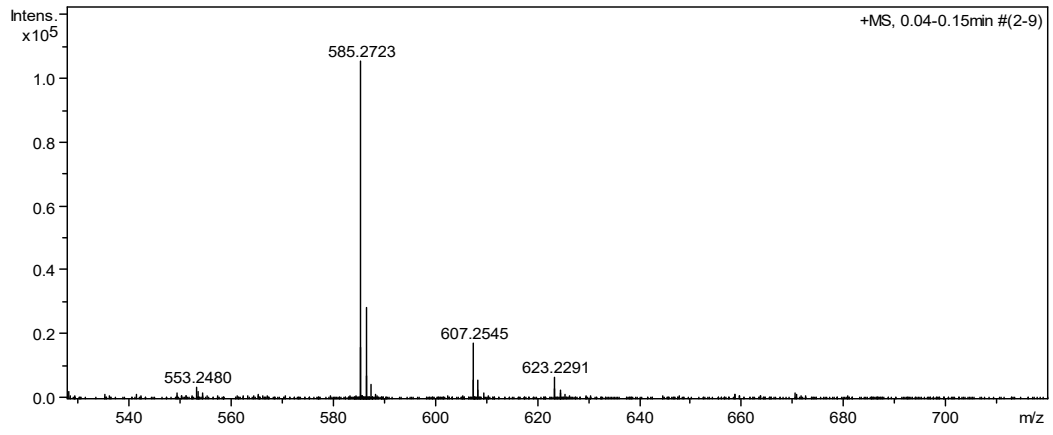
**Methyl-*cis*-1-benzyl-1'-(4-methoxyphenyl)-2'-((*E*)-4-methoxystyryl)-5-methyl-2-oxo-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6j):**



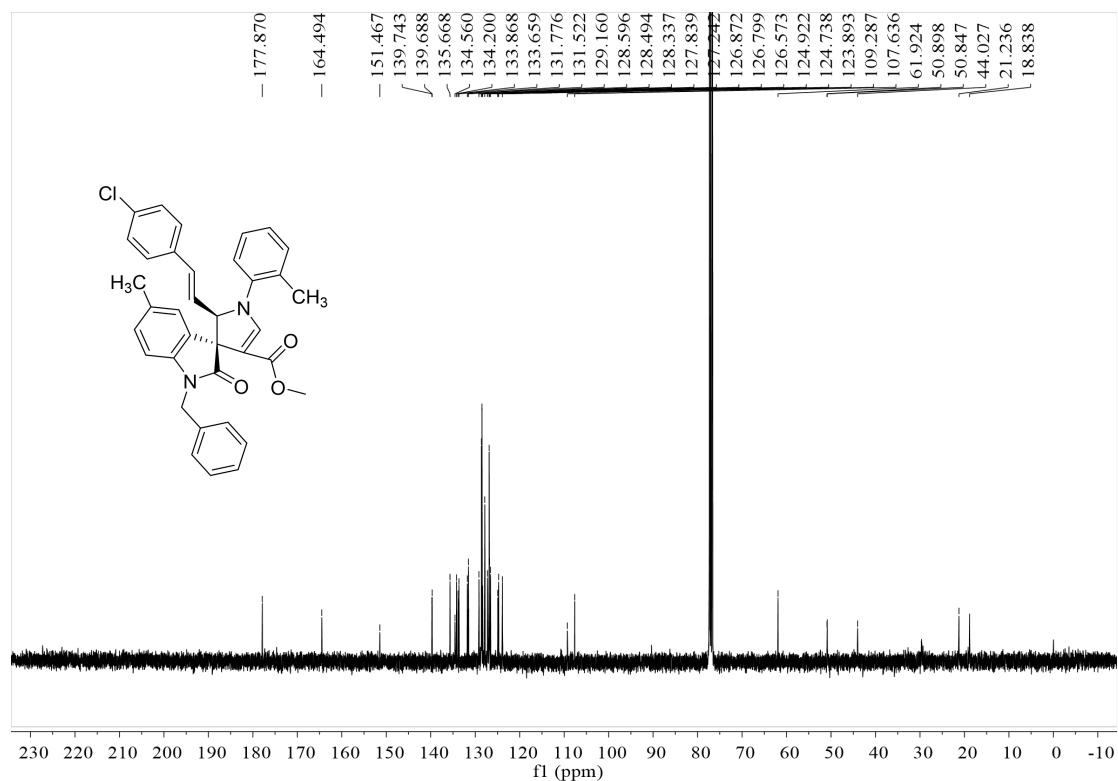
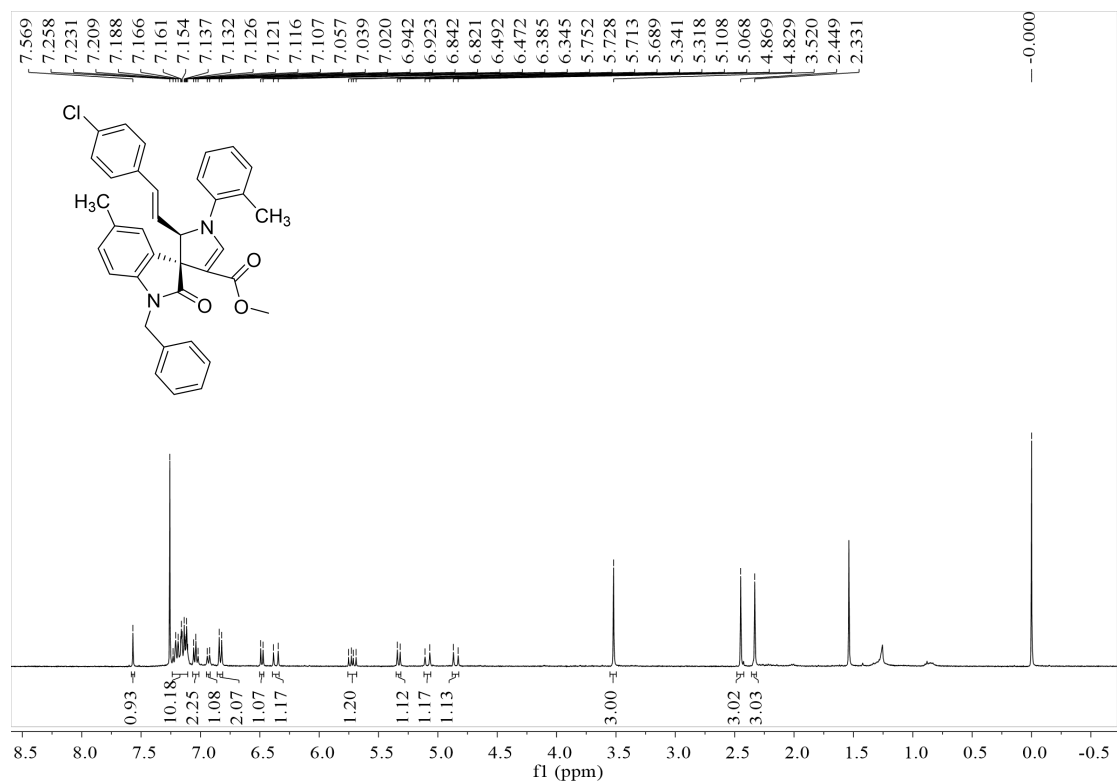


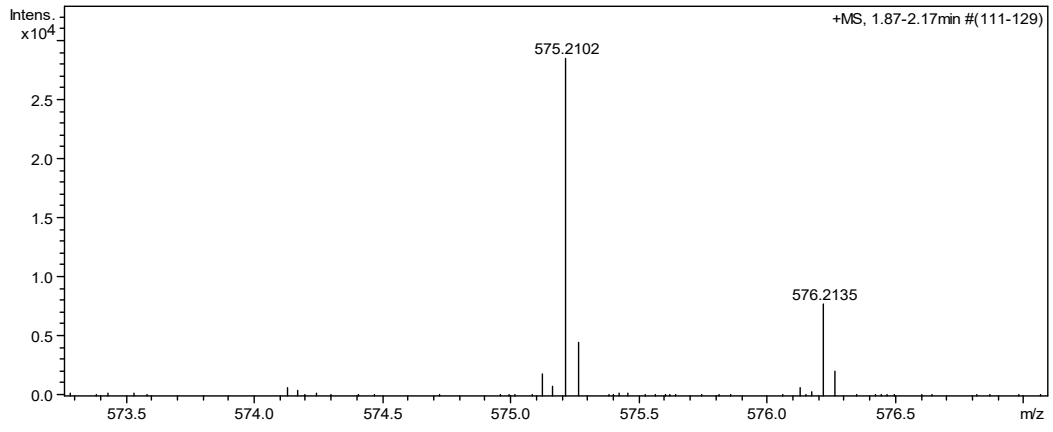
**Methyl-*cis*-1-benzyl-1'-(2,6-dimethylphenyl)-2'-(*E*)-4-methoxystyryl-5-methyl-2-oxo-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate (6k):**



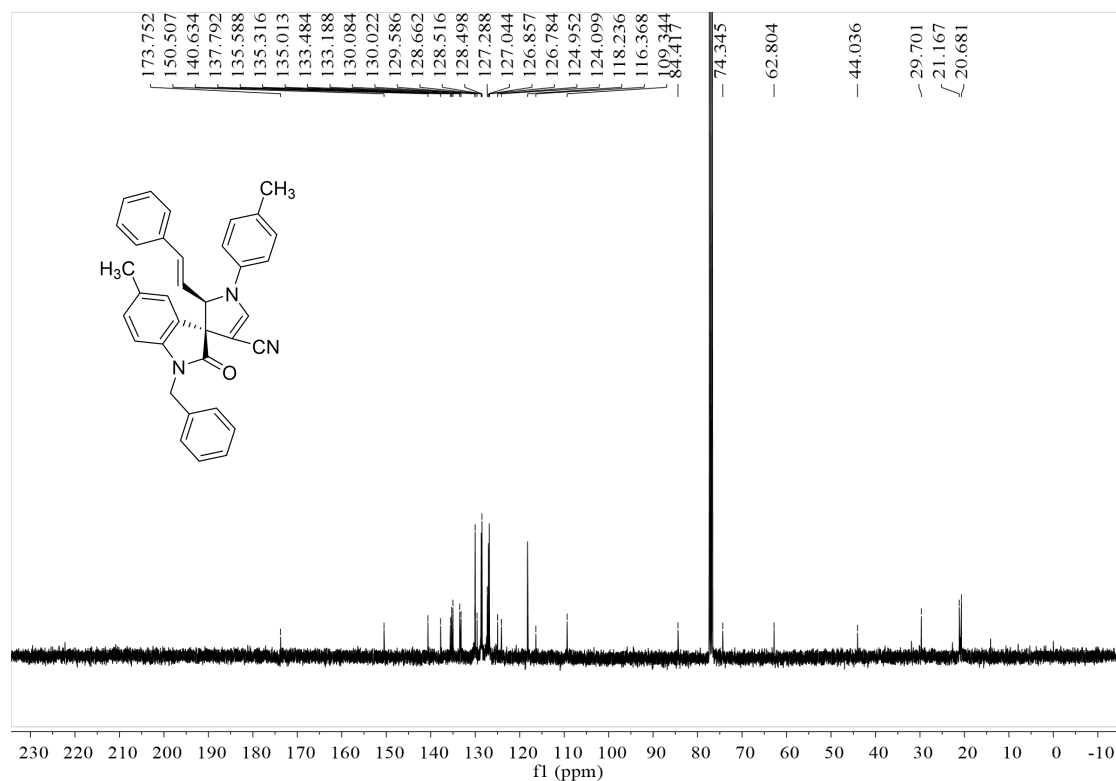
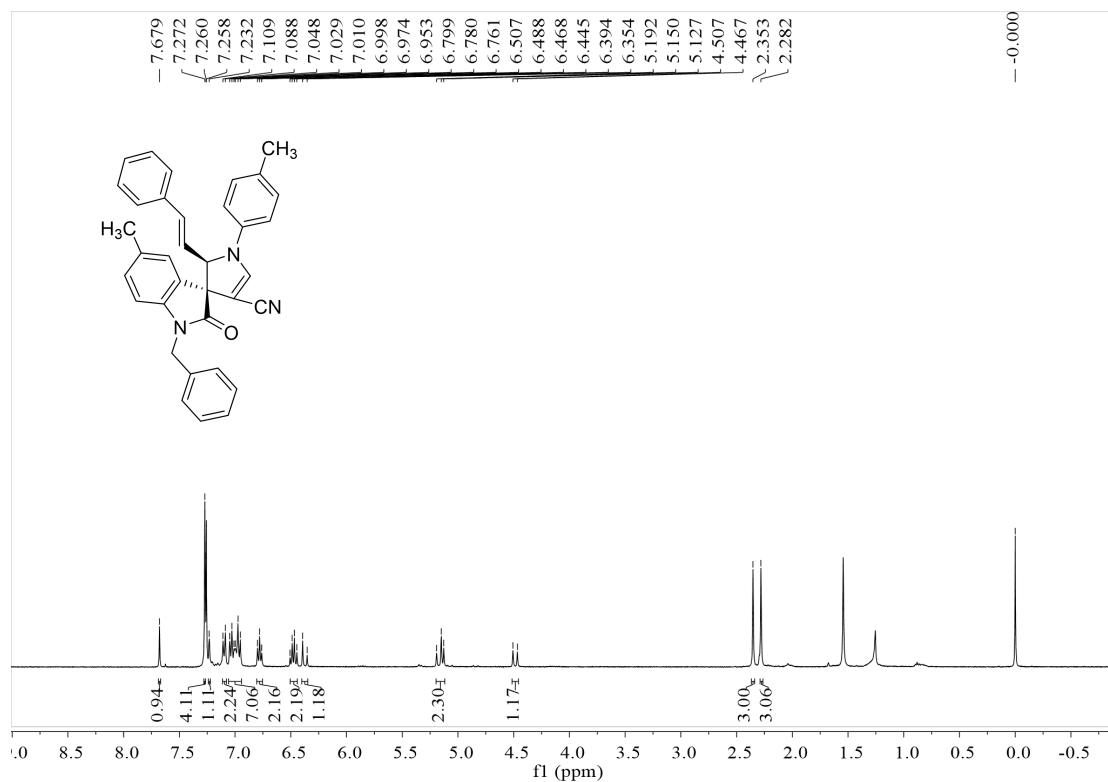


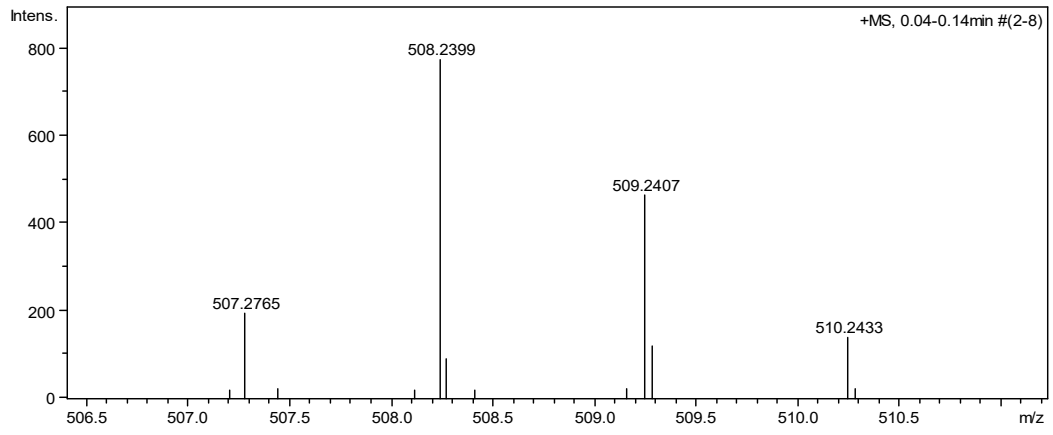
**Methyl-*cis*-1-benzyl-2'-((*E*)-4-chlorostyryl)-5-methyl-2-oxo-1'-(*o*-tolyl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carboxylate(6l):**





**cis-1-benzyl-5-methyl-2-oxo-2'-((E)-styryl)-1'-(p-tolyl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carbonitrile(6m):**







***cis*-1-benzyl-1'-(4-methoxyphenyl)-5-methyl-2-oxo-2'-((*E*)-styryl)-1',2'-dihydrospiro[indoline-3,3'-pyrrole]-4'-carbonitrile (6n):**

