Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Selective Construction of Polycyclic Cyclohepta[*b*]indoles and Cyclopenta[*b*]indoles via Cycloaddition Reaction of 3-(Indol-3-yl)maleimides and (Indol-2-yl)methanols

Jing Sun, Chen Yan, Qiu Sun,* Ying Han, Chao-Guo Yan*

Supporting Information

Single crystals were grown by slow evaporation of concentrated solution in $CHCl_3$ / DCM / EtOH / DMF / DMSO(compounds **3a, 3c, 5c, 5d, 5e, 6k, 8b, 8e, 9a**) in glass vials, which were then sealed by plugs with needles on them.

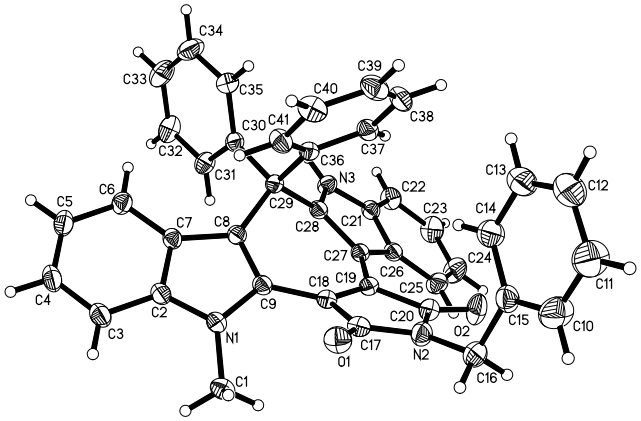


Fig. s1 Single crystal structure of the compound **3c**

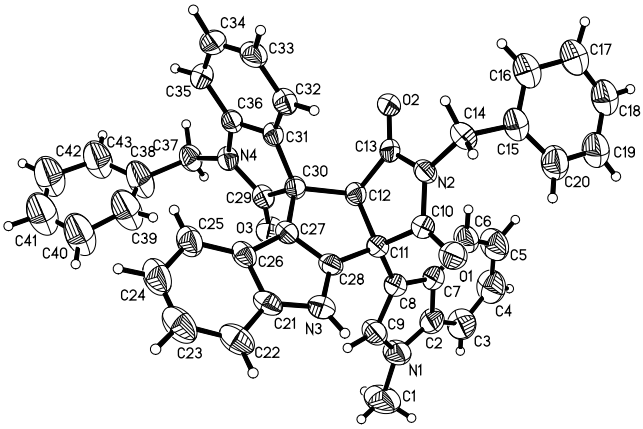


Fig. s2 Single crystal structure of the compound **5d**

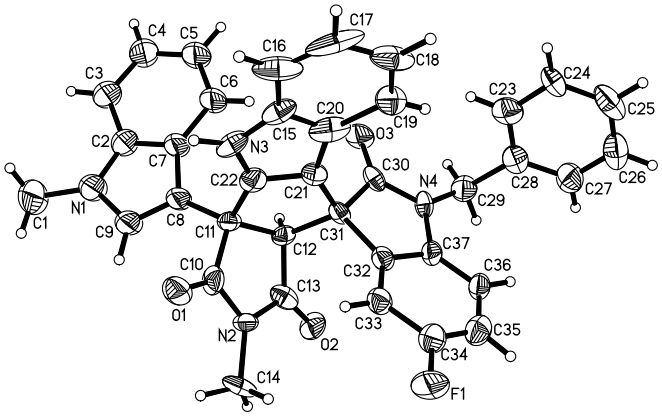


Fig. s3 Single crystal structure of the compound **5e**

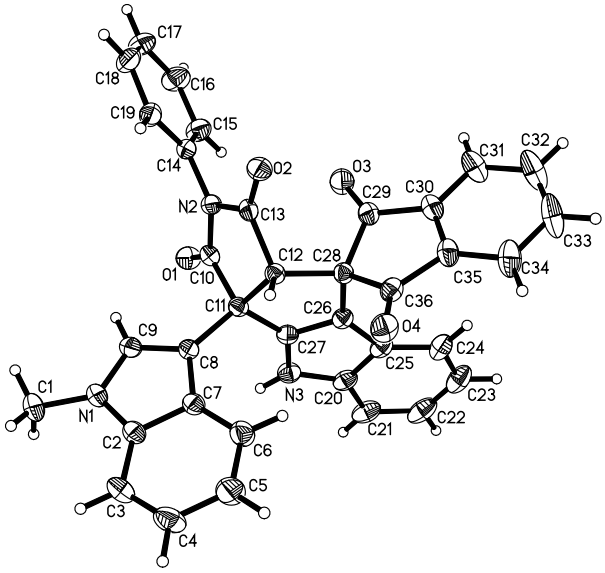


Fig. s5 Single crystal structure of the compound **8b**

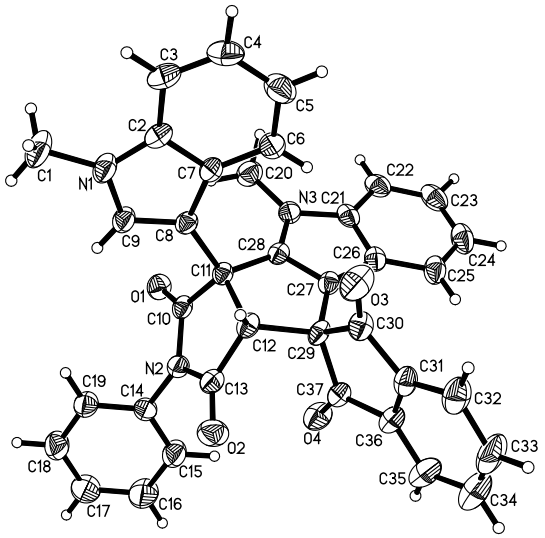


Fig. s6 Single crystal structure of the compound **8e**

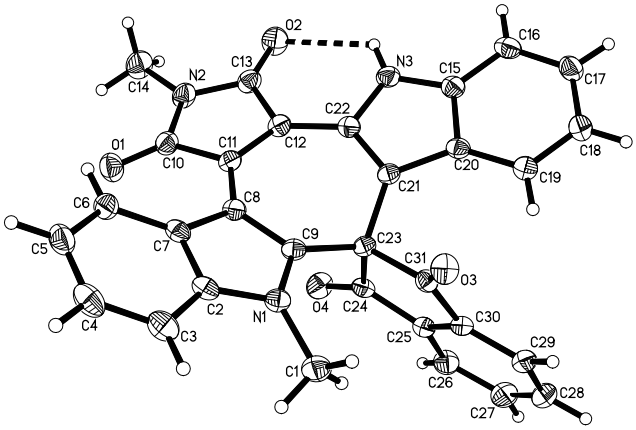


Fig. s7 Single crystal structure of the compound **9a**

Table S1 The single crystal date of compounds **3a, 3c, 5c**

Phase	**3a**	**3c**	**5c**
Empirical formula	$C_{81}H_{55}Cl_3N_6O_4$	$C_{41}H_{29}N_3O_2$	$C_{37}H_{27}FN_4O_3$
Formula weight	1282.66	595.67	594.62
Temperature/K	296(2)	296(2)	296(2)
Wavelength/ Å	0.71073	0.71073	0.71073
Crystal system	Triclinic	Triclinic	Orthorhombic
Space group	P-1	P-1	Pca2(1)
a /Å	13.705(3)	11.3215(4)	30.155(3)
b / Å	13.864(3)	12.4187(5)	9.1523(9)
c / Å	17.875(5)	13.2902(5)	25.150(3)
α (°)	85.290(7)	114.1368(12)	90
β (°)	74.680(7)	110.5982(12)	90
γ (°)	82.395(7)	97.3589(13)	90
V (Å ^{3})	3242.8(14)	1513.15(10)	6941.2(12)
Z	2	2	8
Calculated density	1.314	1.307	1.138
Absorption	0.200	0.081	0.077
F(000)	1332	624	2480
θ range /(°)	2.019 to 24.998	2.293 to 25.998	2.326 to 24.997
Limiting indices	-16<=h<=16, -16<=k<=16, -20<=l<=21	-13<=h<=13, -15<=k<=15, -16<=l<=16	-35<=h<=33, -10<=k<=9, -29<=l<=29
Reflections collected/unique	42255 / 11395 [R(int) = 0.2825]	21473/5890 [R(int) = 0.0299]	67793 / 12103 [R(int) = 0.1808]
Completeness to theta	99.7 %	99.0 %	99.9 %
Max. and min.	0.7456 and 0.4103	0.7455 and 0.7077	0.7456 and 0.6541
Refinement method	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2
Data/restraints/paramet	11395 / 0 / 850	5890/174/462	12103 / 213 / 816
Goodness-of-fit on F^2	0.906	1.027	0.958
Final *R* indices[I>2sigma(I)]	R1 = 0.1218, wR2 = 0.2426	R1 = 0.0456, wR2 = 0.1072	R1 = 0.1035, wR2 = 0.2609
R indices (all data)	R1 = 0.3352, wR2 = 0.2998	R1 = 0.0676, wR2 = 0.1192	R1 = 0.2335, wR2 = 0.3010
Largest diff. peak and hole /(e ·Å^{-3})	0.518 and -0.491	0.208 and -0.283	0.734 and -0.363

Table S2 The single crystal date of compounds **5d, 5e, 6k**

Phase	**5d**	**5e**	**6k**
Empirical formula	$C_{36}H_{33}Cl_3N_4O_3$	$C_{43}H_{32}N_4O_3$	$C_{43}H_{41}N_5O_4$
Formula weight	676.01	652.72	691.81
Temperature/K	296(2)	296(2)	296(2)
Wavelength/ Å	0.71073	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic	Triclinic
Space group	P2(1)/n	C2/c	P-1
a /Å	17.200(4)	29.965(6)	11.3858(5)
b / Å	9.5522(19)	10.1561(19)	11.7752(5)
c / Å	22.075(5)	27.430(6)	15.6044(7)
α (°)	90	90	77.5323(13)
β (°)	110.167(7)	101.793(6)	83.5022(14)
γ (°)	90	90	66.2724(12)
V (Å ^{3})	3404.6(13)	8171(3)	1869.26(14)
Z	4	8	2
Calculated density	1.319	1.061	1.229
Absorption	0.311	0.068	0.080
F(000)	1408	2736	732
θ range /(°)	1.847 to 25.000	2.122 to 24.998	1.337 to 26.000
Limiting indices	-20<=h<=20, -11<=k<=11, -26<=l<=23	-35<=h<=32, -11<=k<=12, -32<=l<=32	-14<=h<=13, -14<=k<=14, -19<=l<=19
Reflections collected/unique	26286/6002 [R(int) = 0.0563]	34118/7151 [R(int) = 0.0533]	26708/7294 [R(int) = 0.0386]
Completeness to theta	99.7 %	99.4 %	99.2 %
Max. and min.	0.7456 and 0.6494	0.7456 and 0.6580	0.7456 and 0.7125
Refinement method	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2
Data/restraints/paramet	6002/23/419	7151/246/499	7294/0/474
Goodness-of-fit on F^2	1.065	0.980	1.018
Final *R* indices[I>2sigma(I)]	R1 = 0.1006, wR2 = 0.3019	R1 = 0.1073, wR2 = 0.3132	R1 = 0.0569, wR2 = 0.1353
R indices (all data)	R1 = 0.1592, wR2 = 0.3344	R1 = 0.1579, wR2 = 0.3406	R1 = 0.0934, wR2 = 0.1567
Largest diff. peak and hole /(e ·Å^{-3})	0.412 and -0.779	0.942 and -0.495	0.481 and -0.292

Table S3 The single crystal date of compounds **8b, 8e, 9a**

Phase	**8b**	**8e**	**9a**
Empirical formula	$C_{39}H_{30}N_4O_5$	$C_{37}H_{25}N_3O_4$	$C_{31}H_{20}Cl_3N_3O_4$
Formula weight	634.67	575.60	616.86
Temperature/K	296(2)	296(2)	296(2)
Wavelength/ Å	0.71073	0.71073	0.71073
Crystal system	Triclinic	Triclinic	Triclinic
Space group	P-1	P-1	P-1
a /Å	10.131(2)	12.284(3)	12.0107(18)
b / Å	11.507(3)	12.500(3)	12.0920(19)
c / Å	15.601(4)	15.924(3)	12.2492(18)
α (°)	72.289(7)	74.639(7)	118.132(4)
β (°)	86.070(8)	86.466(7)	96.981(4)
γ (°)	68.259(7)	65.105(6)	111.445(4)
V (Å ^{3})	1607.3(7)	2135.3(8)	1363.4(4)
Z	2	2	2
Calculated density (g·cm^{-3})	1.311	0.895	1.503
Absorption coefficient(mm^{-1})	0.088	0.059	0.382
F(000)	664	600	632
θ range /(°)	2.167 to 25.997	1.862 to 25.999	1.951 to 25.997
Limiting indices	-12<=h<=12, -14<=k<=14, -19<=l<=19	-15<=h<=15, -14<=k<=15, -19<=l<=19	-14<=h<=14, -14<=k<=14, -14<=l<=15
Reflections collected/unique	22885/6284 [R(int) = 0.0342]	30584/8361 [R(int) = 0.0569]	15501/5270 [R(int) = 0.0402]
Completeness to theta	99.6 %	99.7 %	98.1 %
Max. and min. transmission		0.7456 and 0.6685	0.7456 and 0.5736
Refinement method	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2
Data/restraints/parameters	6284/0/436	8361/0/399	5270/0/381
Goodness-of-fit on F^2	1.020	1.048	1.052
Final *R* indices[I>2sigma(I)]	R1 = 0.0542, wR2 = 0.1306	R1 = 0.0684, wR2 = 0.1902	R1 = 0.0844, wR2 = 0.2283
R indices (all data)	R1 = 0.0882, wR2 = 0.1502	R1 = 0.1274, wR2 = 0.2192	R1 = 0.1165, wR2 = 0.2536
Largest diff. peak and hole /(e ·Å^{-3})	0.284 and -0.301	0.261 and -0.239	0.796 and -0.863

Experimental section

1. General procedure for the preparation of the compound 3a-3k

To around flask was added 3-(indol-3-yl)maleimides (1.0 mmol), (indol-2-yl)diphenylmethanol (1.0 mmol), chloroform (8.0 mL) and *p*-toluenesulfonic acid monohydrate (0.3 mmol). The mixture was heated at 65 ^{o}C for four hours. After removing the solvent at reduced pressure by rotation evaporation, the residue was subjected to column chromatography with petroleum ether and ethyl acetate (V/V = 10:1) as eluent to give the pure product for analysis.

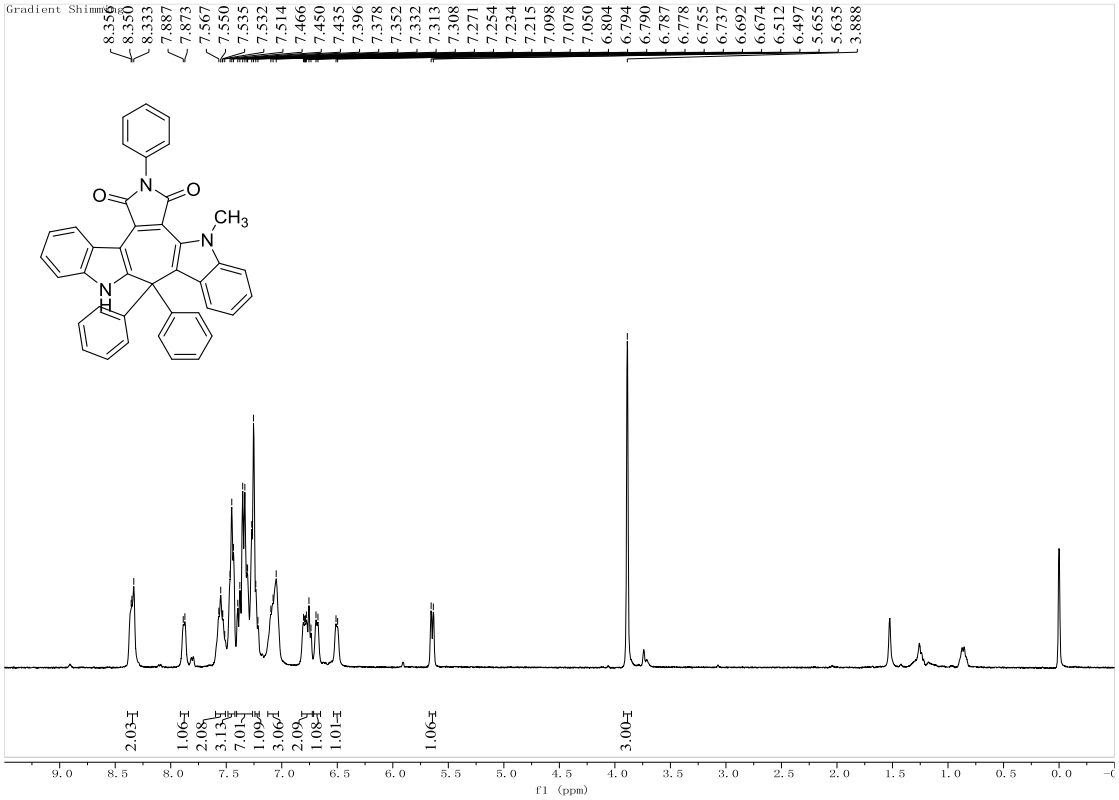
2. General procedure for the preparation of the compound 5a-5k and 6e-6h

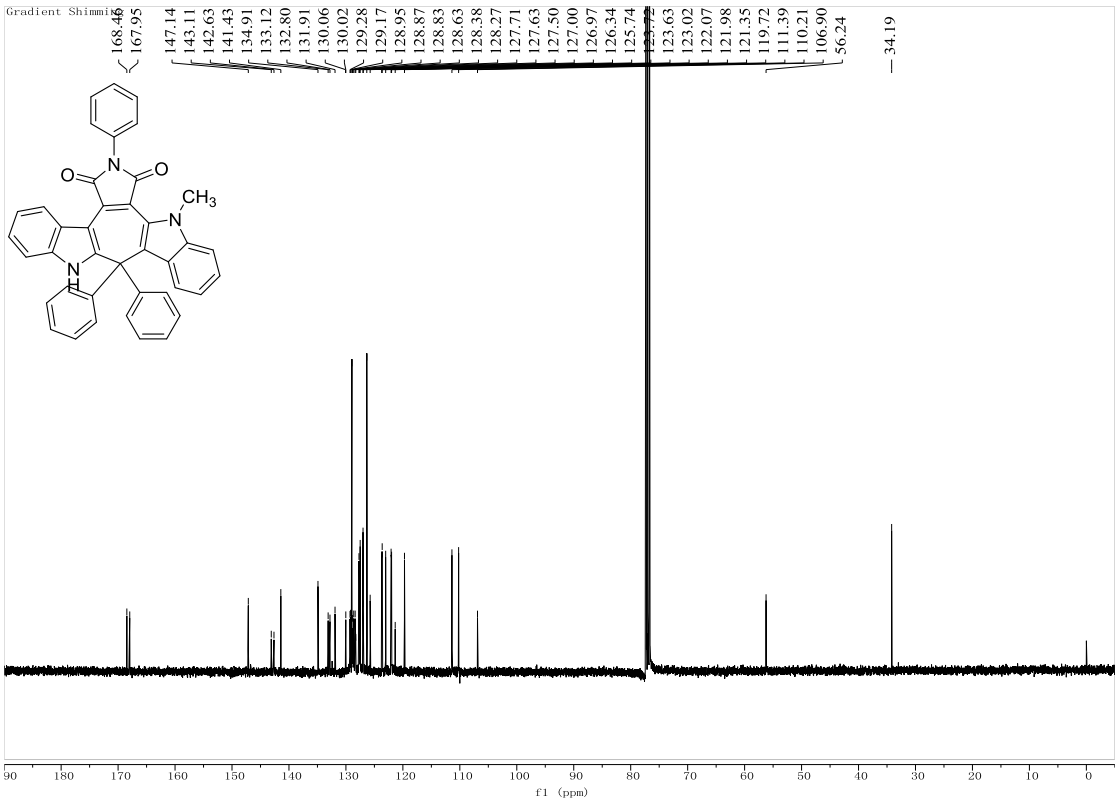
To around flask was added 3-(indol-3-yl)maleimides (1.0 mmol), 3-hydroxy-3-(indol-3-yl)-indolin-2-one (1.0 mmol), acetonitrile (8.0 mL) and *p*-toluenesulfonic acid monohydrate (0.3 mmol). The mixture was heated at 60 ^{o}C for six hours. After removing the solvent at reduced pressure by rotation evaporation, the residue was subjected to column chromatography with petroleum ether and methylene dichloride (V/V = 4:1) as eluent to give the pure product for analysis.

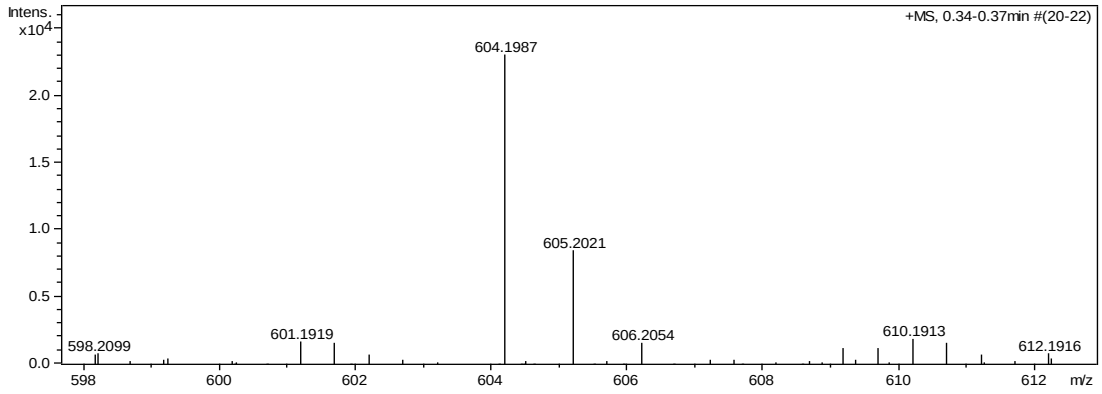
3. General procedure for the preparation of the compound 8a-8f and 9a-9c

To around flask was added 3-(indol-3-yl)maleimides (1.0 mmol), 2-hydroxy-2-(indol-3-yl)-indene-1,3-dione (1.0 mmol), acetonitrile (8.0 mL) and *p*-toluenesulfonic acid monohydrate (0.3 mmol). The mixture was heated at 60 ^{o}C for six hours. After removing the solvent at reduced pressure by rotation evaporation, the residue was subjected to column chromatography with petroleum ether and methylene dichloride (V/V = 4:1) as eluent to give the pure product for analysis.

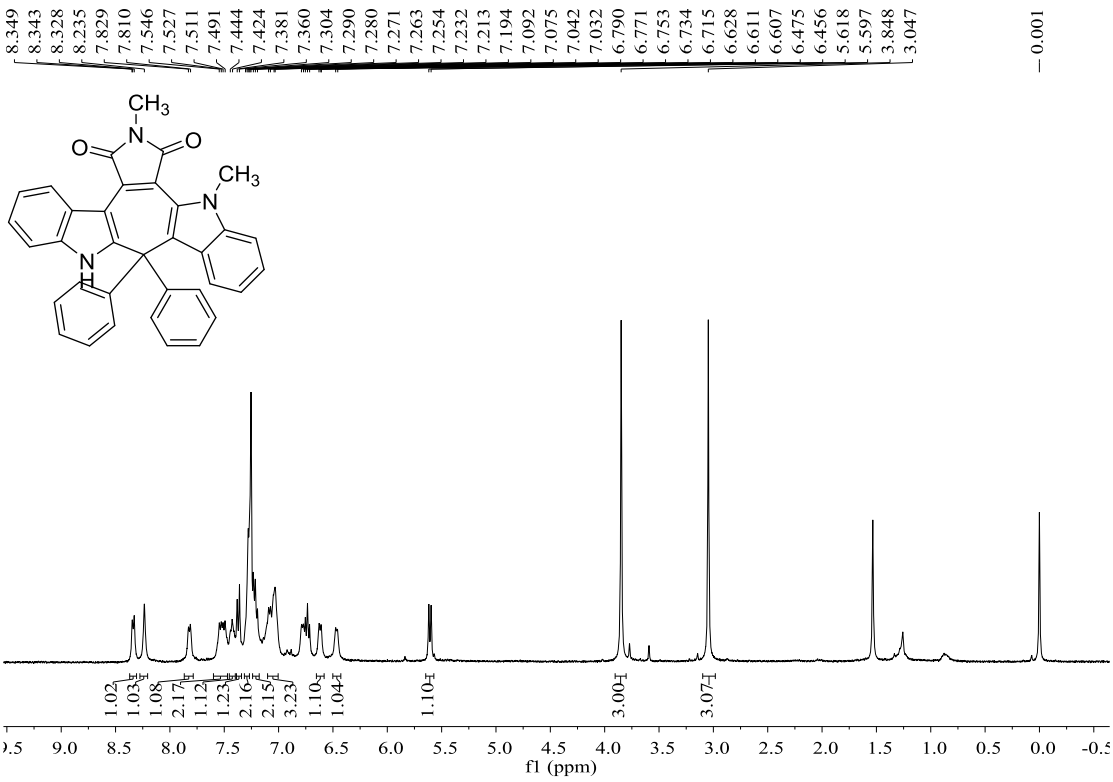
14-methyl-2,9,9-triphenyl-9,14-dihydropyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole-1,3(2*H*,8*H*)-dione (3a): petroleum ether and ethyl acetate (V/V = 10:1) as the eluent, red solid, 394 mg,red solid, 76%, m.p. 235-237 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.36-8.33 (m, 2H, ArH), 7.88 (d, *J* = 6.4 Hz, 1H, ArH), 7.57-7.51 (m, 2H, ArH), 7.45 (t, *J* = 6.0 Hz, 3H, ArH), 7.40-7.22 (m, 8H, ArH), 7.10-7.05 (m, 3H, ArH), 6.80-6.74 (m, 2H, ArH), 6.68 (d, *J* = 7.2 Hz, 1H, ArH), 6.50 (d, *J* = 6.0 Hz, 1H, ArH), 5.65 (d, *J* = 8.0 Hz, 1H, ArH), 3.89 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 168.4, 167.9, 147.1, 143.0, 142.6, 141.4, 134.8, 133.1, 132.7, 131.8, 130.0, 129.9, 129.2, 129.1, 128.9, 128.8, 128.7, 128.6, 128.3, 128.2, 127.7, 127.6, 127.4, 126.9, 126.8, 126.3, 125.7, 123.7, 123.6, 123.0, 122.0, 121.9, 121.3, 119.7, 111.3, 110.2, 106.8, 56.2, 34.1; IR (KBr) υ: 3048, 2911, 2852, 2321, 1890, 1756, 1732, 1628, 1518, 1475, 1448, 1371, 1221, 910, 834, 756 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{40}H_{27}NaN_3O_2$ ([M+Na]$^{+}$): 604.1995, Found: 604.1987.

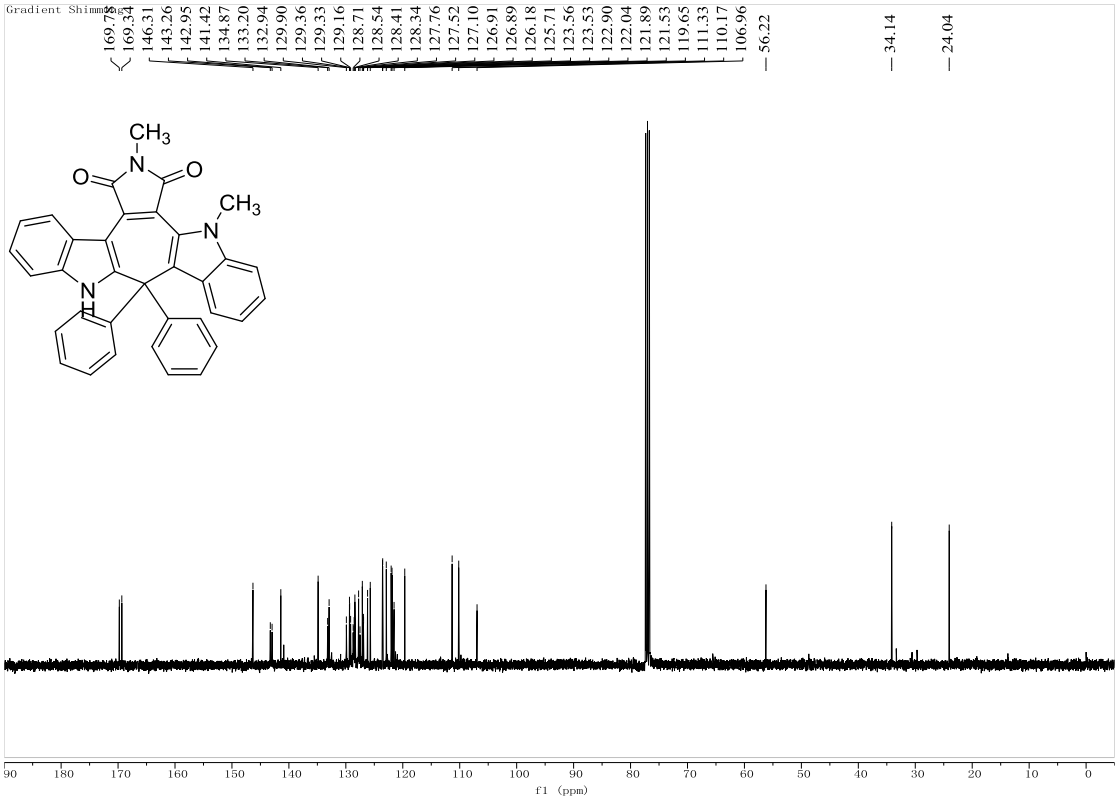


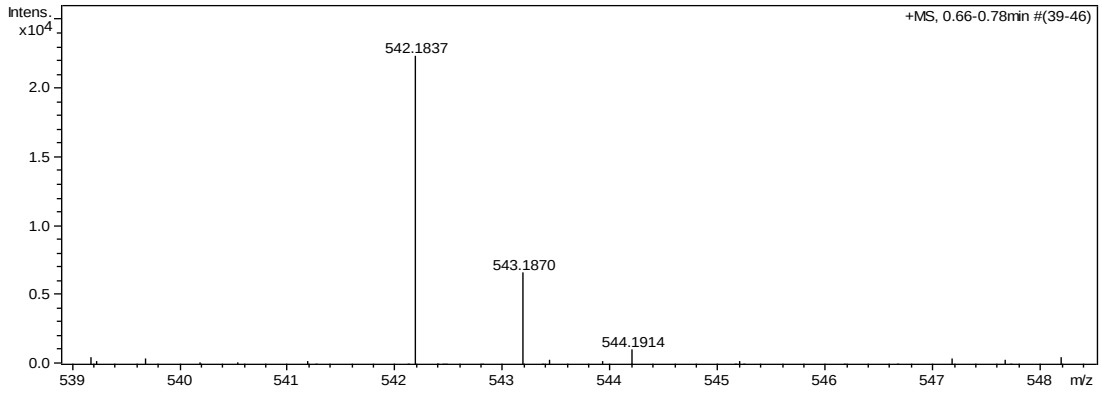

Gradient Shimming
168.46
167.95
147.14
143.11
142.63
141.43
134.91
133.12
132.80
131.91
130.06
130.02
129.28
129.17
128.95
128.87
128.83
128.63
128.38
128.27
127.71
127.63
127.50
127.00
126.97
126.34
125.74
123.63
123.02
122.07
121.98
121.35
119.72
111.39
110.21
106.90
56.24
34.19
O
N
O
CH3
N
N
H
90 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


Intens.
x10^4
+MS, 0.34-0.37min #(20-22)
604.1987
605.2021
601.1919
606.2054
610.1913
598.2099
612.1916
2.0
1.5
1.0
0.5
0.0
598
600
602
604
606
608
610
612
m/z

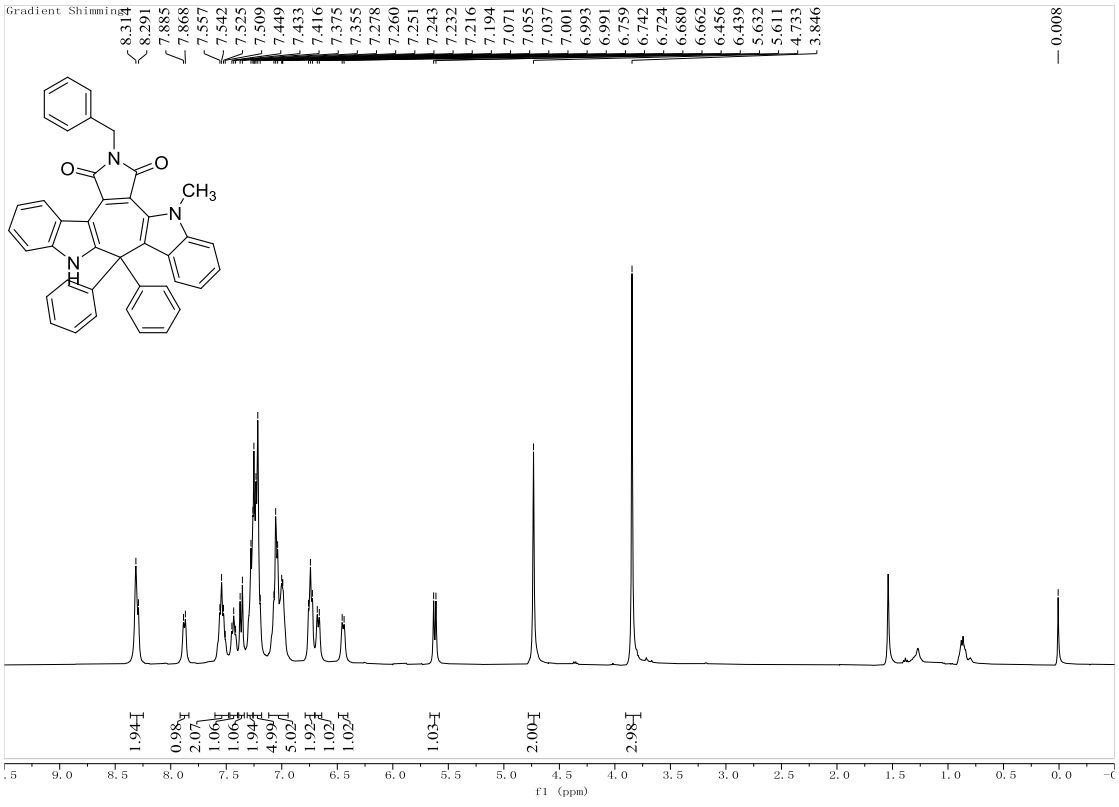
2,14-dimethyl-9,9-diphenyl-9,14-dihydropyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole-1,3(2*H*,8*H*)-dione (3b): petroleum ether and ethyl acetate (V/V = 10:1) as the eluent, red solid, 453 mg, red solid, 78%, m.p. 244-246 ºC; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.34 (d, J = 6.8 Hz, 1H, ArH), 8.24 (s, 1H, NH), 7.82 (d, J = 6.8 Hz, 1H, ArH), 7.55-7.49 (m, 2H, ArH), 7.44-7.41 (m, 1H, ArH), 7.37 (d, J = 8.0 Hz, 1H, ArH), 7.30-7.27 (m, 2H, ArH), 7.23-7.19 (m, 2H, ArH), 7.09-7.03 (m, 3H, ArH), 6.79-6.71 (m, 2H, ArH), 6.62 (d, J = 6.8 Hz, 1H, ArH), 6.47 (d, J = 6.4 Hz, 1H, ArH), 5.61 (d, J = 8.4 Hz, 1H, ArH), 3.85 (s, 3H, CH_3), 3.05 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 169.7, 169.3, 146.2, 143.2, 142.9, 141.4, 134.8, 133.1, 132.9, 129.8, 129.4, 129.3, 129.1, 128.6, 128.5, 128.4, 128.3, 127.7, 127.5, 127.0, 126.8, 126.7, 126.1, 125.6, 123.6, 123.5, 122.8, 122.0, 121.8, 121.5, 119.6, 111.3, 110.1, 106.9, 56.2, 34.1, 24.0; IR (KBr) υ: 3052, 2924, 2862, 2366, 1881, 1770, 1712, 1601, 1542, 1450, 1371, 1244, 1188, 998, 820, 734 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{35}H_{25}NaN_3O_2$ ([M+Na]$^{+}$): 542.1839, Found: 542.1837.

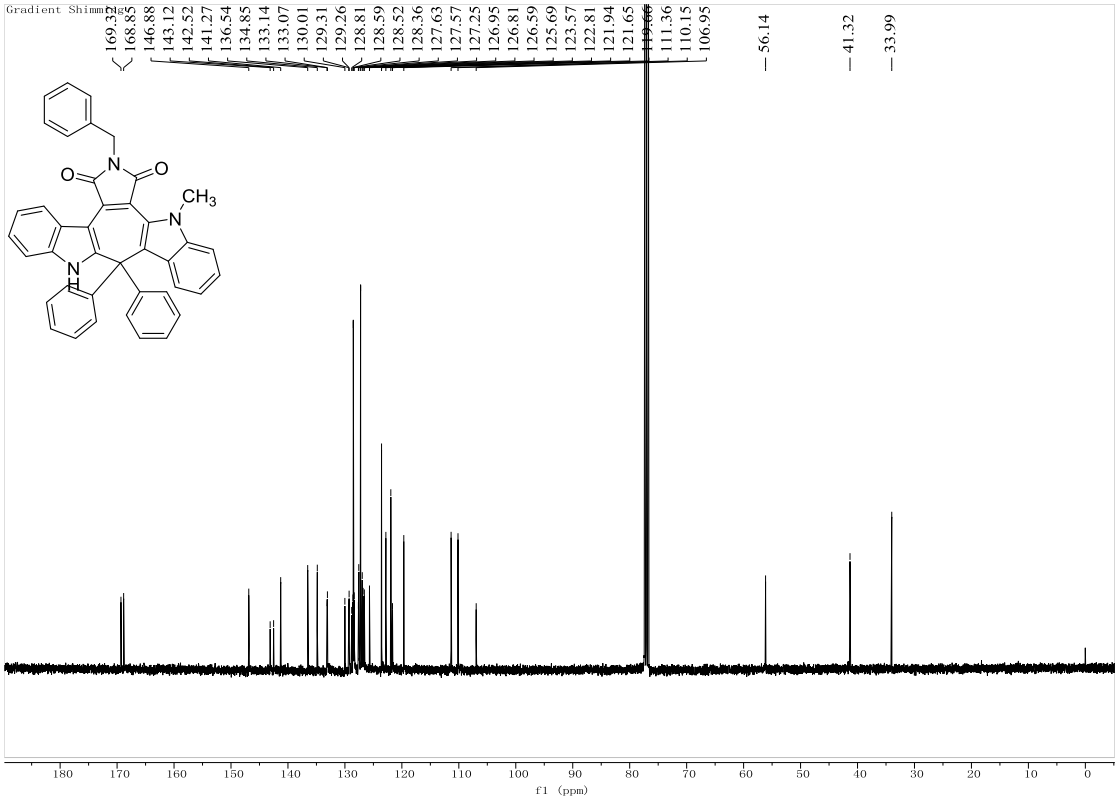


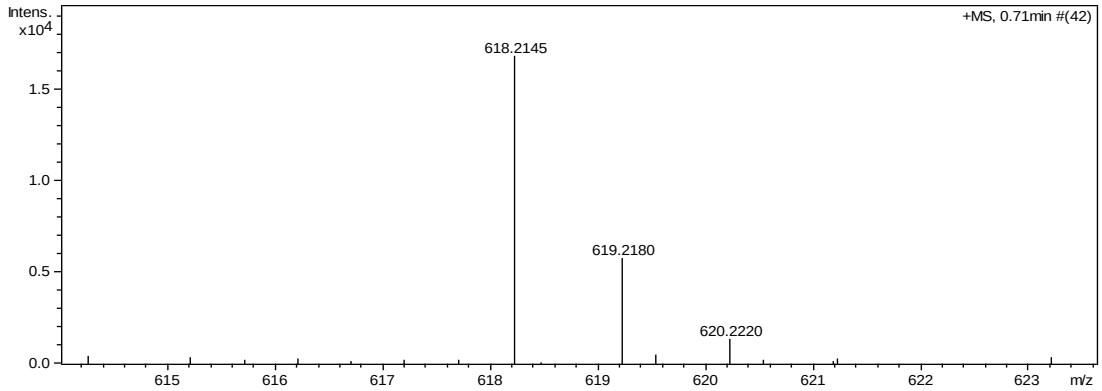

Gradient Shimming
169.75
169.34
146.31
143.26
142.95
141.42
134.87
133.20
132.94
129.90
129.36
129.33
129.16
128.71
128.54
128.41
128.34
127.76
127.52
127.10
126.91
126.89
126.18
125.71
123.56
123.53
122.90
122.04
121.89
121.53
119.65
111.33
110.17
106.96
56.22
34.14
24.04
CH3
N
O
O
CH3
N
N
H
90 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


Intens.
x10^4
+MS, 0.66-0.78min #(39-46)
542.1837
543.1870
544.1914
2.0
1.5
1.0
0.5
0.0
539 540 541 542 543 544 545 546 547 548 m/z

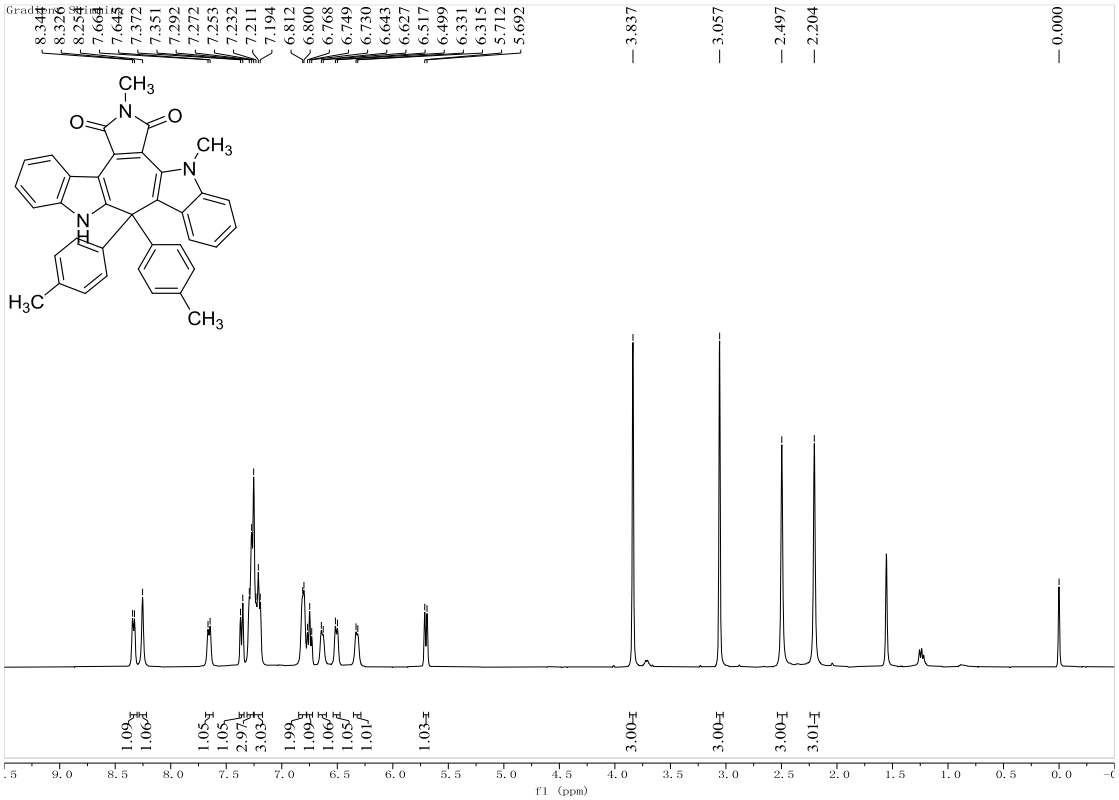
2-benzyl-14-methyl-9,9-diphenyl-9,14-dihydropyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole-1,3(2*H*,8*H*)-dione (3c): petroleum ether and ethyl acetate (V/V = 10:1) as the eluent, red solid, 464 mg, red solid, 78%, m.p. 212-215 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.31-8.29 (m, 2H, ArH), 7.88 (d, *J* = 6.8 Hz, 1H, ArH), 7.56-7.51 (m, 2H, ArH), 7.43 (t, *J* = 6.8 Hz, 3H, ArH), 7.37 (d, *J* = 8.0 Hz, 1H, ArH), 7.28-7.19 (m, 7H, ArH), 7.07-6.99 (m, 5H, ArH), 6.74 (t, *J* = 7.2 Hz, 2H, ArH), 6.67 (d, *J* = 7.2 Hz, 1H, ArH), 6.45 (d, *J* = 6.8 Hz, 1H, ArH), 5.62 (d, *J* = 8.4 Hz, 1H, ArH), 4.73 (s, 2H, CH), 3.85 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 169.3, 168.8, 146.8, 143.1, 142.5, 141.2, 136.5, 134.8, 133.1, 133.0, 129.9, 129.3, 129.2, 128.7, 128.5, 128.5, 128.3, 127.6, 127.5, 127.2, 126.9, 126.7, 126.5, 125.6, 123.5, 122.7, 121.9, 121.6, 119.6, 111.3, 110.1, 106.9, 56.1, 41.3, 33.9; IR (KBr) υ: 3032, 2956, 2878, 2334, 1876, 1765, 1730, 1589, 1521, 1476, 1325, 1230, 1180, 920, 856, 774 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{41}H_{29}NaN_3O_2$ ([M+Na]$^{+}$): 618.2152, Found: 618.2145.

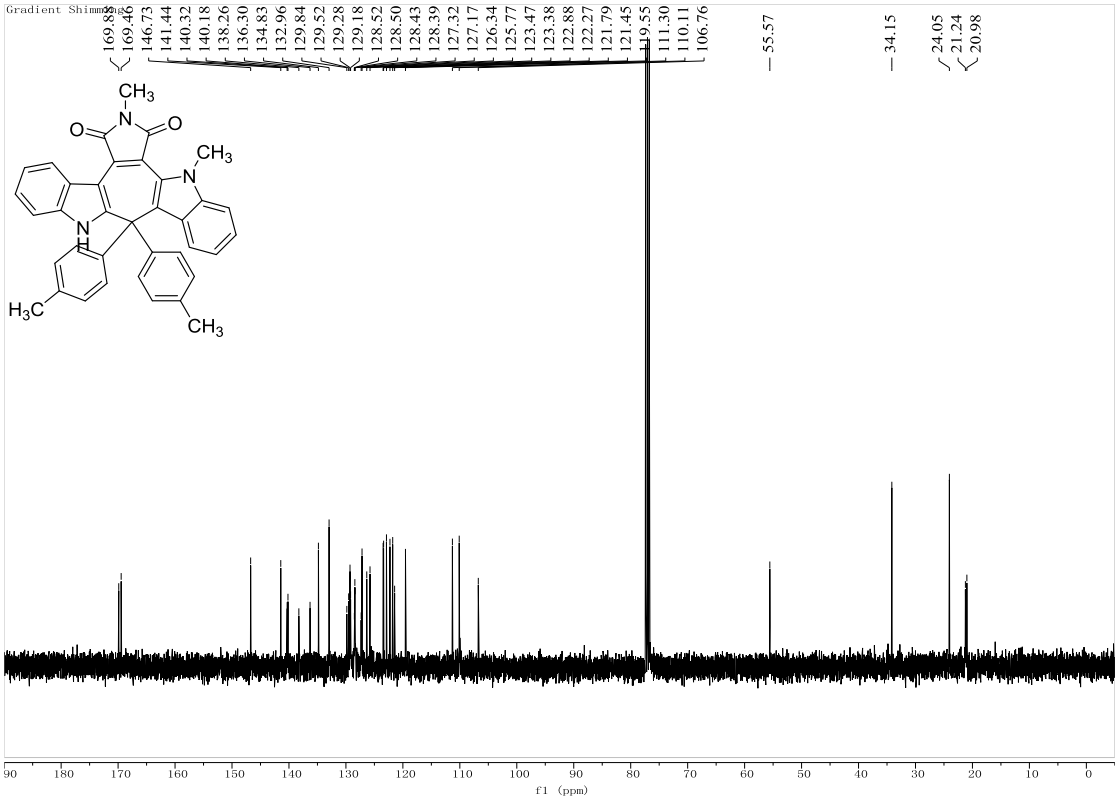


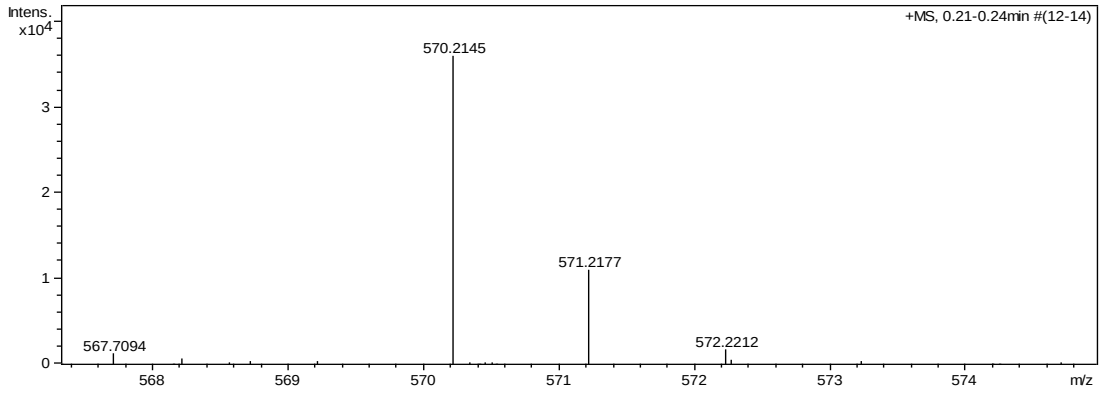

Gradient Shimming
169.32
168.85
146.88
143.12
142.52
141.27
136.54
134.85
133.14
133.07
130.01
129.31
129.26
128.81
128.59
128.52
128.36
127.63
127.57
127.25
126.95
126.81
126.59
125.69
123.57
122.81
121.94
121.65
111.36
110.15
106.95
56.14
41.32
33.99
O
N
O
CH3
N
N
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Intens.
x10^4
+MS, 0.71min #(42)
618.2145
619.2180
620.2220
1.5
1.0
0.5
0.0
615
616
617
618
619
620
621
622
623
m/z

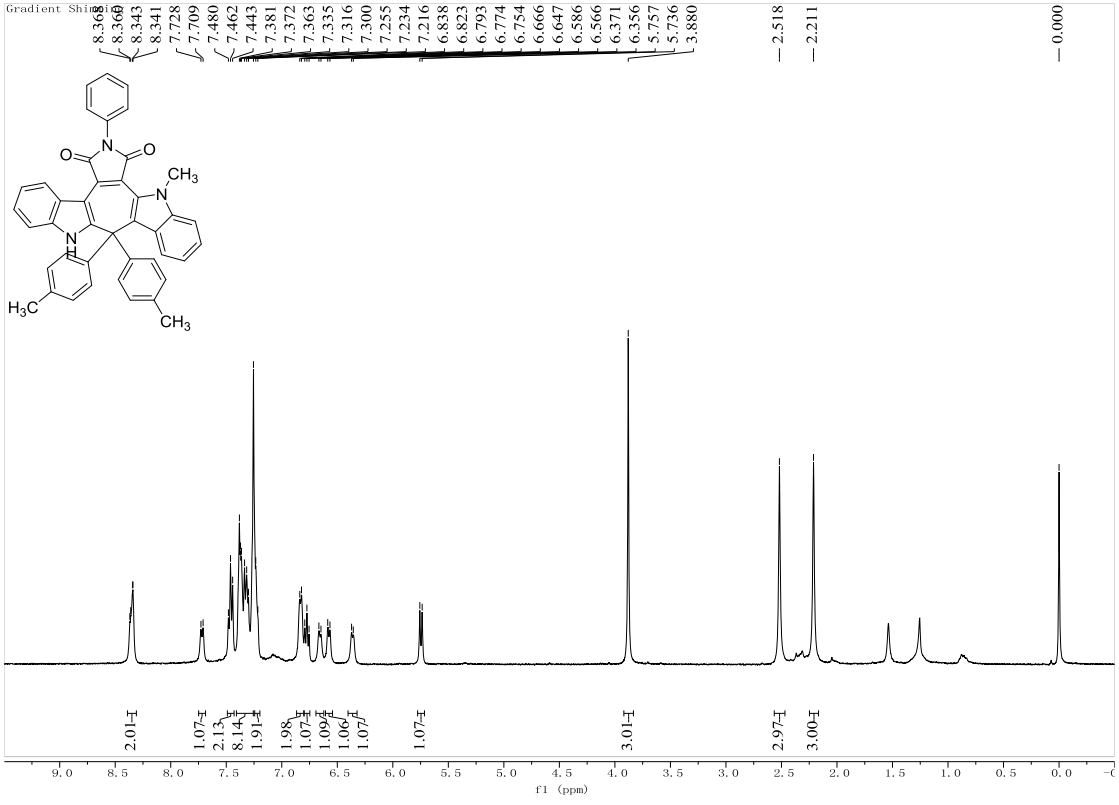
2,14-dimethyl-9,9-di-p-tolyl-9,14-dihydropyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole-1,3(2*H*,8*H*)-dione (3d): petroleum ether and ethyl acetate (V/V = 10:1) as the eluent, red solid, 405 mg, red solid, 74%, m.p. 233-236 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.33 (d, *J* = 6.8 Hz, 1H, ArH), 8.25 (s, 1H, NH), 7.65 (d, *J* = 7.6 Hz, 1H, ArH), 7.36 (d, *J* = 8.4 Hz, 1H, ArH), 7.29-7.19 (m, 6H, ArH), 6.81 (d, *J* = 4.8 Hz, 2H, ArH), 6.75 (t, *J* = 7.6 Hz, 1H, ArH), 6.64 (d, *J* = 6.4 Hz, 1H, ArH), 6.32 (d, *J* = 6.4 Hz, 1H, ArH), 5.70 (d, *J* = 8.0 Hz, 1H, ArH), 3.84 (s, 3H, CH_3), 3.06 (s, 3H, CH_3), 2.50 (s, 3H, CH_3), 2.20 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 169.8, 169.4, 146.7, 141.4, 140.3, 140.1, 138.2, 136.2, 134.8, 132.9, 129.8, 129.5, 129.2, 129.1, 128.5, 128.4, 128.4, 128.3, 127.3, 127.1, 126.3, 125.7, 123.4, 123.3, 122.8, 122.2, 121.7, 121.4, 119.5, 111.2, 110.0, 106.7, 55.5, 34.1, 24.0, 21.2, 20.9; IR (KBr) υ: 3078, 2934, 2852, 2735, 2545, 2356, 1887, 1775, 1721, 1632, 1534, 1445, 1356, 1265, 1156, 912, 820, 721 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{37}H_{29}NaN_3O_2$ ([M+Na]$^{+}$): 570.2152, Found: 570.2145.

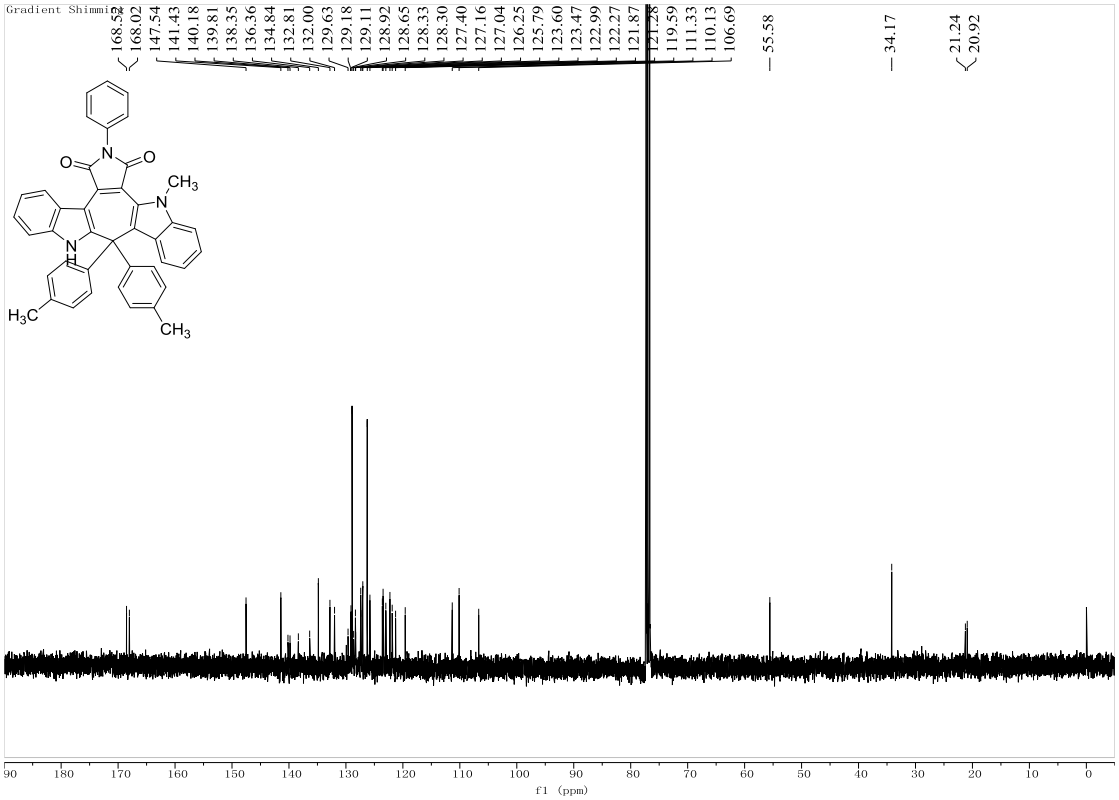


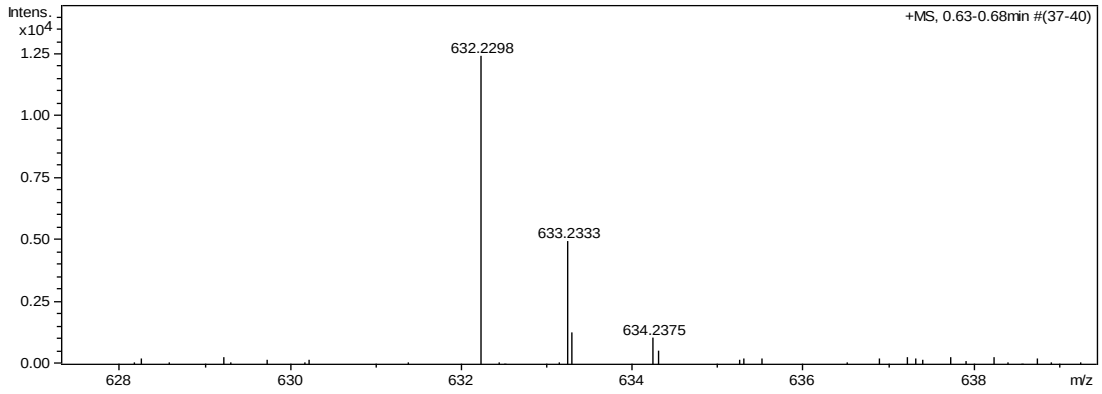

Gradient Shimming
169.88
169.46
146.73
141.44
140.32
140.18
138.26
136.30
134.83
132.96
129.84
129.52
129.28
129.18
128.52
128.50
128.43
128.39
127.32
127.17
126.34
125.77
123.47
123.38
122.88
122.27
121.79
121.45
119.55
111.30
110.11
106.76
55.57
34.15
24.05
21.24
20.98
CH3
O
N
O
CH3
N
N
H3C
CH3
90
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Intens.
x10^4
+MS, 0.21-0.24min #(12-14)
570.2145
571.2177
567.7094
572.2212
0
1
2
3
568
569
570
571
572
573
574
m/z

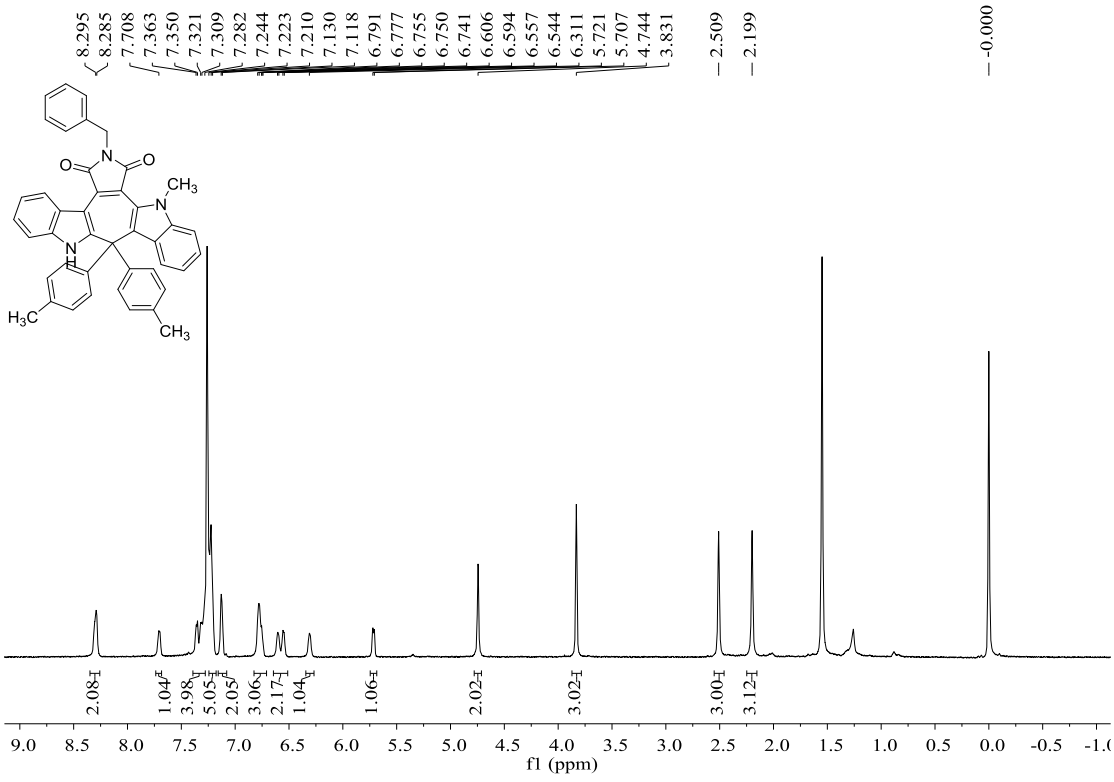
14-methyl-2-phenyl-9,9-di-p-tolyl-9,14-dihydropyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole-1,3(2*H*,8*H*)-dione (3e): petroleum ether and ethyl acetate (V/V = 10:1) as the eluent, red solid, 439 mg, red solid, 72%, m.p. 208-211 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.37-8.34 (m, 2H, ArH), 7.72 (d, *J* = 7.6 Hz, 1H, ArH), 7.46 (t, *J* = 7.6 Hz, 2H, ArH), 7.38-7.22 (m, 10H, ArH), 6.83 (d, *J* = 6.0 Hz, 2H, ArH), 6.77 (t, *J* = 8.0 Hz, 1H, ArH), 6.66 (d, *J* = 7.6 Hz, 1H, ArH), 6.58 (d, *J* = 8.0 Hz, 1H, ArH), 6.36 (d, *J* = 6.0 Hz, 1H, ArH), 5.75 (d, *J* = 8.4 Hz, 1H, ArH), 3.88 (s, 3H, CH_3), 2.52 (s, 3H, CH_3), 2.21 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 168.4, 167.9, 147.5, 141.4, 140.1, 139.7, 138.3, 136.3, 134.8, 132.7, 131.9, 129.6, 129.1, 129.0, 128.9, 128.6, 128.3, 128.2, 127.3, 127.1, 127.0, 126.2, 125.7, 123.5, 123.4, 122.9, 122.2, 121.8, 121.2, 119.5, 111.3, 110.1, 106.6, 55.5, 34.1, 21.2, 20.9; IR (KBr) υ: 3034, 2938, 2867, 2345, 1876, 1764, 1711, 1631, 1538, 1475, 1453, 1376, 1238, 1178, 911, 852, 747 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{42}H_{31}NaN_3O_2$ ([M+Na]$^{+}$): 632.2308, Found: 632.2298.

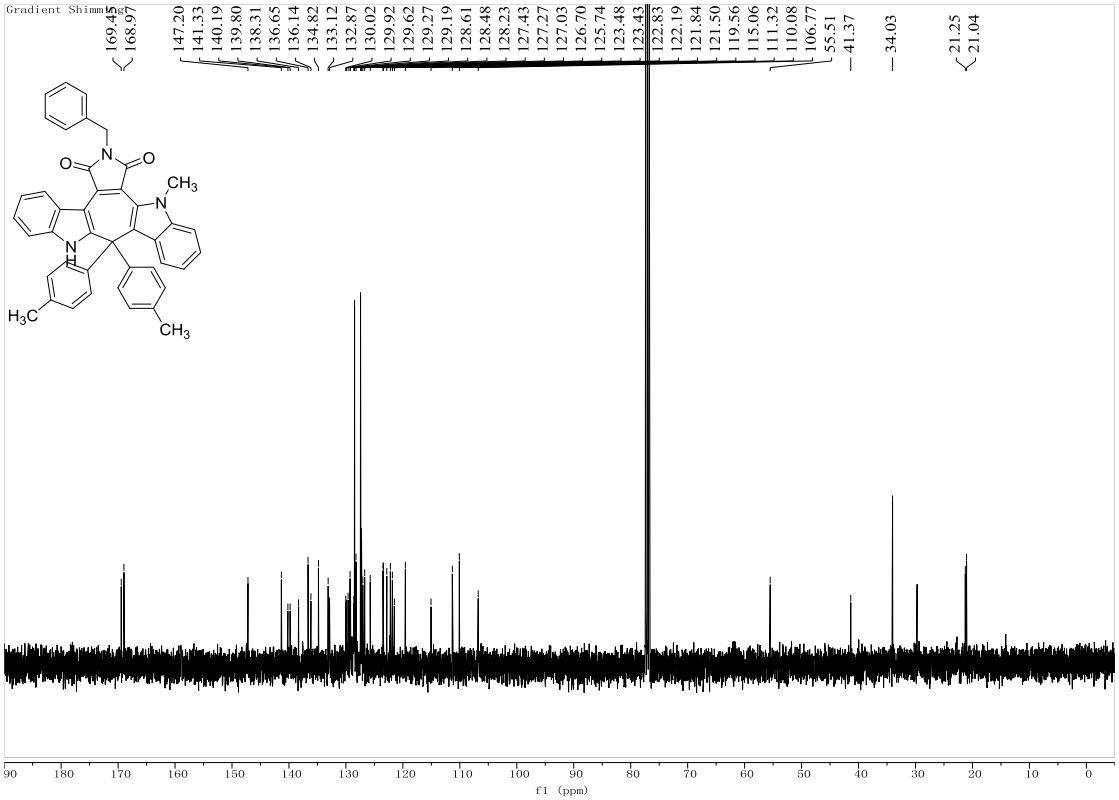


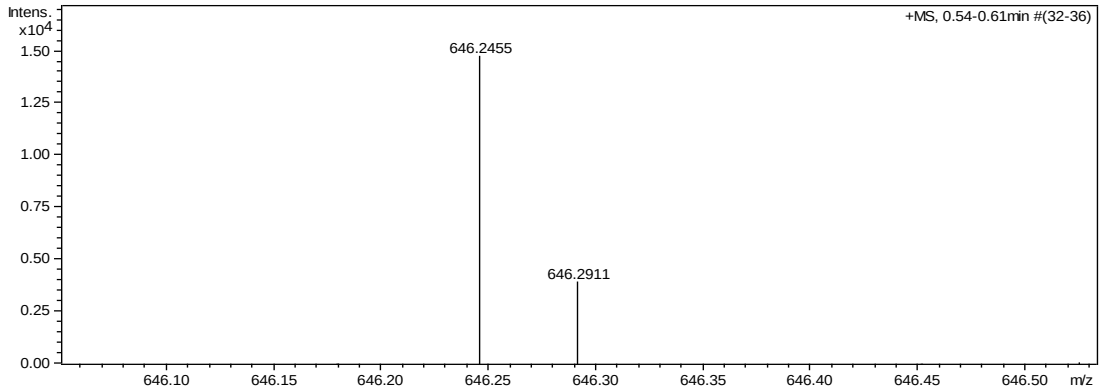

Gradient Shimming
168.52
168.02
147.54
141.43
140.18
139.81
138.35
136.36
134.84
132.81
132.00
129.63
129.18
129.11
128.92
128.65
128.33
128.30
127.40
127.16
127.04
126.25
125.79
123.60
123.47
122.99
122.27
121.87
121.28
119.59
111.33
110.13
106.69
55.58
34.17
21.24
20.92
O
O
N
CH3
N
H
H3C
CH3
90
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Intens.
x10^4
1.25
1.00
0.75
0.50
0.25
0.00
+MS, 0.63-0.68min #(37-40)
632.2298
633.2333
634.2375
628
630
632
634
636
638
m/z

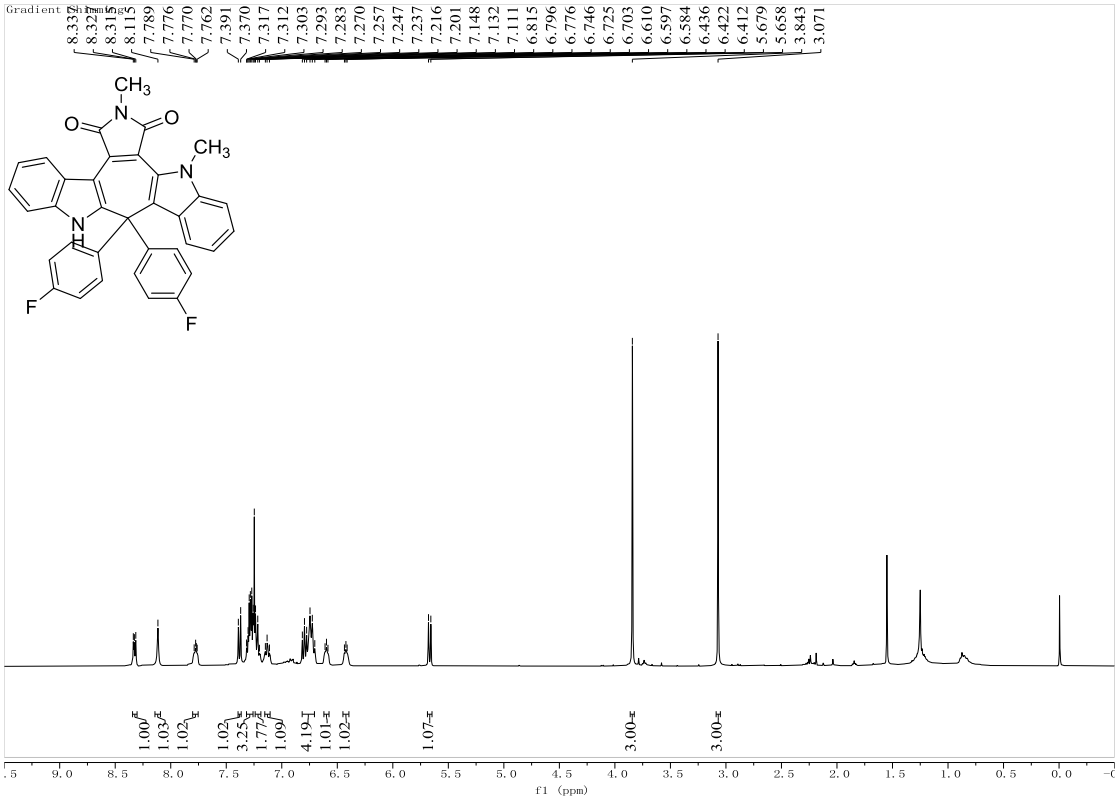
2-benzyl-14-methyl-9,9-di-p-tolyl-9,14-dihydropyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole-1,3(2*H*,8*H*)-dione (3f): petroleum ether and ethyl acetate (V/V = 10:1) as the eluent, red solid, 449 mg, red solid, 72%, m.p. 252-255 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ: 8.30-8.29 (m, 2H, ArH), 7.71 (s, 1H, ArH), 7.36-7.28 (m, 4H, ArH), 7.24-7.21 (m, 5H, ArH), 7.13-7.12 (m, 2H, ArH), 6.79-6.74 (m, 3H, ArH), 6.61-6.54 (m, 2H, ArH), 6.31 (s, 1H, ArH), 5.71 (d, *J* = 8.4 Hz, 1H, ArH), 4.74 (s, 2H, CH), 3.83 (s, 3H, CH_3), 2.51 (s, 3H, CH_3); 2.20 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 169.4, 168.9, 147.1, 141.3, 140.1, 139.7, 138.2, 136.6, 136.1, 134.8, 133.1, 132.8, 130.0, 129.9, 129.6, 129.2, 129.1, 128.5, 128.4, 128.2, 127.4, 127.2, 127.0, 126.6, 125.7, 123.5, 123.4, 122.8, 122.1, 121.8, 121.4, 119.5, 115.0, 111.3, 110.0, 106.7, 55.5, 41.3, 34.0, 21.2, 21.0; IR (KBr) υ: 3021, 2933, 2854, 2357, 1880, 1776, 1605, 1532, 1455, 1447, 1356, 1276, 1165, 956, 837, 754 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{43}H_{33}NaN_3O_2$ ([M+Na]$^{+}$): 646.2465, Found: 646.2455.

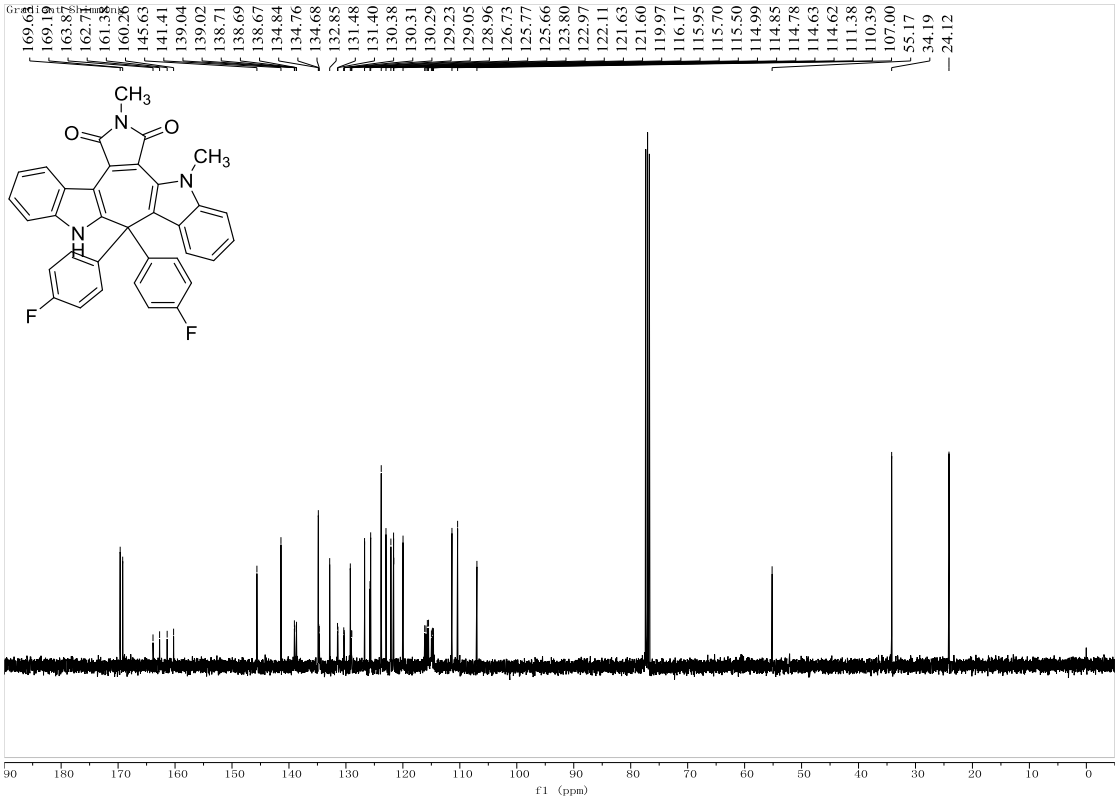


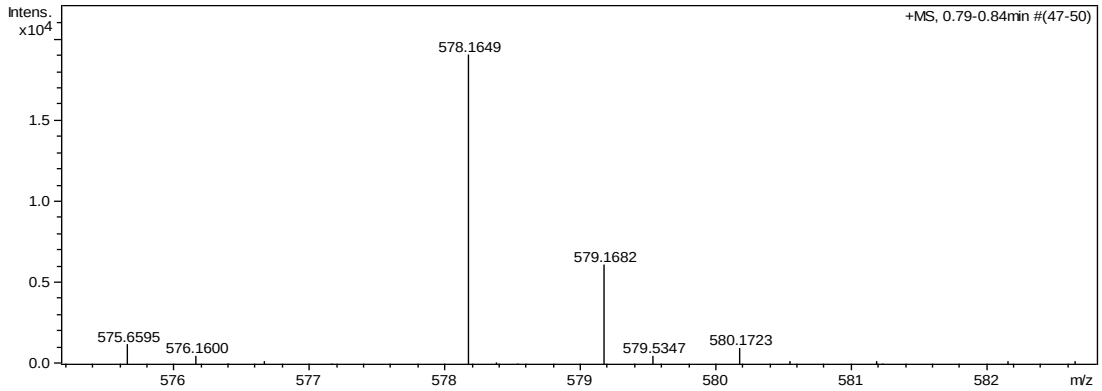

Gradient Shimmol.gf
169.45
168.97
147.20
141.33
140.19
139.80
138.31
136.65
136.14
134.82
133.12
132.87
130.02
129.92
129.62
129.27
129.19
128.61
128.48
128.23
127.43
127.27
127.03
126.70
125.74
123.48
123.43
122.83
122.19
121.84
121.50
119.56
115.06
111.32
110.08
106.77
55.51
41.37
34.03
21.25
21.04
O
O
N
CH3
N
H
H3C
CH3
90 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


Intens.
x10^4
1.50
1.25
1.00
0.75
0.50
0.25
0.00
+MS, 0.54-0.61min #(32-36)
646.2455
646.2911
646.10 646.15 646.20 646.25 646.30 646.35 646.40 646.45 646.50 m/z

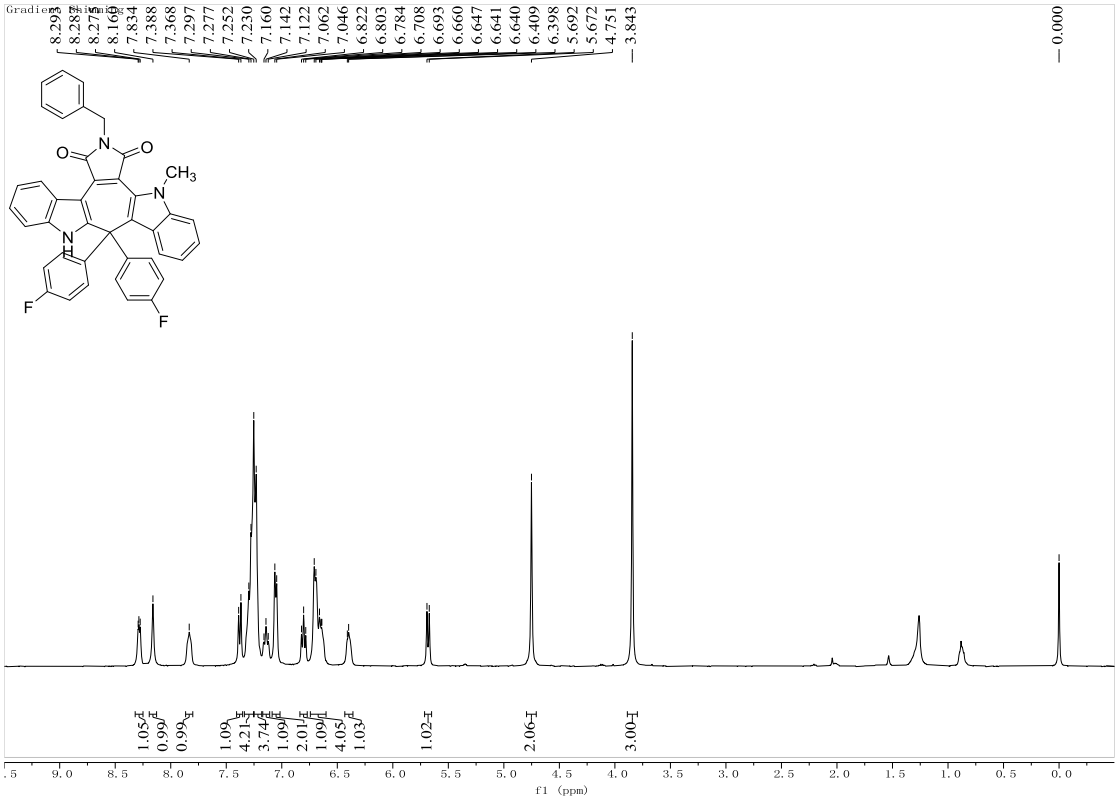
9,9-bis(4-fluorophenyl)-2,14-dimethyl-9,14-dihydropyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole-1,3(2*H*,8*H*)-dione (3g): petroleum ether and ethyl acetate (V/V = 10:1) as the eluent, red solid, 472 mg, red solid, 85%, m.p. 236-238 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.34-8.32 (m, 1H, ArH), 8.12 (s, 1H, NH), 7.79-7.76 (m, 1H, ArH), 7.38 (d, *J* = 8.8 Hz, 1H, ArH), 7.32-7.20 (m, 5H, ArH), 7.15-7.11 (m, 1H, ArH), 6.82-6.70 (m, 4H, ArH), 6.61-6.58 (m, 1H, ArH), 6.44-6.41 (m, 1H, ArH), 5.67 (d, *J* = 8.4 Hz, 1H, ArH), 3.84 (s, 3H, CH_3), 3.07 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 169.6, 169.2, 162.6 (d, J = 248.5 Hz), 161.5 (d, J = 245.7 Hz), 145.6, 141.4, 139.1, 139.0, 138.7, 138.6, 134.8, 134.7, 134.7, 132.8, 131.5, 131.4, 130.3, 130.2, 129.2, 129.0, 128.9, 126.7, 125.8, 125.6, 123.8, 122.9, 122.1, 121.6, 121.5, 120.0, 116.0 (d, J = 22.2 Hz), 115.6 (d, J = 20.3 Hz), 114.9 (d, J = 21.0 Hz), 114.7 (d, J = 21.4 Hz), 111.3, 110.3, 107.0, 55.2, 34.2, 24.1; IR (KBr) υ: 3024, 2936, 2863, 2356, 1877, 1722, 1601, 1533, 1479, 1445, 1368, 1232, 1156, 1125, 833, 737 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{35}H_{23}F_2NaN_3O_2$ ([M+Na]$^{+}$): 578.1651, Found: 578.1469.

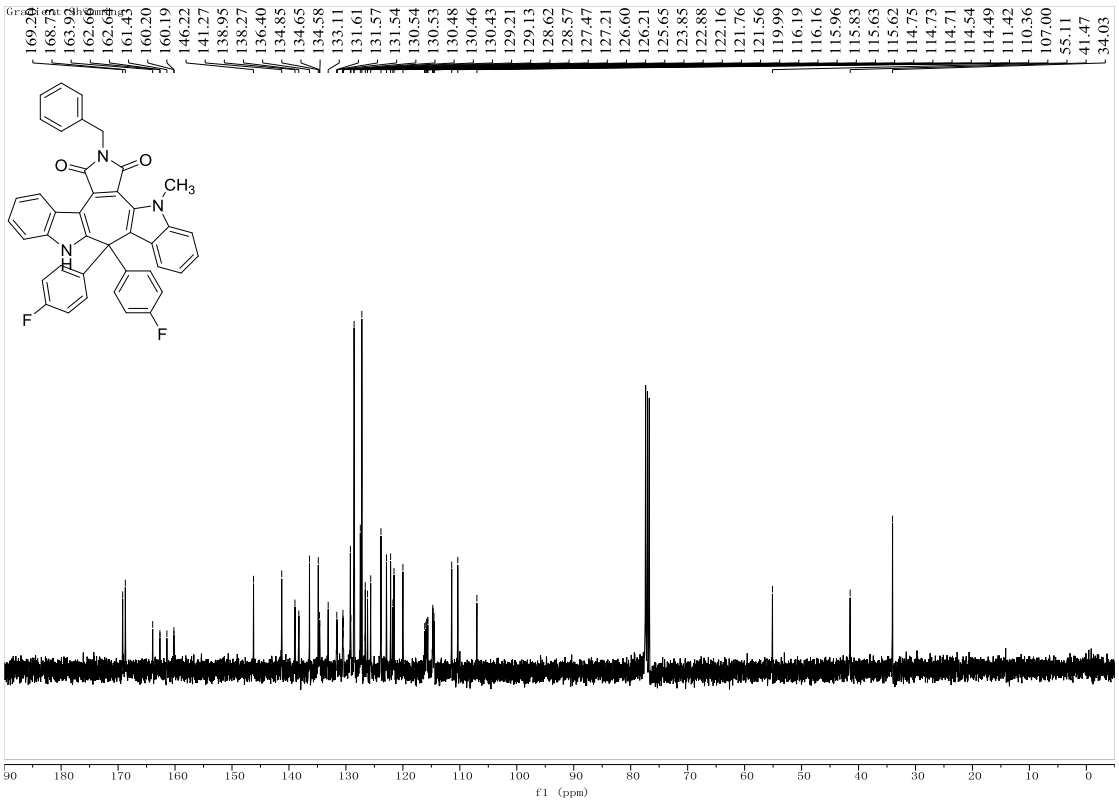


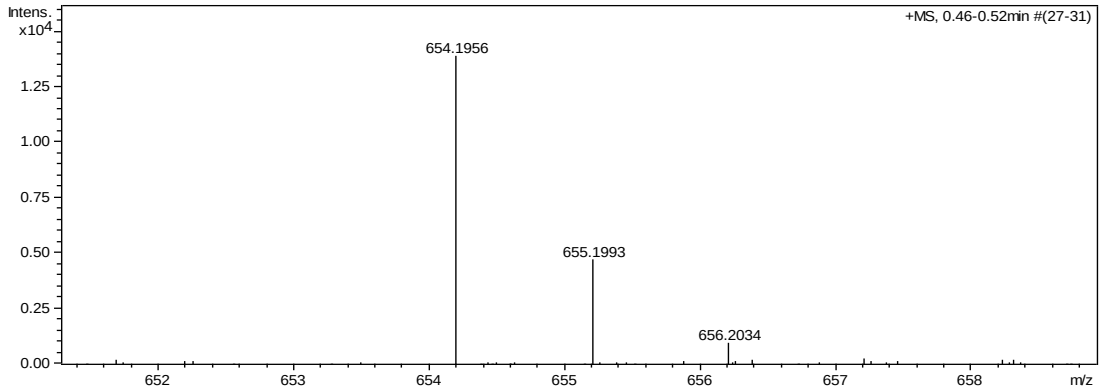

169.66
169.19
163.87
162.77
161.35
160.26
145.63
141.41
139.04
139.02
138.71
138.69
138.67
134.84
134.76
134.68
132.85
131.48
131.40
130.38
130.31
130.29
129.23
129.05
128.96
126.73
125.77
125.66
123.80
122.97
122.11
121.63
121.60
119.97
116.17
115.95
115.70
115.50
114.99
114.85
114.78
114.63
114.62
111.38
110.39
107.00
55.17
34.19
24.12
CH3
O
N
O
CH3
N
N
H
F
F
90
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Intens.
x10^4
+MS, 0.79-0.84min #(47-50)
578.1649
579.1682
575.6595
576.1600
579.5347
580.1723
1.5
1.0
0.5
0.0
576
577
578
579
580
581
582
m/z

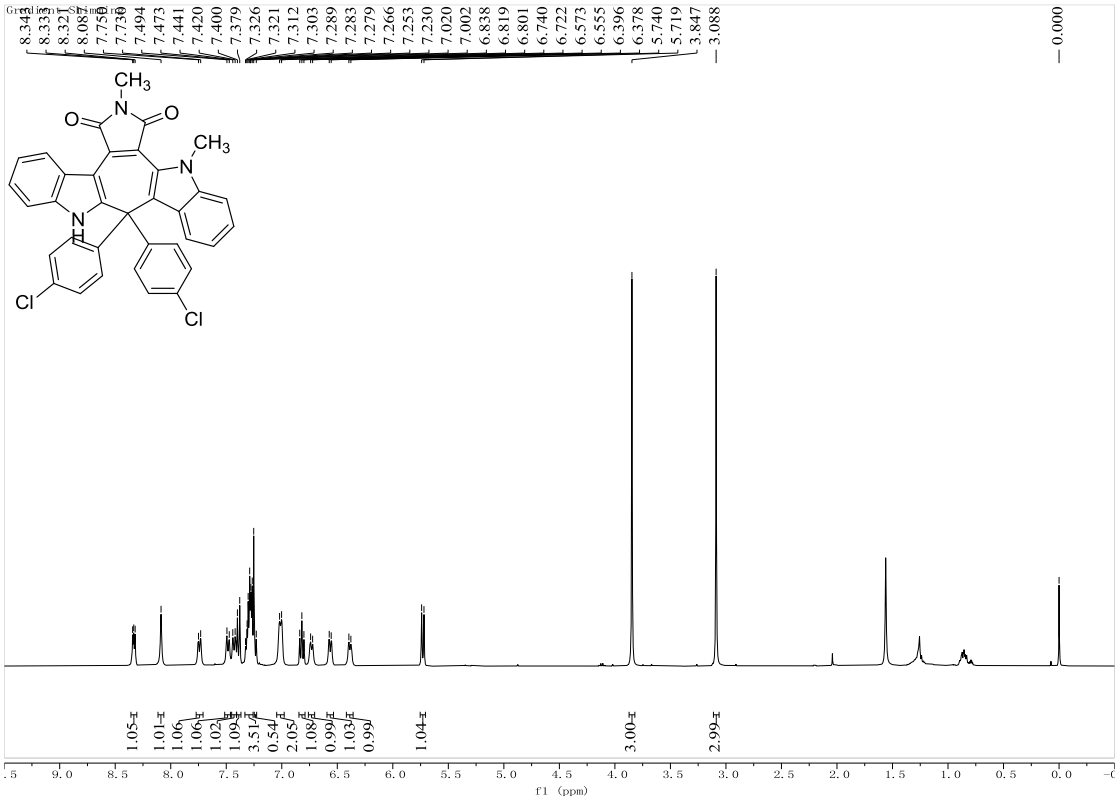
2-benzyl-9,9-bis(4-fluorophenyl)-14-methyl-9,14-dihydropyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole-1,3(2*H*,8*H*)-dione (3h): petroleum ether and ethyl acetate (V/V = 10:1) as the eluent, red solid, 543 mg, red solid, 86%, m.p. 222-224 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.29-8.28 (m, 1H, ArH), 8.16 (s, 1H, NH), 7.83 (s, 1H, ArH), 7.38 (d, *J* = 8.0 Hz, 1H, ArH), 7.30-7.23 (m, 8H, ArH), 7.16-7.12 (m, 1H, ArH), 7.05 (d, *J* = 6.4 Hz, 2H, ArH), 6.80 (d, *J* = 7.6 Hz, 1H, ArH), 6.71-6.64 (m, 4H, ArH), 6.41-6.40 (m, 1H, ArH), 5.68 (d, *J* = 8.0 Hz, 1H, ArH), 4.75 (s, 2H, CH), 3.84 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 169.1, 168.7, 162.7 (d, *J* = 249.3 Hz), 161.4 (d, *J* = 245.9 Hz), 146.2, 141.2, 138.9, 138.2, 136.3, 134.8, 134.6, 134.5, 133.0, 131.5, 131.5, 130.5, 130.5, 130.4, 130.4, 129.1, 129.1, 128.5, 127.4, 127.1, 126.6, 126.5, 126.1, 125.6, 123.8, 122.8, 122.1, 121.7, 121.5, 119.9, 116.1 (d, *J* = 20.2 Hz), 115.7 (d, *J* = 21.4 Hz), 114.6 (d, *J* = 21.4 Hz), 114.6 (d, *J* = 21.4 Hz), 110.3, 106.9, 55.1, 41.4, 34.0; IR (KBr) υ: 3045, 2935, 2866, 1879, 1770, 1714, 1615, 1546, 1465, 1445, 1367, 1244, 1176, 820, 734 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{41}H_{27}F_2NaN_3O_2$ ([M+Na]$^+$): 654.1964, Found: 654.1956.

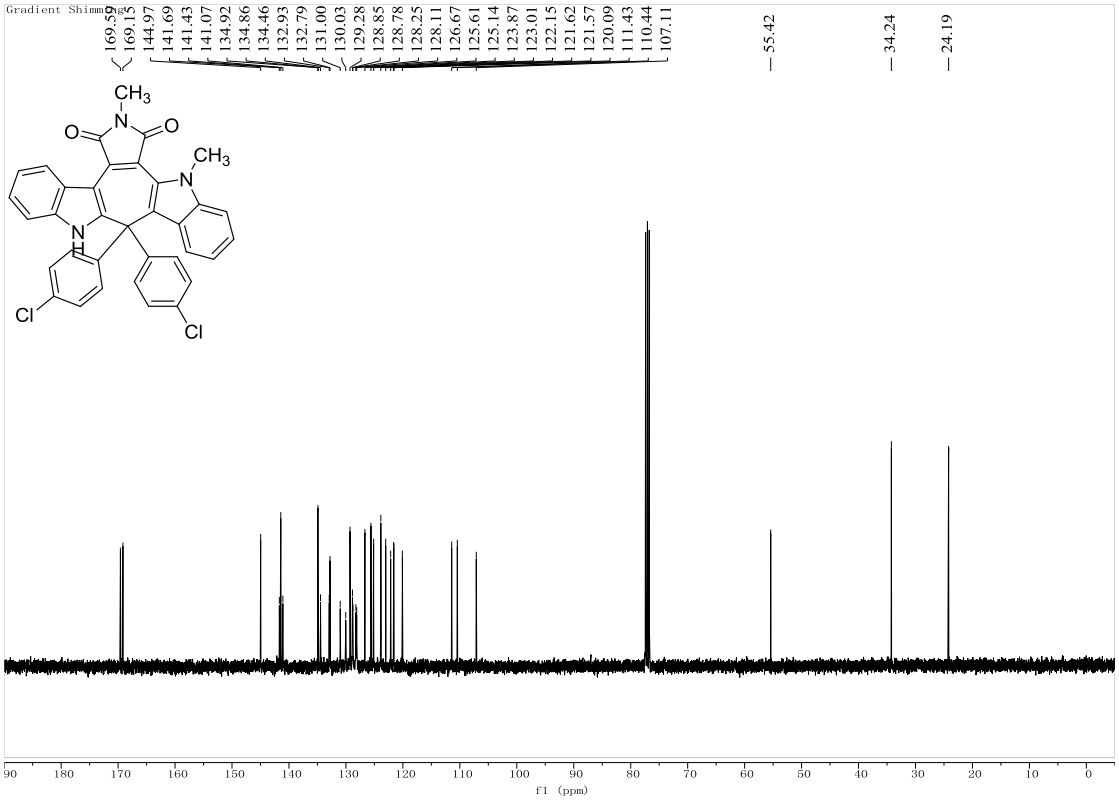


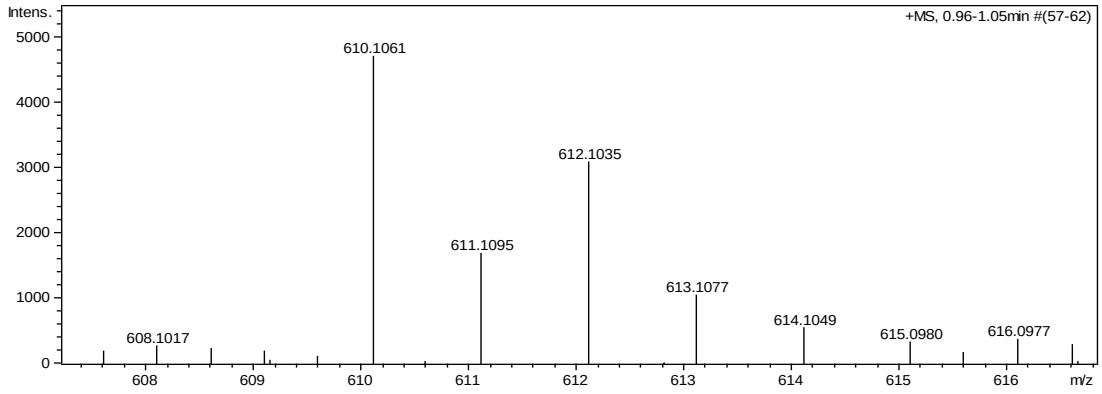

169.26
168.73
163.92
162.60
162.66
161.43
160.20
160.19
146.22
141.27
138.95
138.27
136.40
134.85
134.65
134.58
133.11
131.61
131.57
131.54
130.54
130.53
130.48
130.46
130.43
129.21
129.13
128.62
128.57
127.47
127.21
126.60
126.21
125.65
123.85
122.88
122.16
121.76
121.56
119.99
116.19
116.16
115.96
115.83
115.63
115.62
114.75
114.73
114.71
114.54
114.49
111.42
110.36
107.00
55.11
41.47
34.03
O
O
N
CH3
N
H
F
F
90
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Intens.
x10^4
+MS, 0.46-0.52min #(27-31)
654.1956
655.1993
656.2034
1.25
1.00
0.75
0.50
0.25
0.00
652
653
654
655
656
657
658
m/z

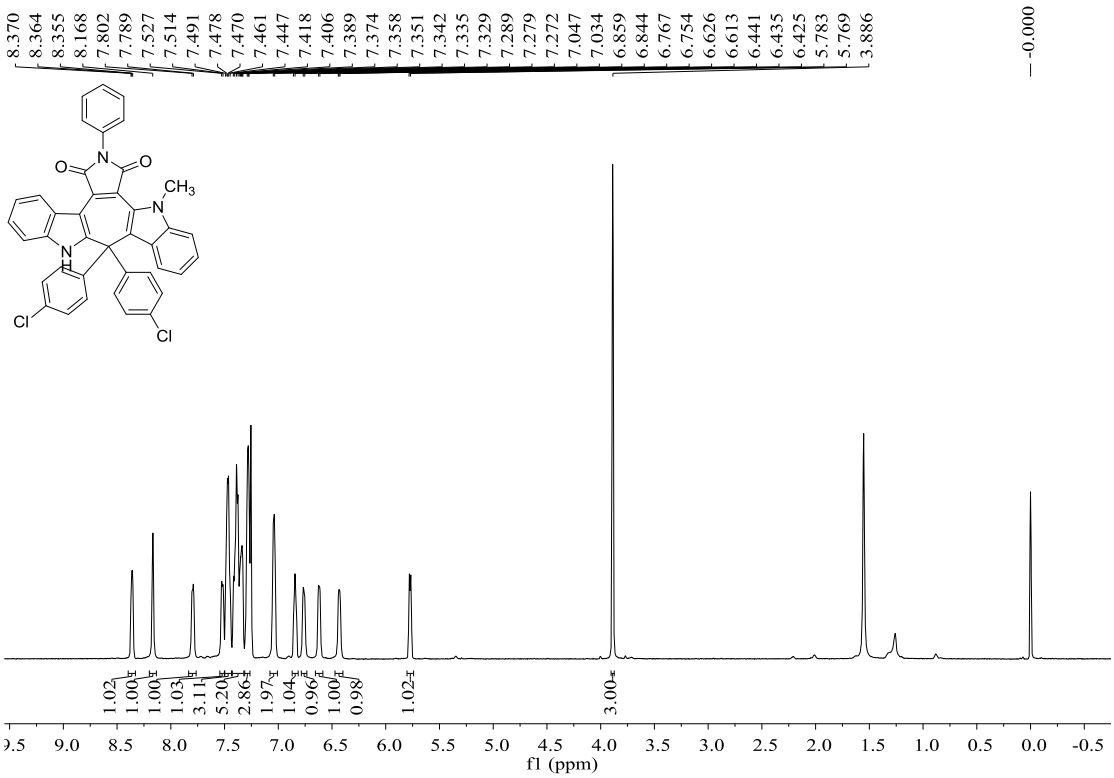
9,9-bis(4-chlorophenyl)-2,14-dimethyl-9,14-dihydropyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole-1,3(2*H*,8*H*)-dione (3i): petroleum ether and ethyl acetate (V/V = 10:1) as the eluent, red solid, 517 mg, red solid, 88%, m.p. 246-248 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.34-8.32 (m, 1H, ArH), 8.09 (s, 1H, NH), 7.74 (d, *J* = 8.0 Hz, 1H, ArH), 7.48 (d, *J* = 8.4 Hz, 1H, ArH), 7.43 (d, *J* = 7.6 Hz, 1H, ArH), 7.39 (d, *J* = 8.0 Hz, 1H, ArH), 7.33-7.23 (m, 4H, ArH), 7.01 (d, *J* = 7.2 Hz, 2H, ArH), 6.82 (t, *J* = 7.6 Hz, 1H, ArH), 6.73 (d, *J* = 7.2 Hz, 1H, ArH), 6.56 (d, *J* = 7.2 Hz, 1H, ArH), 6.39 (d, *J* = 7.2 Hz, 1H, ArH), 5.73 (d, *J* = 8.4 Hz, 1H, ArH), 3.85 (s, 3H, CH_3), 3.09 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 169.5, 169.1, 144.9, 141.6, 141.4, 141.0, 134.9, 134.8, 134.4, 132.9, 132.7, 130.9, 130.0, 129.2, 128.8, 128.7, 128.2, 128.0, 126.6, 125.5, 125.1, 123.8, 123.0, 122.1, 121.6, 121.5, 120.0, 111.4, 110.4, 107.1, 55.4, 34.2, 24.1; IR (KBr) υ: 3044, 2934, 2856, 2356, 1851, 1756, 1614, 1553, 1478, 1443, 1381, 1244, 1198, 1122, 833, 745 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{35}H_{23}Cl_2NaN_3O_2$ ([M+Na]$^{+}$): 610.1060, Found: 610.1061.

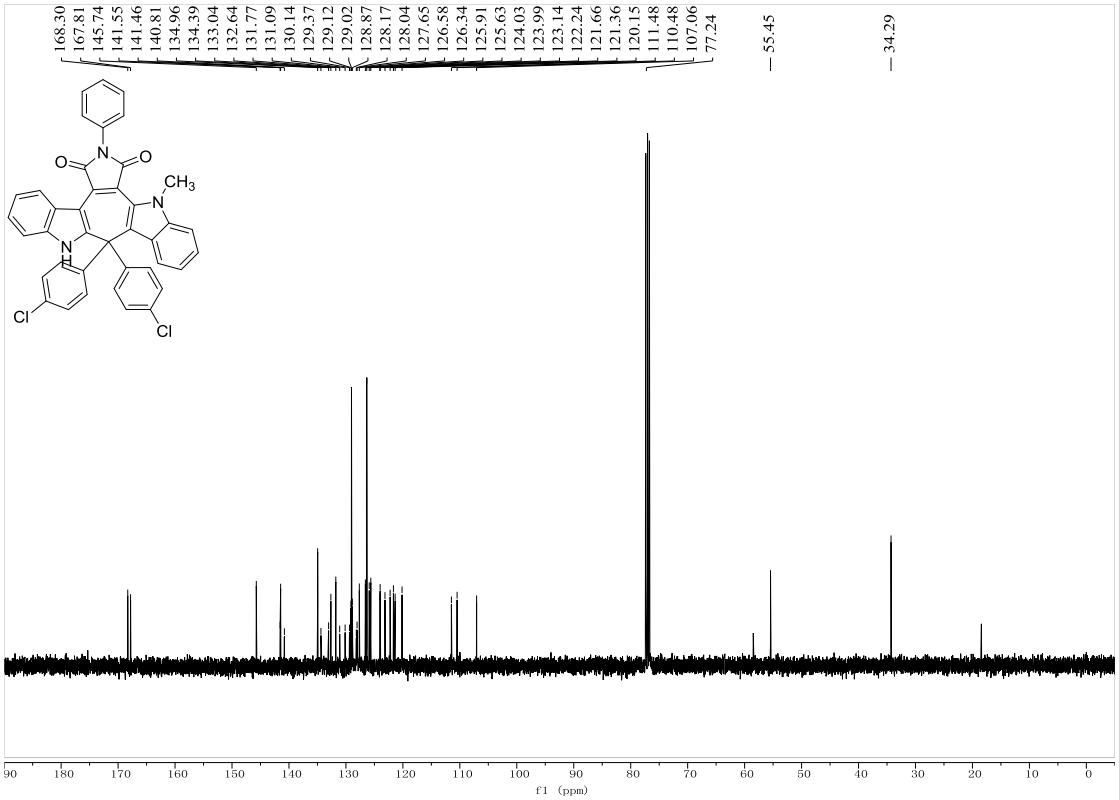


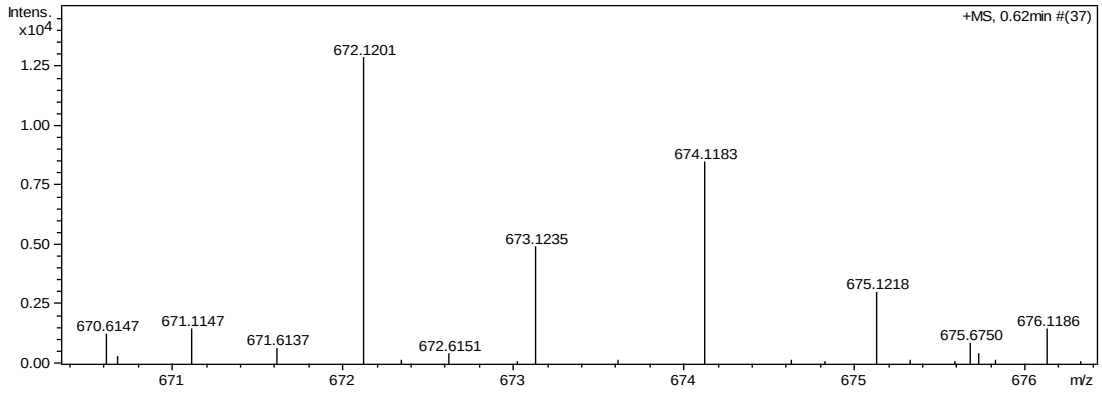

Gradient Shimming
169.59
169.15
144.97
141.69
141.43
141.07
134.92
134.86
134.46
132.93
132.79
131.00
130.03
129.28
128.85
128.78
128.25
128.11
126.67
125.61
125.14
123.87
123.01
122.15
121.62
121.57
120.09
111.43
110.44
107.11
55.42
34.24
24.19
CH3
N
O
O
CH3
N
N
H
Cl
Cl
90
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Intens.
+MS, 0.96-1.05min #(57-62)
5000
4000
3000
2000
1000
0
610.1061
612.1035
611.1095
613.1077
614.1049
608.1017
615.0980
616.0977
608
609
610
611
612
613
614
615
616
m/z

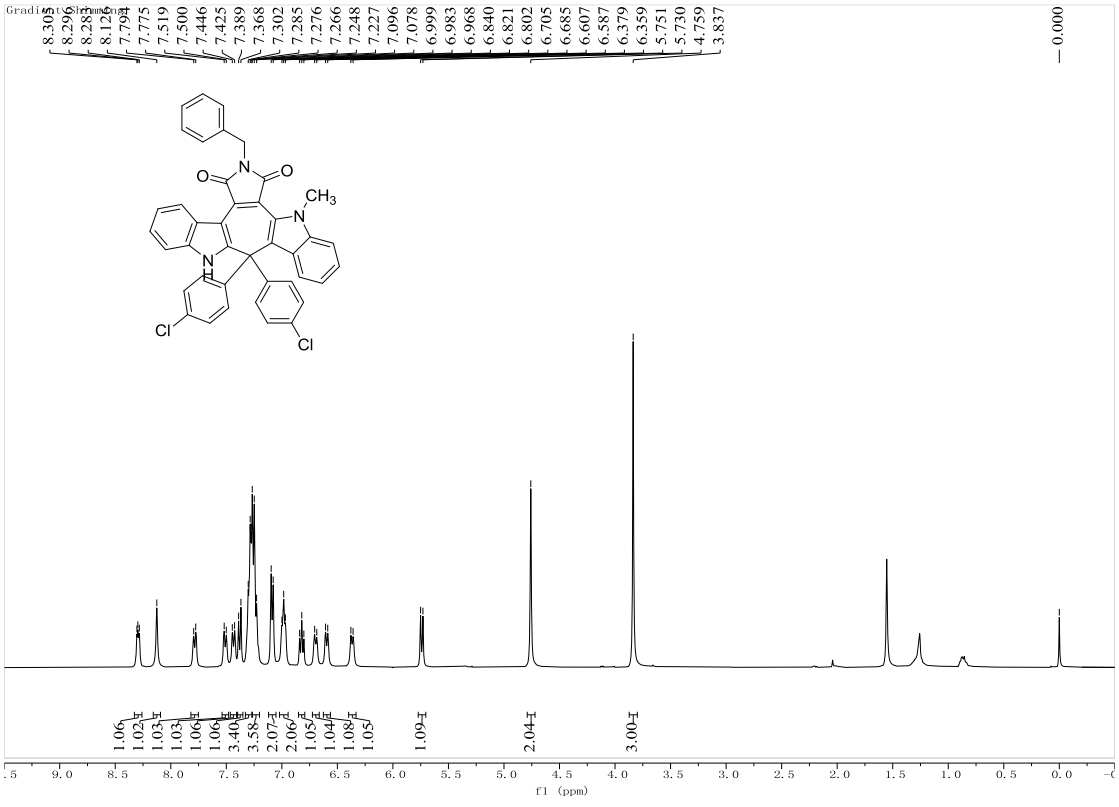
9,9-bis(4-chlorophenyl)-14-methyl-2-phenyl-9,14-dihydropyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole-1,3(2*H*,8*H*)-dione (3j): petroleum ether and ethyl acetate (V/V = 10:1) as the eluent, red solid, 576 mg, red solid, 88%, m.p. 231-233 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ: 8.37-8.36 (m, 1H, ArH), 8.17 (s, 1H, NH), 7.80 (d, *J* = 7.8 Hz, 1H, ArH), 7.52 (d, *J* = 7.8 Hz, 1H, ArH), 7.49-7.45 (m, 3H, ArH), 7.42-7.33 (m, 5H, ArH), 7.29-7.27 (m, 3H, ArH), 7.04 (d, *J* = 7.8 Hz, 2H, ArH), 6.86-6.84 (m, 1H, ArH), 6.76 (d, *J* = 7.8 Hz, 1H, ArH), 6.62 (d, *J* = 7.8 Hz, 1H, ArH), 6.44-6.43 (m, 1H, ArH), 5.78 (d, *J* = 8.4 Hz, 1H, ArH), 3.89 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 168.2, 167.7, 145.7, 141.5, 141.4, 140.7, 134.9, 134.3, 133.0, 132.6, 131.7, 131.0, 130.1, 129.3, 129.1, 129.0, 128.8, 128.1, 128.0, 127.6, 126.5, 126.3, 125.8, 125.6, 124.0, 123.9, 123.1, 122.2, 121.6, 121.3, 120.1, 111.4, 110.4, 107.0, 77.2, 55.4, 34.2; IR (KBr) υ: 3033, 2938, 2856, 2356, 1871, 1780, 1722, 1605, 1556, 1490, 1434, 1382, 1235, 1179, 1118, 845, 738 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{40}H_{25}Cl_2NaN_3O_2$ ([M+Na]$^+$): 672.1216, Found: 672.1201.

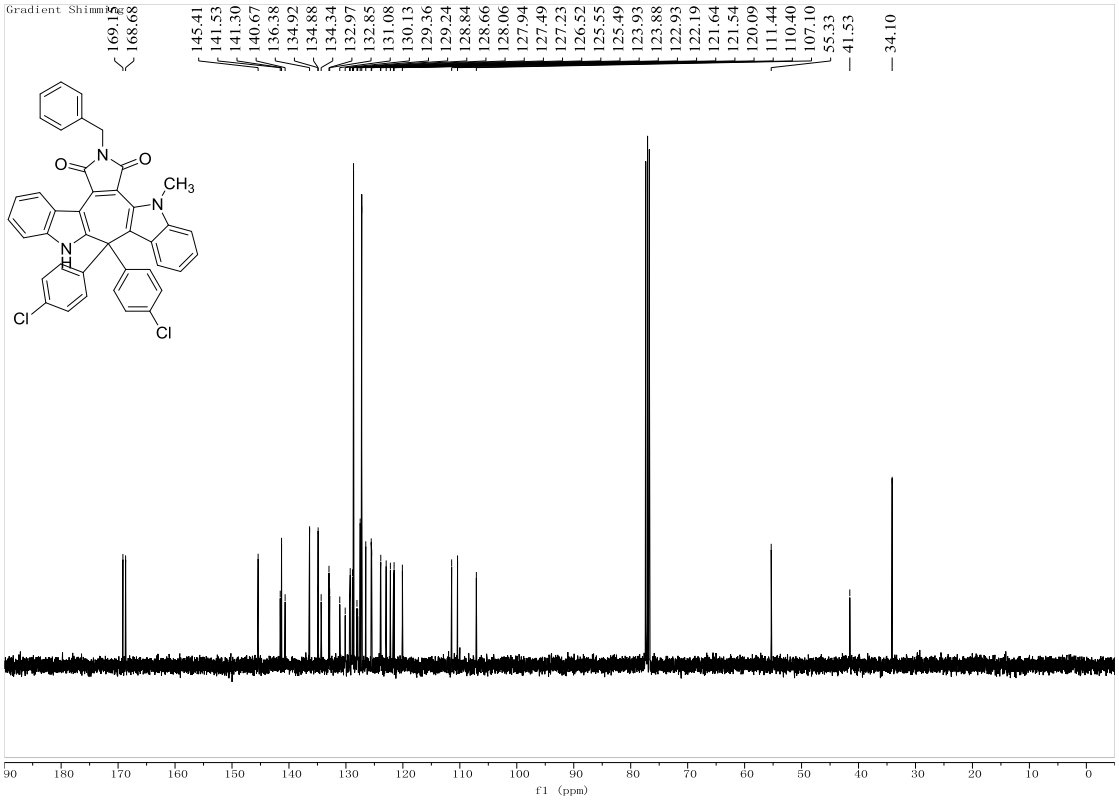


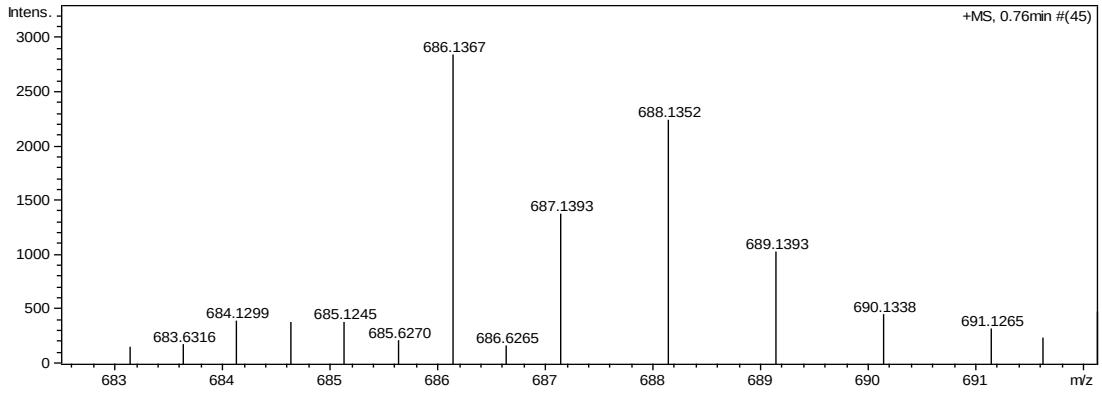

168.30
167.81
145.74
141.55
141.46
140.81
134.96
134.39
133.04
132.64
131.77
131.09
130.14
129.37
129.12
129.02
128.87
128.17
128.04
127.65
126.58
126.34
125.91
125.63
124.03
123.99
123.14
122.24
121.66
121.36
120.15
111.48
110.48
107.06
77.24
55.45
34.29
O
O
N
CH3
N
H
Cl
Cl
90 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


Intens.
x10^4
+MS, 0.62min #(37)
1.25
1.00
0.75
0.50
0.25
0.00
672.1201
674.1183
673.1235
675.1218
670.6147
671.1147
671.6137
672.6151
675.6750
676.1186
671
672
673
674
675
676
m/z

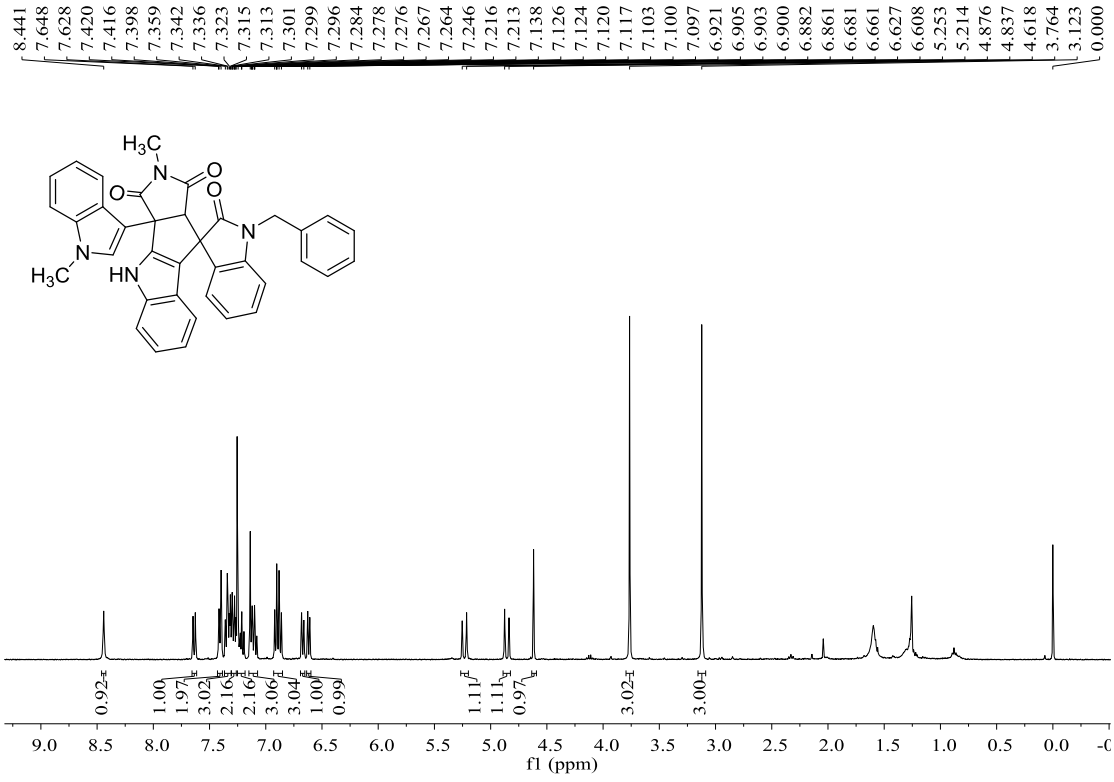
2-benzyl-9,9-bis(4-chlorophenyl)-14-methyl-9,14-dihydropyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole-1,3(2*H*,8*H*)-dione (3k): petroleum ether and ethyl acetate (V/V = 10:1) as the eluent, red solid, 590 mg, red solid, 89%, m.p. 216-218 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.31-8.28 (m, 1H, ArH), 8.13 (s, 1H, NH), 7.78 (d, *J* = 7.6 Hz, 1H, ArH), 7.51 (d, *J* = 7.6 Hz, 1H, ArH), 7.44 (d, *J* = 7.6 Hz, 1H, ArH), 7.38 (d, *J* = 8.4 Hz, 1H, ArH), 7.30-7.23 (m, 7H, ArH), 7.09 (d, *J* = 7.2 Hz, 2H, ArH), 6.98 (t, *J* = 6.8 Hz, 2H, ArH), 6.82 (t, *J* = 7.6 Hz, 1H, ArH), 6.70 (d, *J* = 8.0 Hz, 1H, ArH), 6.60 (d, *J* = 8.0 Hz, 1H, ArH), 6.37 (d, *J* = 8.0 Hz, 1H, ArH), 5.74 (d, *J* = 7.6 Hz, 1H, ArH), 4.76 (s, 2H, CH), 3.84 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 169.1, 168.6, 145.3, 141.5, 141.2, 140.6, 136.3, 134.9, 134.8, 134.3, 132.9, 132.8, 131.0, 130.1, 129.3, 129.2, 128.8, 128.6, 128.0, 127.9, 127.4, 127.2, 126.5, 125.5, 125.4, 123.9, 123.8, 122.9, 122.1, 121.6, 121.5, 120.0, 111.4, 110.3, 107.0, 55.3, 41.5, 34.0; IR (KBr) υ: 3035, 2934, 2852, 2366, 1878, 1765, 1723, 1643, 1556, 1424, 1443, 1371, 1256, 1190, 1122, 832, 744 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{41}H_{27}Cl_2NaN_3O_2$ ($[M+Na]^+$): 686.1373, Found: 686.1367.

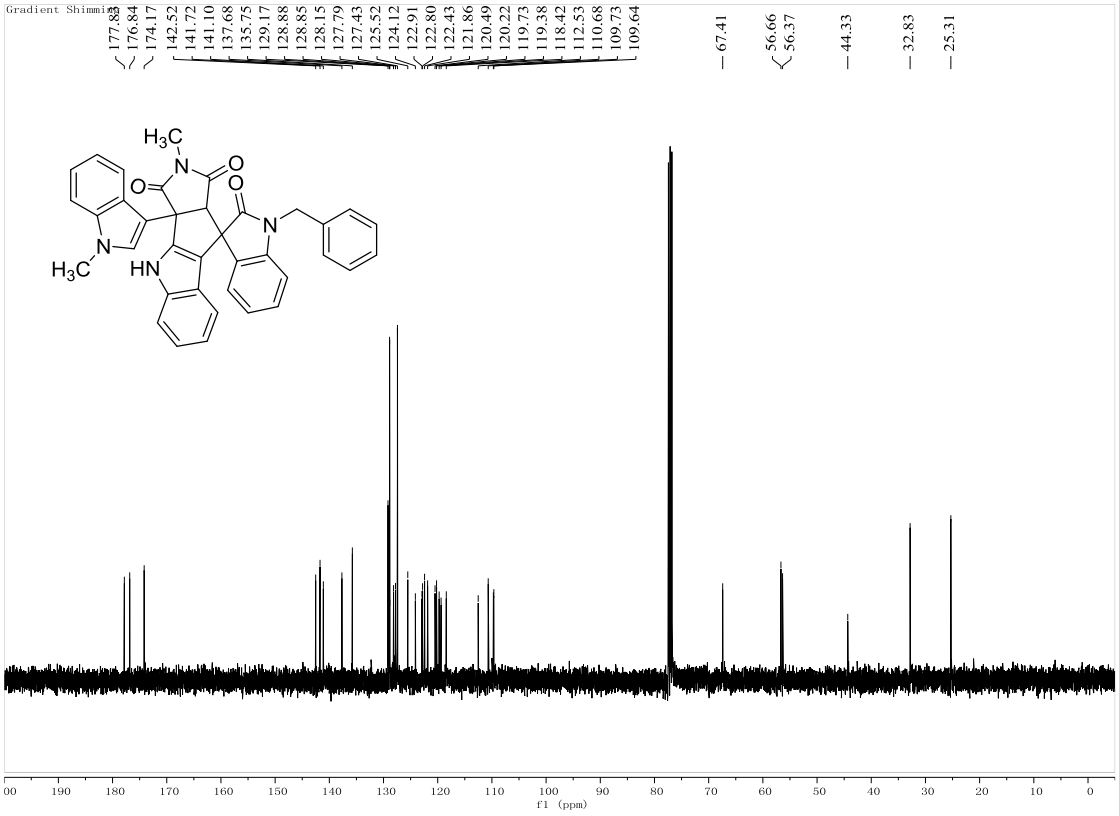


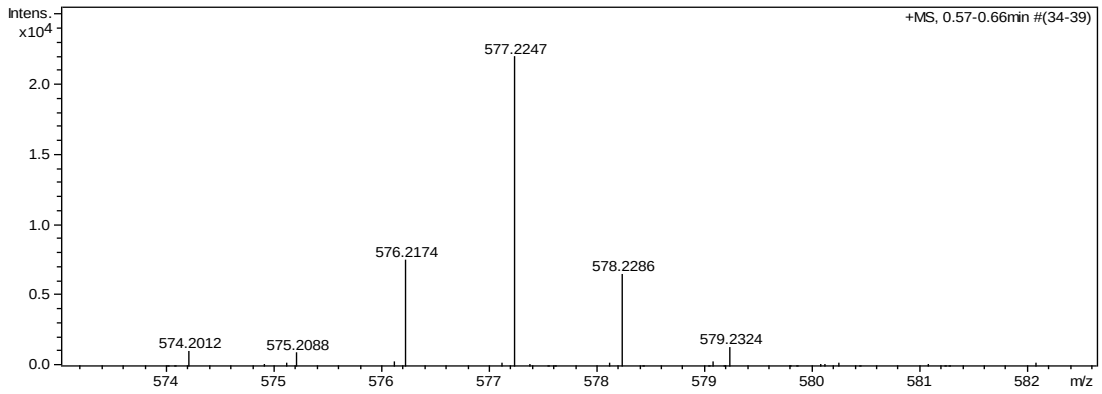

Gradient Shimming
169.16
168.68
145.41
141.53
141.30
140.67
136.38
134.92
134.88
134.34
132.97
132.85
131.08
130.13
129.36
129.24
128.84
128.66
128.06
127.94
127.49
127.23
126.52
125.55
125.49
123.93
123.88
122.93
122.19
121.64
121.54
120.09
111.44
110.40
107.10
55.33
41.53
34.10
O
O
N
CH3
N
H
Cl
Cl
90
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Intens.
+MS, 0.76min #(45)
3000
2500
2000
1500
1000
500
0
686.1367
688.1352
687.1393
689.1393
690.1338
691.1265
684.1299
685.1245
683.6316
685.6270
686.6265
683
684
685
686
687
688
689
690
691
m/z

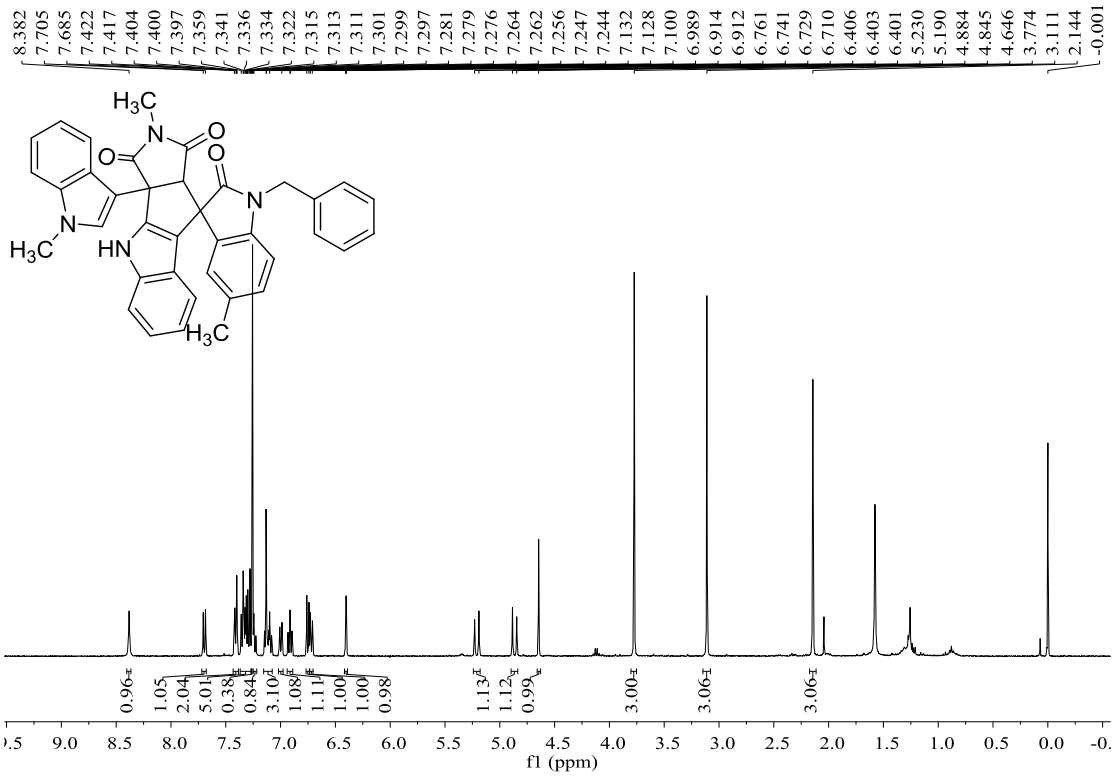
1-benzyl-2'-methyl-3a'-(1-methyl-1*H*-indol-3-yl)-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,4'*H*)-trione (5a): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 507 mg, white solid, 88%, m.p. 248-250 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.44 (s, 1H, NH), 7.64 (d, *J* = 8.0 Hz, 1H, ArH), 7.42-7.40 (m, 2H, ArH), 7.36-7.31 (m, 3H, ArH), 7.31-7.26 (m, 2H, ArH), 7.25-7.19 (m, 2H, ArH), 7.14-7.08 (m, 3H, ArH), 6.92-6.86 (m, 3H, ArH), 6.67 (d, J = 8.0 Hz, 1H, ArH), 6.62 (d, J = 7.6 Hz, 1H, ArH), 5.23 (d, *J* = 15.6 Hz, 1H, CH), 4.86 (d, *J* = 15.6 Hz, 1H, CH), 4.62 (s, 1H, CH), 3.76 (s, 3H, CH_3), 3.12 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 177.8, 176.8, 174.1, 142.5, 141.7, 141.0, 137.6, 135.7, 129.1, 128.8, 128.8, 128.1, 127.7, 127.4, 125.5, 124.1, 122.8, 122.7, 122.4, 121.8, 120.4, 120.2, 119.7, 119.3, 118.4, 112.5, 110.6, 109.7, 109.6, 67.4, 56.6, 56.3, 44.3, 32.8, 25.3; IR (KBr) υ: 3051, 2924, 2862, 2727, 2527, 2366, 1881, 1770, 1712, 1601, 1542, 1450, 1371, 1244, 1188, 1112, 820, 734 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{37}H_{29}N_4O_3$ ([M+H]$^+$): 577.2234, Found: 577.2247.

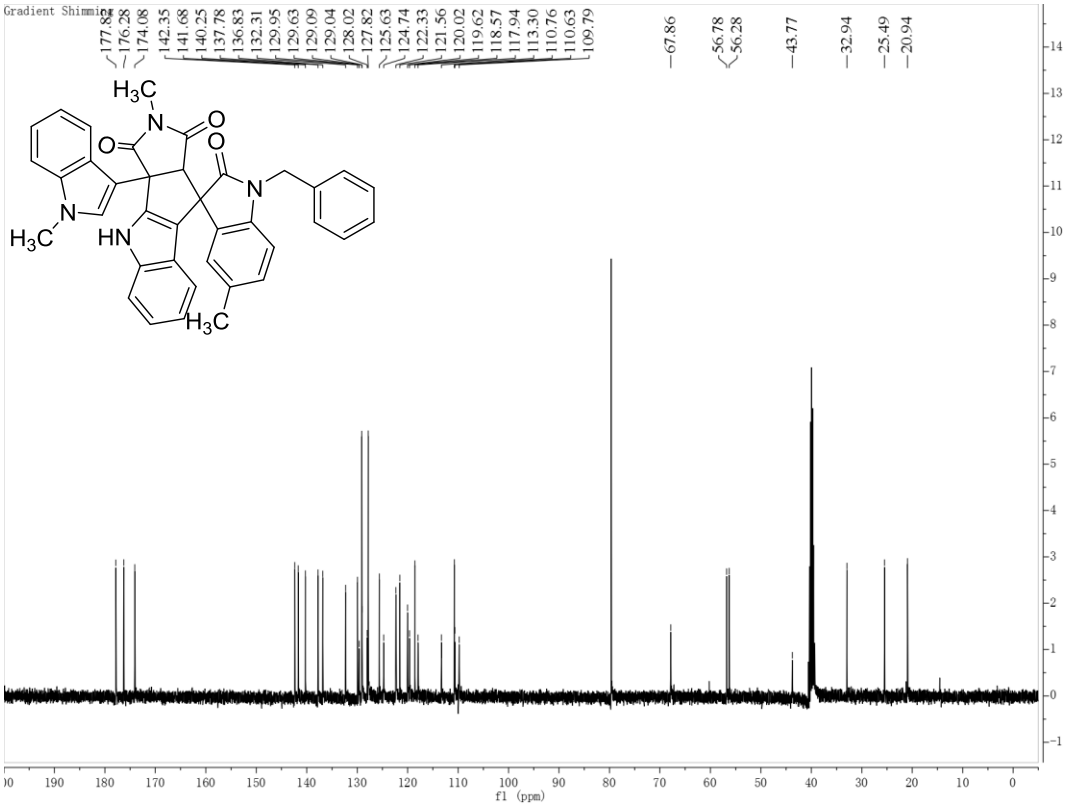


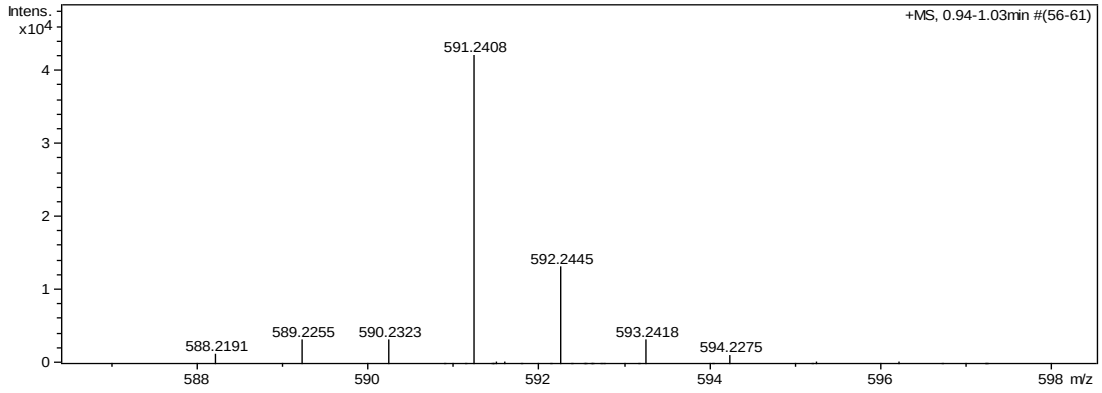

Gradient Shimming
177.83
176.84
174.17
142.52
141.72
141.10
137.68
135.75
129.17
128.88
128.85
128.15
127.79
127.43
125.52
124.12
122.91
122.80
122.43
121.86
120.49
120.22
119.73
119.38
118.42
112.53
110.68
109.73
109.64
67.41
56.66
56.37
44.33
32.83
25.31
H3C
N
O
O
O
N
N
H3C
HN
f1 (ppm)


Intens.
x10^4
+MS, 0.57-0.66min #(34-39)
577.2247
576.2174
578.2286
574.2012
575.2088
579.2324
m/z

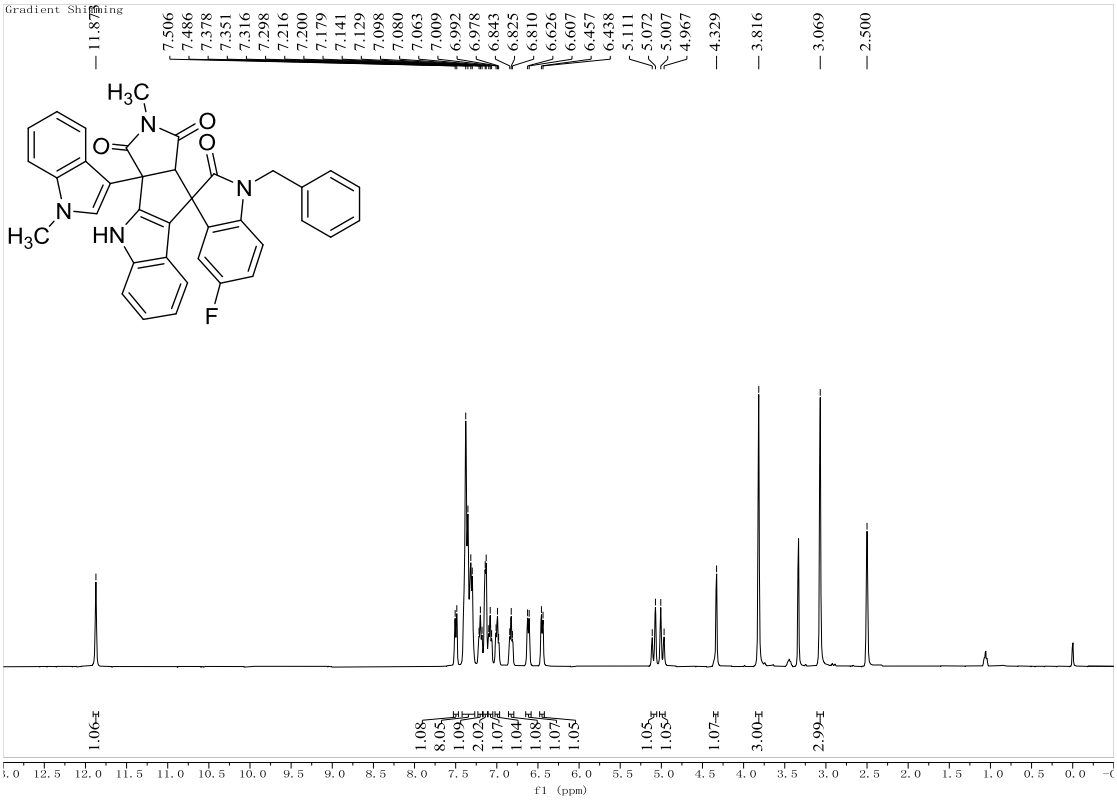
1-benzyl-2',5-dimethyl-3a'-(1-methyl-1*H*-indol-3-yl)-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',4':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,4'*H*)-trione (5b): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 513 mg, white solid, 87%, m.p. 232-234 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.38 (s, 1H, NH), 7.71-7.69 (m, 1H, ArH), 7.42-7.40 (m, 2H, ArH), 7.36-7.28 (m, 5H, ArH), 7.26-7.22 (m, 1H, ArH), 7.15-7.08 (m, 3H, ArH), 7.01-6.99 (m, 1H, ArH), 6.93-6.89 (m, 1H, ArH), 6.75 (d, J = 8.0 Hz, 1H, ArH), 6.72 (d, *J* = 7.6 Hz, 1H, ArH), 6.41-6.40 (m, 1H, ArH), 5.21 (d, *J* = 15.6 Hz, 1H, CH), 4.86 (d, *J* = 15.6 Hz, 1H, CH), 4.65 (s, 1H, CH), 3.77 (s, 3H, CH_3), 3.11 (s, 3H, CH_3); 2.14 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 177.8, 176.2, 174.0, 142.3, 141.6, 140.2, 137.7, 136.8, 132.2, 129.9, 129.6, 129.1, 129.0, 128.0, 127.8, 125.6, 124.7, 122.3, 121.5, 120.0, 119.6, 118.5, 117.9, 113.2, 110.7, 110.6, 109.7, 67.8, 56.7, 56.2, 43.7, 32.9, 25.4, 20.9; IR (KBr) υ: 3054, 2927, 2866, 2745, 2534, 2365, 1871, 1769, 1715, 1608, 1543, 1485, 1443, 1324, 1235, 1181, 1120, 823, 756 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{38}H_{31}N_4O_3$ ([M+H]$^+$): 591.2391, Found: 591.2408.

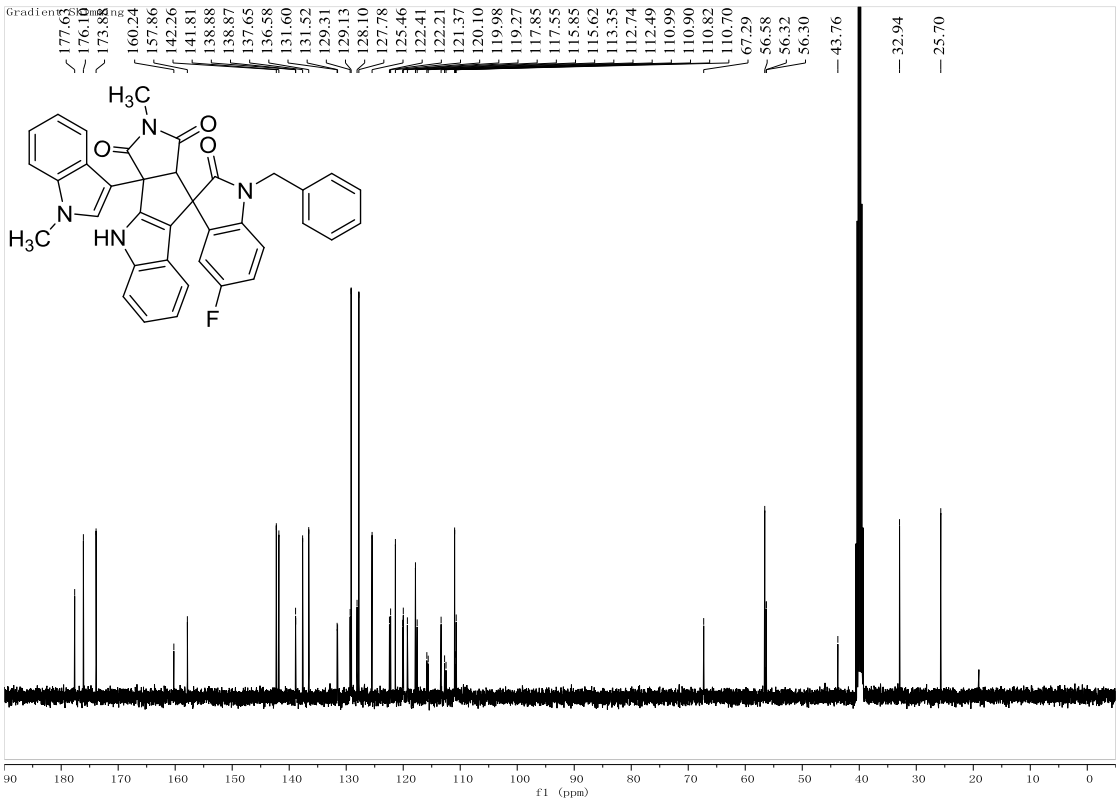


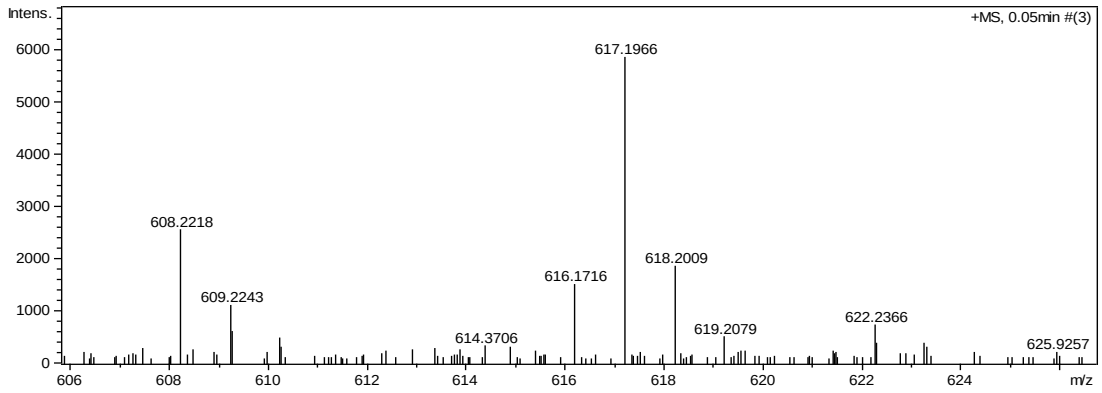

Gradient Shimming
177.80
176.28
174.08
142.35
141.68
140.25
137.78
136.83
132.31
129.95
129.63
129.09
129.04
128.02
127.82
125.63
124.74
122.33
121.56
120.02
119.62
118.57
117.94
113.30
110.76
110.63
109.79
67.86
56.78
56.28
43.77
32.94
25.49
20.94
f1 (ppm)


Intens.
x10^4
+MS, 0.94-1.03min #(56-61)
591.2408
592.2445
589.2255
590.2323
588.2191
593.2418
594.2275
m/z

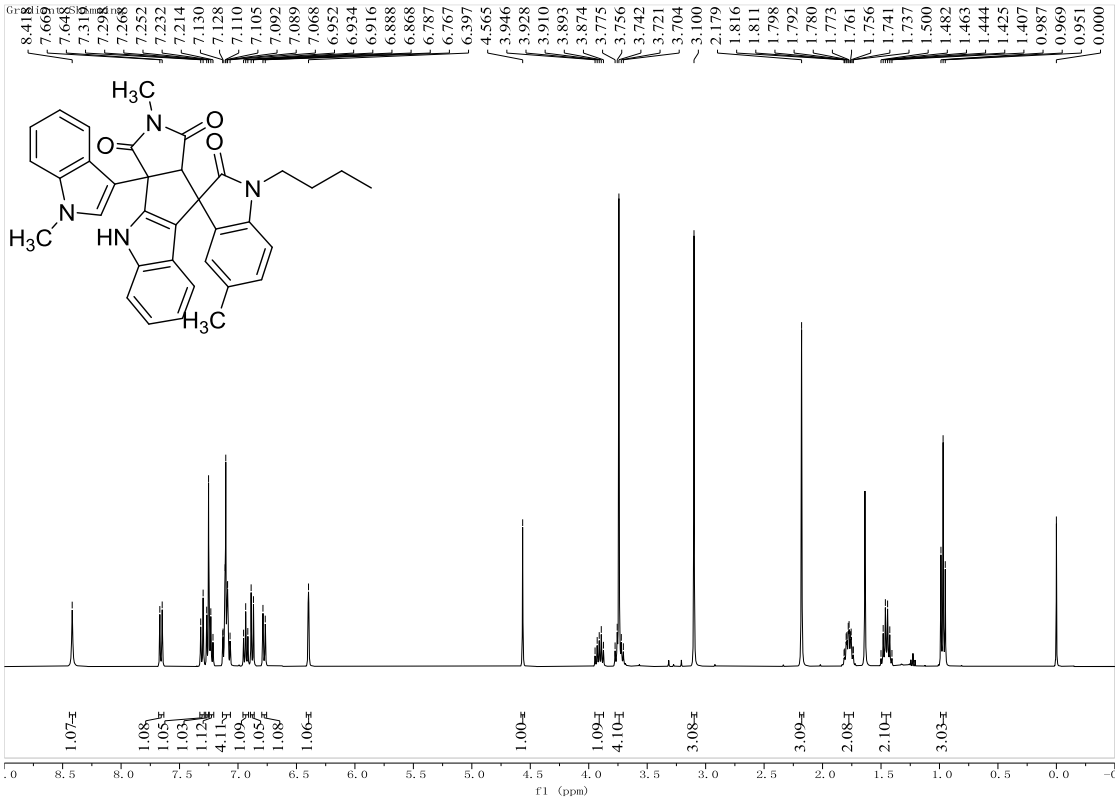
1-benzyl-5-fluoro-2'-methyl-3a'-(1-methyl-1*H*-indol-3-yl)-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,*4*'*H*)-trione (5c): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 535 mg, white solid, 90%, m.p. 227-229 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 11.87 (s, 1H, NH), 7.50 (d, *J* = 8.0 Hz, 1H, ArH), 7.38-7.30 (m, 8H, ArH), 7.22-7.18 (m, 1H, ArH), 7.24 (d, *J* = 4.8 Hz, 2H, ArH), 7.08 (t, *J* = 6.8 Hz, 1H, ArH), 6.99 (t, *J* = 6.8 Hz, 1H, ArH), 6.83 (t, *J* = 6.4 Hz, 1H, ArH), 6.62 (d, *J* = 7.6 Hz, 1H, ArH), 6.45 (d, *J* = 7.6 Hz, 1H, ArH), 5.09 (d, *J* = 15.6 Hz, 1H, CH), 4.99 (d, *J* = 15.6 Hz, 1H, CH), 4.33 (s, 1H, CH), 3.82 (s, 3H, CH_3), 3.07 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 177.6, 176.0, 173.8, 159.0 (d, *J* = 237.5 Hz), 142.2, 141.7, 138.9 (d, *J* = 1.8 Hz), 137.6, 136.5, 131.5 (d, *J* = 7.9 Hz), 129.2, 129.1, 128.0, 127.7, 125.4, 122.3, 122.1, 121.3, 120.0, 119.9, 119.2, 117.8, 117.5, 115.7 (d, *J* = 23.4 Hz), 113.3, 112.6 (d, *J* = 25.1 Hz), 110.9, 110.8, 110.8, 110.6, 67.2, 56.5, 56.3 (d, *J* = 1.5 Hz), 43.7, 32.9, 25.6; IR (KBr) υ: 3045, 2934, 2852, 2734, 2533, 2376, 1881, 1765, 1722, 1607, 1532, 1478, 1444, 1361, 1228, 1179, 1112, 828, 731 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{37}H_{27}NaFN_4O_3$ ([M+Na]$^+$): 617.1959, Found: 617.1966.

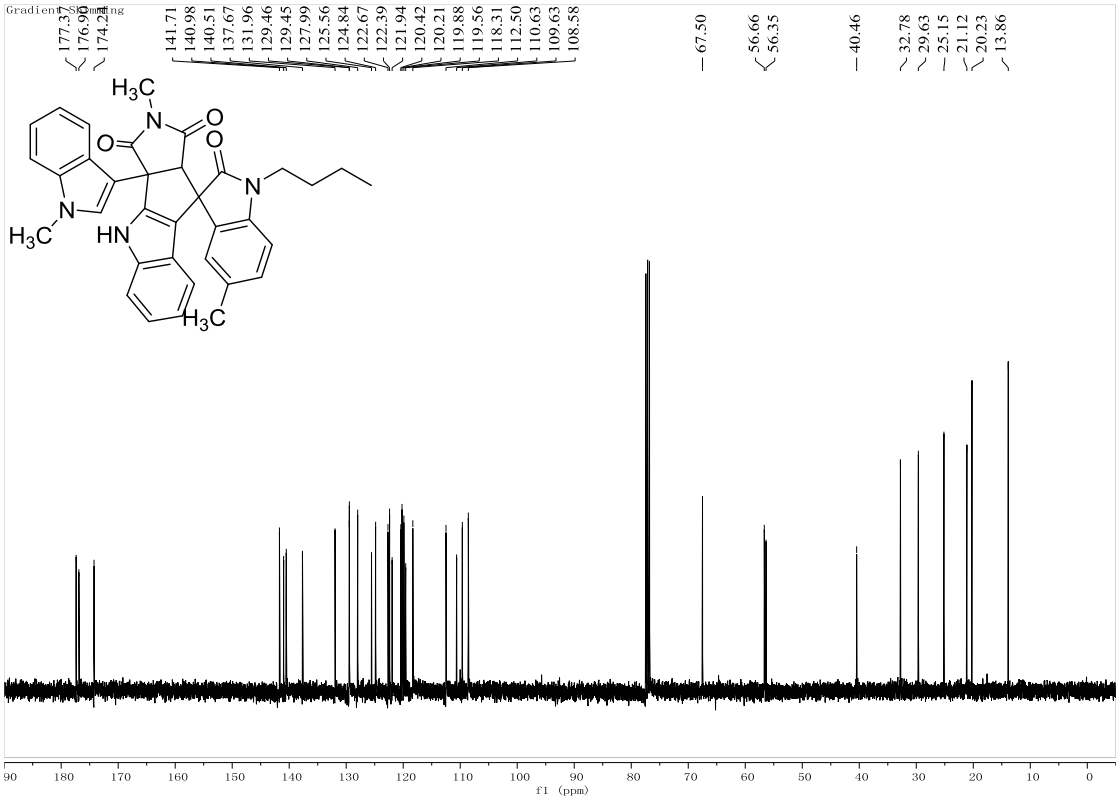


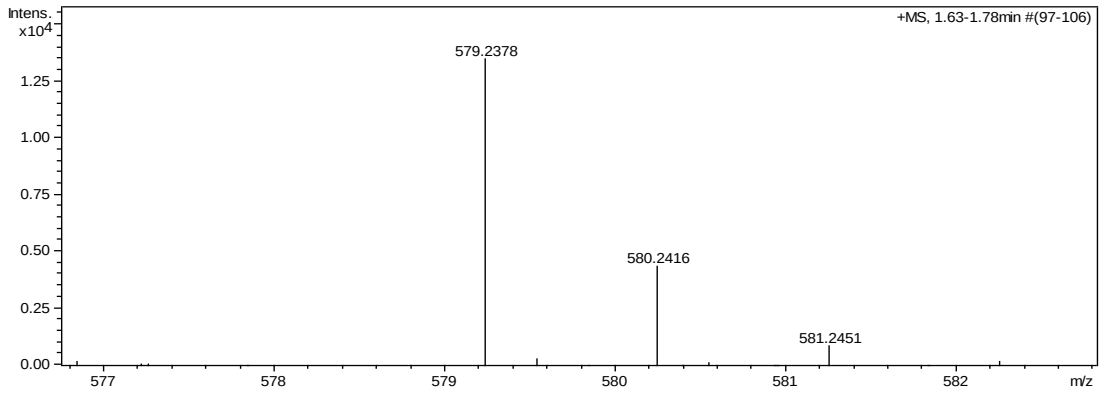

Gradient 13Cmang
177.67
176.10
173.88
160.24
157.86
142.26
141.81
138.88
138.87
137.65
136.58
131.60
131.52
129.31
129.13
128.10
127.78
125.46
122.41
122.21
121.37
120.10
119.98
119.27
117.85
117.55
115.85
115.62
113.35
112.74
112.49
110.99
110.90
110.82
110.70
67.29
56.58
56.32
56.30
43.76
32.94
25.70
H3C
N
O
O
O
N
N
H3C
HN
F
90
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Intens.
+MS, 0.05min #(3)
6000
5000
4000
3000
2000
1000
0
617.1966
608.2218
609.2243
616.1716
618.2009
614.3706
619.2079
622.2366
625.9257
606
608
610
612
614
616
618
620
622
624
m/z

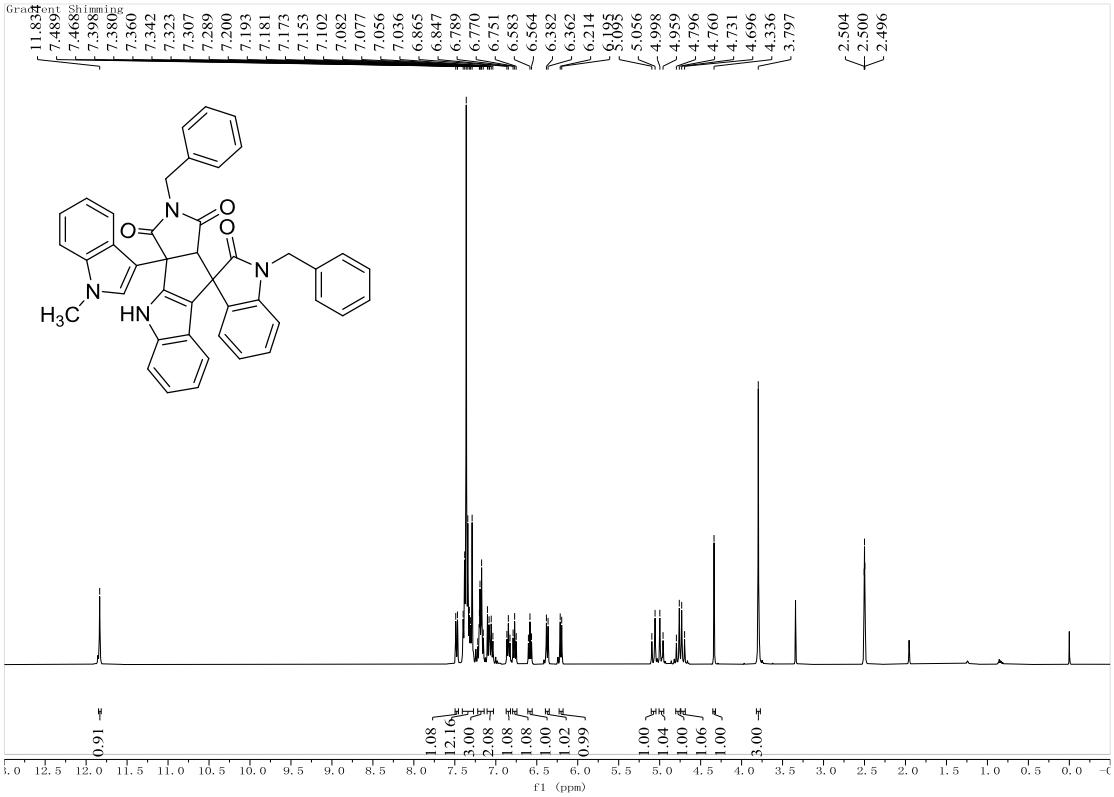
1-butyl-2',5-dimethyl-3a'-(1-methyl-1*H*-indol-3-yl)-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,*4*'*H*)-trione (5d): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 495 mg, white solid, 89%, m.p. 221-224 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.42 (s, 1H, NH), 7.66 (d, *J* = 8.0 Hz, 1H, ArH), 7.27-7.21 (m, 2H, ArH), 7.13-7.07 (m, 4H, ArH), 6.93 (t, *J* = 7.2 Hz, 1H, ArH), 6.88 (d, *J* = 8.0 Hz, 1H, ArH), 6.78 (t, *J* = 7.6 Hz, 1H, ArH), 6.40 (s, 1H, ArH), 4.57 (s, 1H, CH), 3.95-3.87 (m, 1H, CH), 3.76-3.70 (m, 1H, CH), 3.74 (s, 3H, CH_3), 3.10 (s, 3H, CH_3), 2.18 (s, 3H, CH_3), 1.82-1.74 (m, 2H, CH), 1.50-1.41 (m, 2H, CH), 0.97 (t, *J* = 7.2 Hz, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 177.3, 176.8, 174.2, 141.6, 140.9, 140.4, 137.6, 131.9, 129.5, 129.4, 127.9, 125.5, 124.8, 122.6, 122.3, 121.9, 120.4, 120.1, 119.8, 119.5, 118.2, 112.4, 110.6, 109.6, 108.5, 67.4, 56.6, 56.3, 40.4, 32.7, 29.6, 25.1, 21.1, 20.2, 13.8; IR (KBr) υ: 3048, 2921, 2860, 2734, 2537, 2356, 1871, 1768, 1723, 1611, 1534, 1479, 1445, 1378, 1248, 1178, 1122, 832, 743 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{35}H_{32}NaN_4O_3$ ([M+Na]$^{+}$): 579.2367, Found: 579.2378.

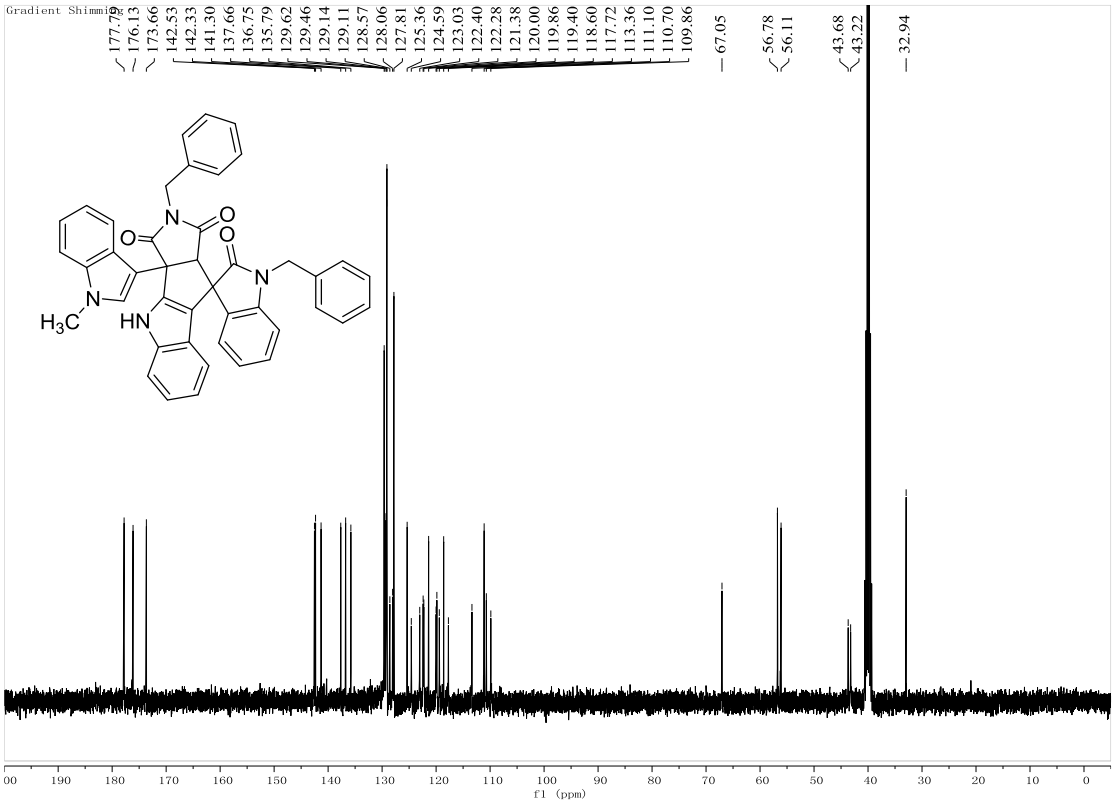


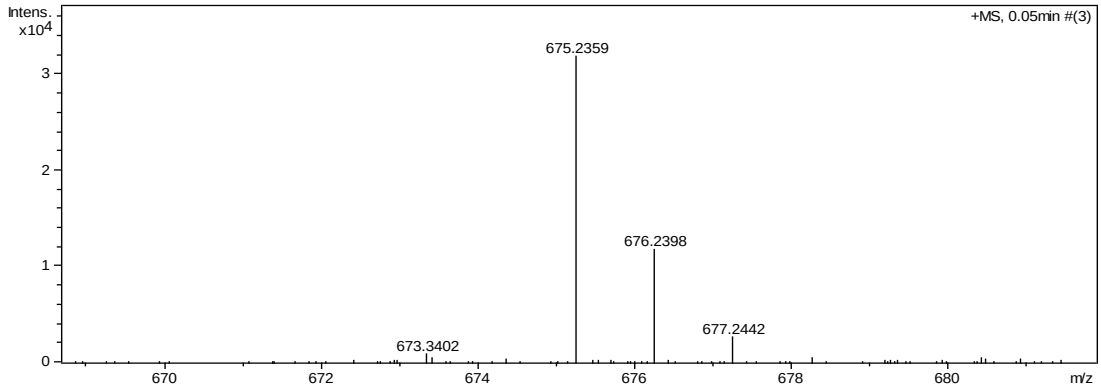

Gradient Spacing
177.37
176.90
174.23
141.71
140.98
140.51
137.67
131.96
129.46
129.45
127.99
125.56
124.84
122.67
122.39
121.94
120.42
120.21
119.88
119.56
118.31
112.50
110.63
109.63
108.58
67.50
56.66
56.35
40.46
32.78
29.63
25.15
21.12
20.23
13.86
H3C
N
O
O
O
N
N
H3C
HN
H3C
90
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Intens.
x10^4
+MS, 1.63-1.78min #(97-106)
579.2378
580.2416
581.2451
1.25
1.00
0.75
0.50
0.25
0.00
577
578
579
580
581
582
m/z

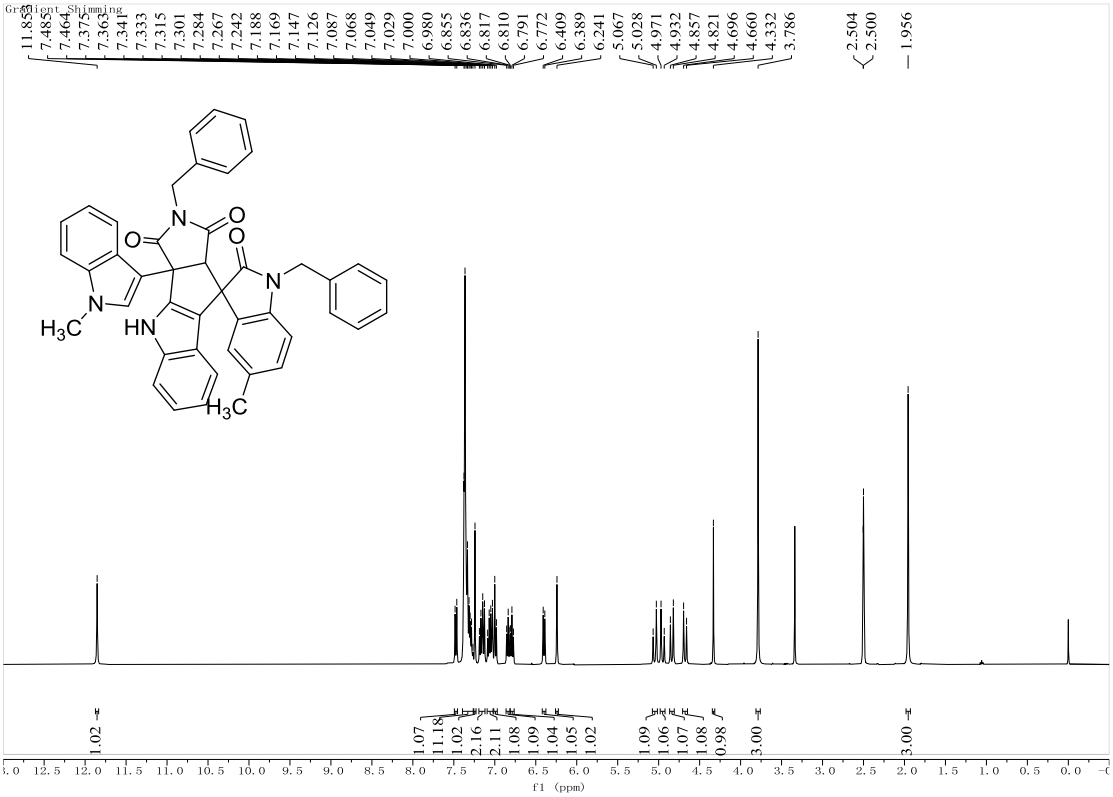
1,2'-dibenzyl-3a'-(1-methyl-1*H*-indol-3-yl)-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,4'*H*)-trione (5e): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 580 mg, white solid, 89%, m.p. 228-230 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 11.83 (s, 1H, NH), 7.48 (d, *J* = 8.4 Hz, 1H, ArH), 7.40-7.29 (m, 12H, ArH), 7.22-7.15 (m, 3H, ArH), 7.10-7.04 (m, 2H, ArH), 6.85 (t, *J* = 8.0 Hz, 1H, ArH), 6.77 (t, *J* = 7.6 Hz, 1H, ArH), 6.58 (t, *J* = 7.6 Hz, 1H, ArH), 6.37 (d, *J* = 8.0 Hz, 1H, ArH), 6.20 (d, *J* = 7.6 Hz, 1H, ArH), 5.08 (d, *J* = 15.6 Hz, 1H, CH), 4.98 (d, *J* = 15.6 Hz, 1H, CH), 4.78 (d, *J* = 14.2 Hz, 1H, CH), 4.71 (d, *J* = 14.2 Hz, 1H, CH), 4.34 (s, 1H, CH), 3.80 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 177.7, 176.1, 173.6, 142.5, 142.3, 141.2, 137.6, 136.7, 135.7, 129.6, 129.4, 129.1, 129.0, 128.5, 128.0, 127.8, 125.3, 124.5, 123.0, 122.3, 122.2, 121.3, 119.9, 119.8, 119.3, 118.5, 117.7, 113.3, 111.0, 110.6, 109.8, 67.0, 56.7, 56.0, 43.6, 43.2, 32.9; IR (KBr) υ: 3055, 2921, 2858, 2731, 2535, 2378, 1871, 1775, 1724, 1611, 1537, 1490, 1445, 1375, 1244, 1179, 1115, 825, 731 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{43}H_{32}NaN_4O_3$ ([M+Na]$^+$): 675.2367, Found: 675.2359.

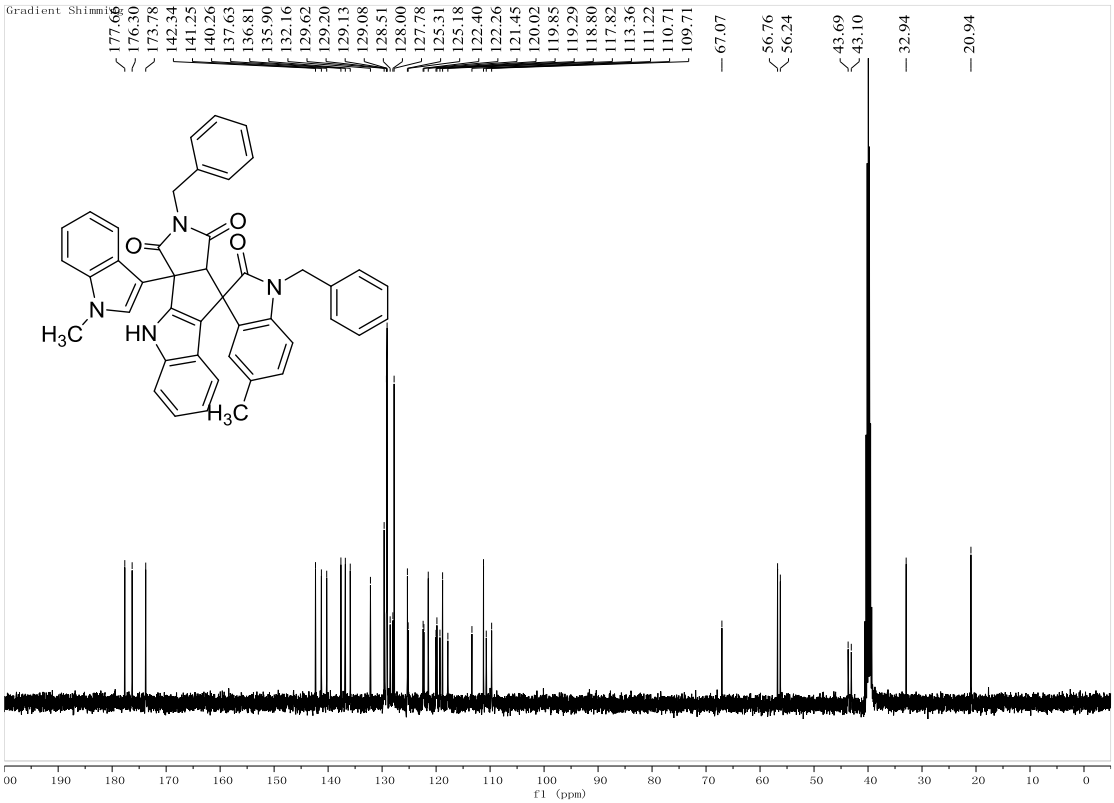


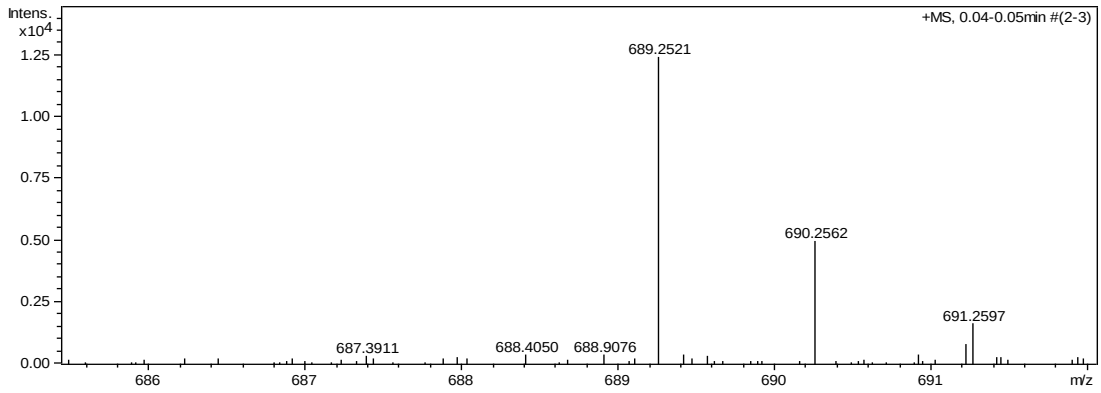

Gradient Shimming
177.79
176.13
173.66
142.53
142.33
141.30
137.66
136.75
135.79
129.62
129.46
129.14
129.11
128.57
128.06
127.81
125.36
124.59
123.03
122.40
122.28
121.38
120.00
119.86
119.40
118.60
117.72
113.36
111.10
110.70
109.86
67.05
56.78
56.11
43.68
43.22
32.94
O
O
O
N
N
HN
H_3C
f1 (ppm)


Intens.
x10^4
+MS, 0.05min #(3)
675.2359
676.2398
677.2442
673.3402
m/z

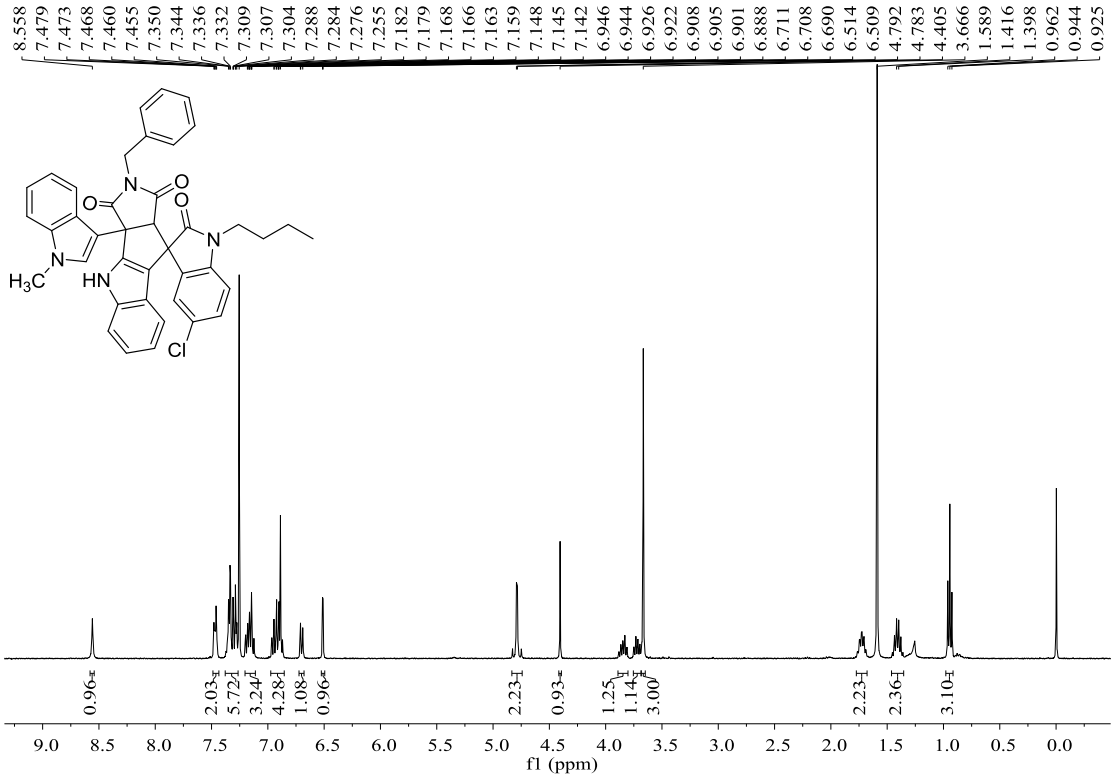
1,2'-dibenzyl-5-methyl-3a'-(1-methyl-1*H*-indol-3-yl)-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',4':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,4'*H*)-trione (5f): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 573 mg, white solid, 86%, m.p. 231-234 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 11.85 (s, 1H, NH), 7.47 (d, *J* = 8.0 Hz, 1H, ArH), 7.38-7.27 (m, 11H, ArH), 7.24 (s, 1H, ArH), 7.19-7.13 (m, 2H, ArH), 7.09-7.03 (m, 2H, ArH), 6.99 (d, *J* = 8.0 Hz, 1H, ArH), 6.84 (t, *J* = 7.6 Hz, 1H, ArH), 6.79 (t, *J* = 7.6 Hz, 1H, ArH), 6.40 (d, *J* = 8.0 Hz, 1H, ArH), 6.24 (s, 1H, ArH), 5.05 (d, *J* = 15.6 Hz, 1H, CH), 4.95 (d, *J* = 15.6 Hz, 1H, CH), 4.84 (d, *J* = 14.4 Hz, 1H, CH), 4.68 (d, *J* = 14.4 Hz, 1H, CH), 4.33 (s, 1H, CH), 3.79 (s, 3H, CH_3), 1.96 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 177.6, 176.2, 173.7, 142.3, 141.2, 140.2, 137.6, 136.7, 135.8, 132.1, 129.6, 129.2, 129.1, 129.0, 128.4, 127.9, 127.7, 125.2, 125.1, 122.3, 122.2, 121.4, 120.0, 119.8, 119.2, 118.7, 117.8, 113.3, 111.2, 110.6, 109.7, 67.0, 56.7, 56.2, 43.6, 43.0, 32.9, 20.9; IR (KBr) υ: 3060, 2931, 2858, 2727, 2528, 2367, 1878, 1765, 1718, 1608, 1545, 1482, 1458, 1375, 1248, 1185, 1124, 825, 731 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{44}H_{34}NaN_4O_3$ ([M+Na]$^+$): 689.2523, Found: 689.2521.

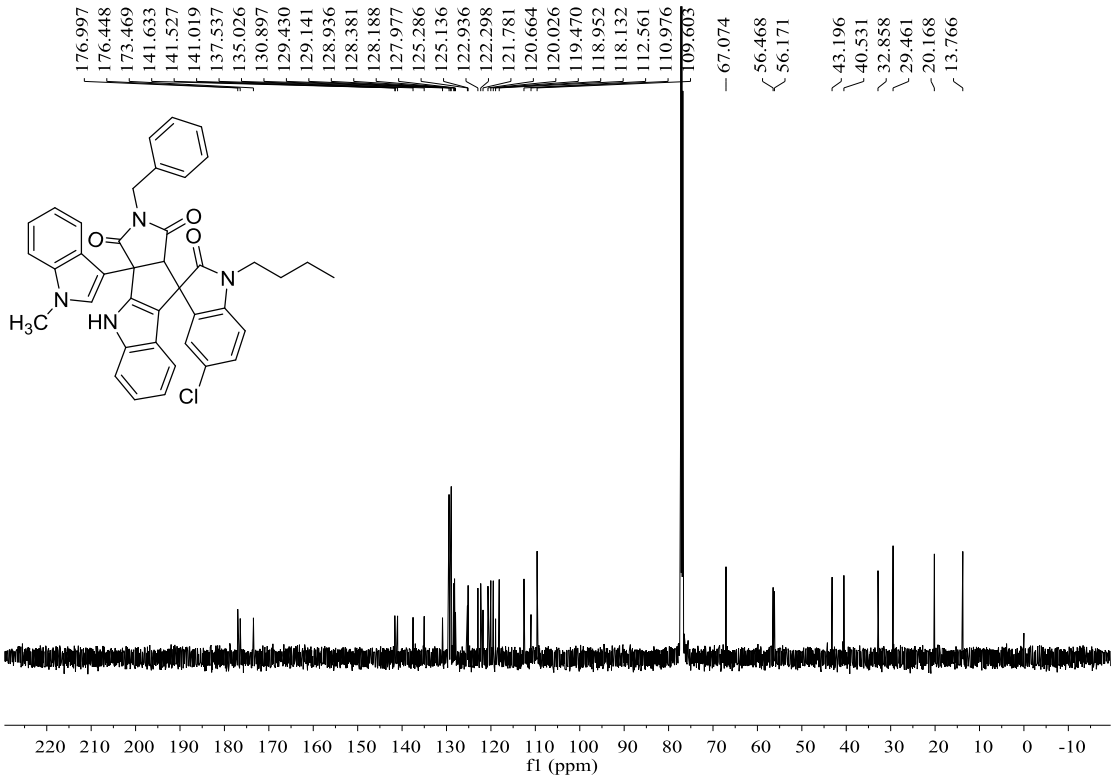


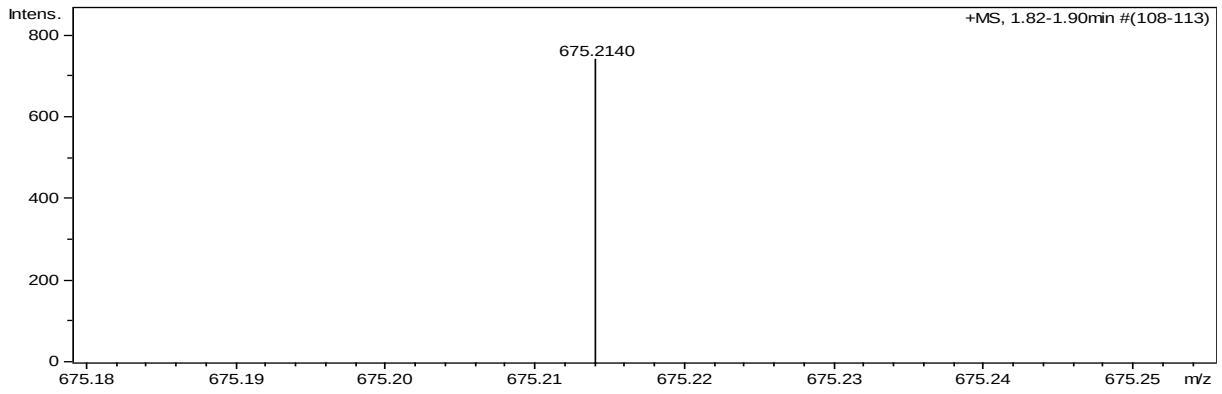

Gradient Shimming
177.66
176.30
173.78
142.34
141.25
140.26
137.63
136.81
135.90
132.16
129.62
129.20
129.13
129.08
128.51
128.00
127.78
125.31
125.18
122.40
122.26
121.45
120.02
119.85
119.29
118.80
117.82
113.36
111.22
110.71
109.71
67.07
56.76
56.24
43.69
43.10
32.94
20.94
O
O
O
N
N
N
HN
H3C
H3C
f1 (ppm)


Intens.
x10^4
1.25
1.00
0.75
0.50
0.25
0.00
+MS, 0.04-0.05min #(2-3)
689.2521
690.2562
691.2597
687.3911
688.4050
688.9076
686
687
688
689
690
691
m/z

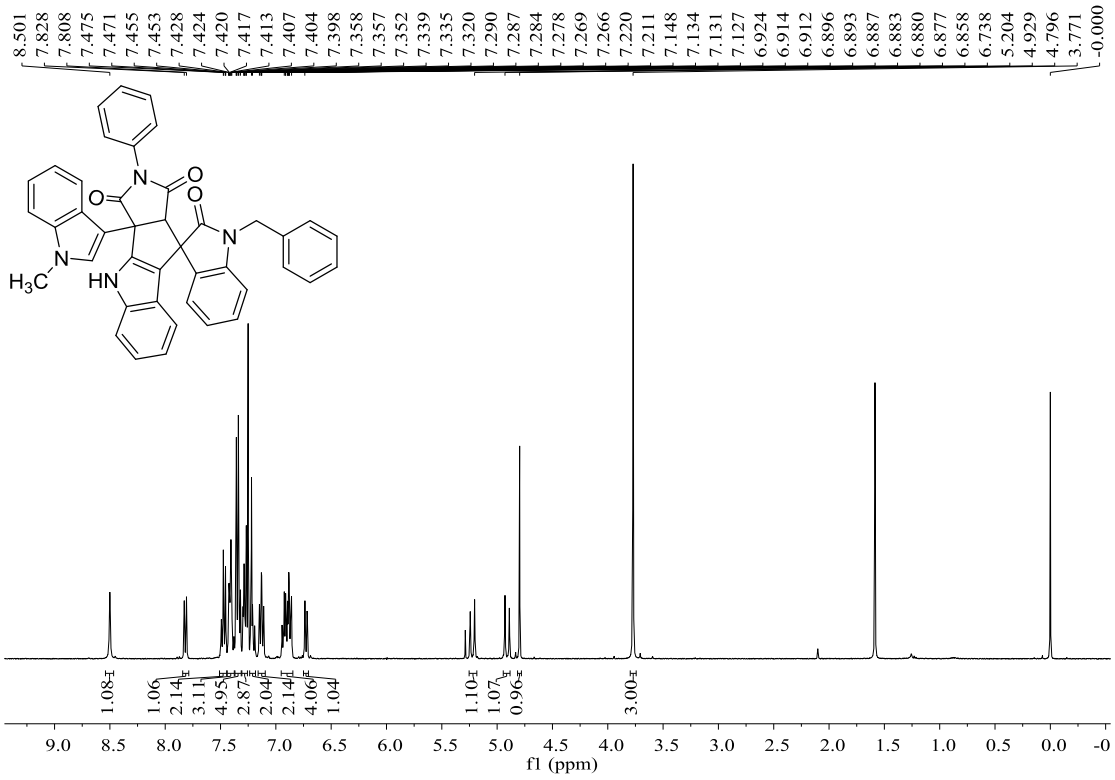
2'-benzyl-1-butyl-5-chloro-3a'-(1-methyl-1*H*-indol-3-yl)-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,4'*H*)-trione (5g): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 550 mg, white solid, 87%, m.p. >300 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.56 (s, 1H, NH), 7.48-7.46 (m, 2H, ArH), 7.37-7.32 (m, 3H, ArH), 7.31-7.28 (m, 2H, ArH), 7.20-7.12 (m, 3H, ArH), 6.97-6.87 (m, 4H, ArH), 6.70 (d, J = 8.0 Hz, 1H, ArH), 6.51 (d, J = 2.0 Hz, 1H, ArH), 4.81 (d, J = 13.6 Hz, 1H, CH), 4.77 (d, J = 13.6 Hz, 1H, CH), 4.41 (s, 1H, CH), 3.89-3.81 (m, 1H, CH), 3.75-3.70 (m, 1H, CH), 3.67 (s, 3H, CH_3); 1.77-1.69 (m, 2H, CH), 1.46-1.36 (m, 2H, CH), 0.95 (t, J = 7.2 Hz, 3H, CH); ^{13}C NMR (150 MHz, $CDCl_3$) δ: 177.0, 176.5, 173.5, 141.6, 141.5, 141.0, 137.5, 135.0, 130.9, 129.4, 129.1, 128.9, 128.4, 128.2, 128.0, 125.3, 125.1, 122.9, 122.3, 121.8, 120.7, 120.0, 119.5, 119.0, 118.1, 112.6, 111.0, 109.6, 67.1, 56.5, 56.2, 43.2, 40.5, 32.9, 29.5, 20.2, 13.8; IR (KBr) υ: 3054, 2931, 2856, 2731, 2532, 2368, 1873, 1766, 1709, 1611, 1548, 1478, 1460, 1372, 1250, 1175, 1124, 831, 732 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{40}H_{33}ClNaN_4O_3$ ([M+Na]$^{+}$): 675.2133, Found: 675.2133.

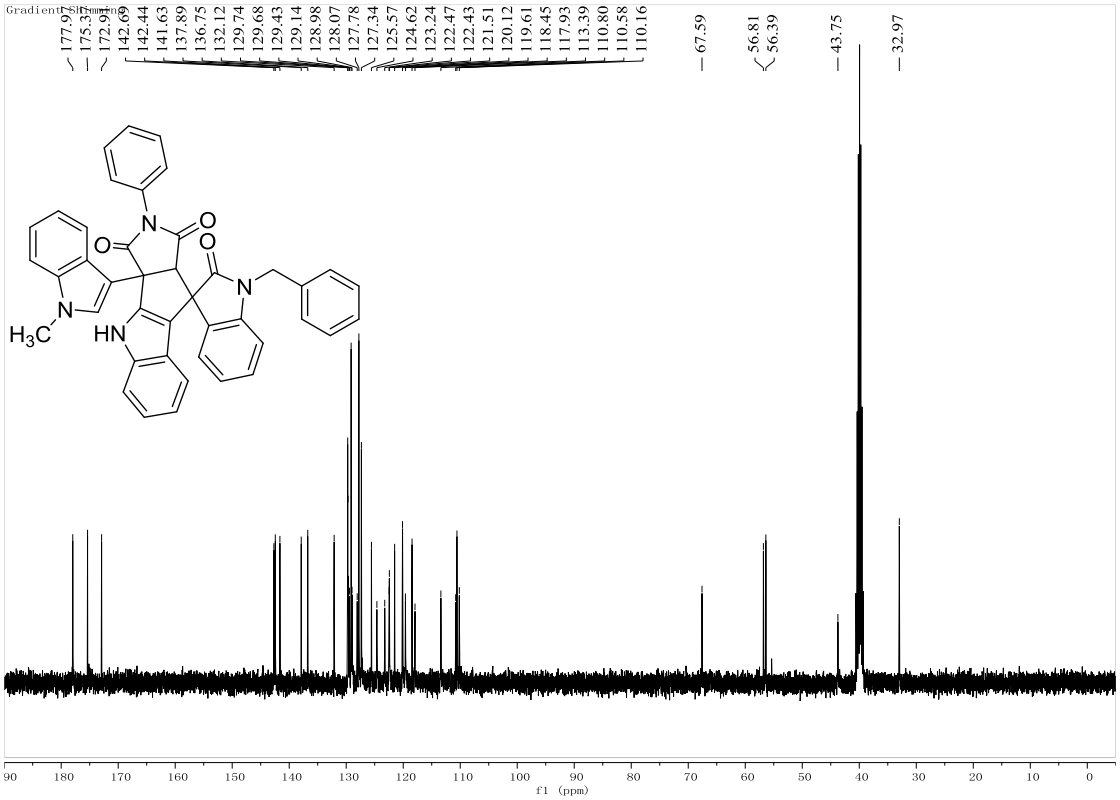


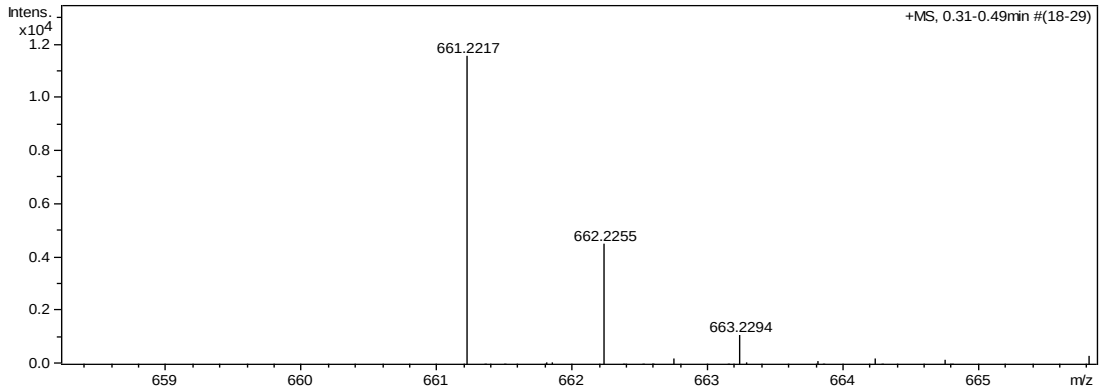

176.997
176.448
173.469
141.633
141.527
141.019
137.537
135.026
130.897
129.430
129.141
128.936
128.381
128.188
127.977
125.286
125.136
122.936
122.298
121.781
120.664
120.026
119.470
118.952
118.132
112.561
110.976
109.603
67.074
56.468
56.171
43.196
40.531
32.858
29.461
20.168
13.766
O
O
O
N
N
HN
H3C
Cl
220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


Intens.
+MS, 1.82-1.90min #(108-113)
675.2140
800
600
400
200
0
675.18
675.19
675.20
675.21
675.22
675.23
675.24
675.25
m/z

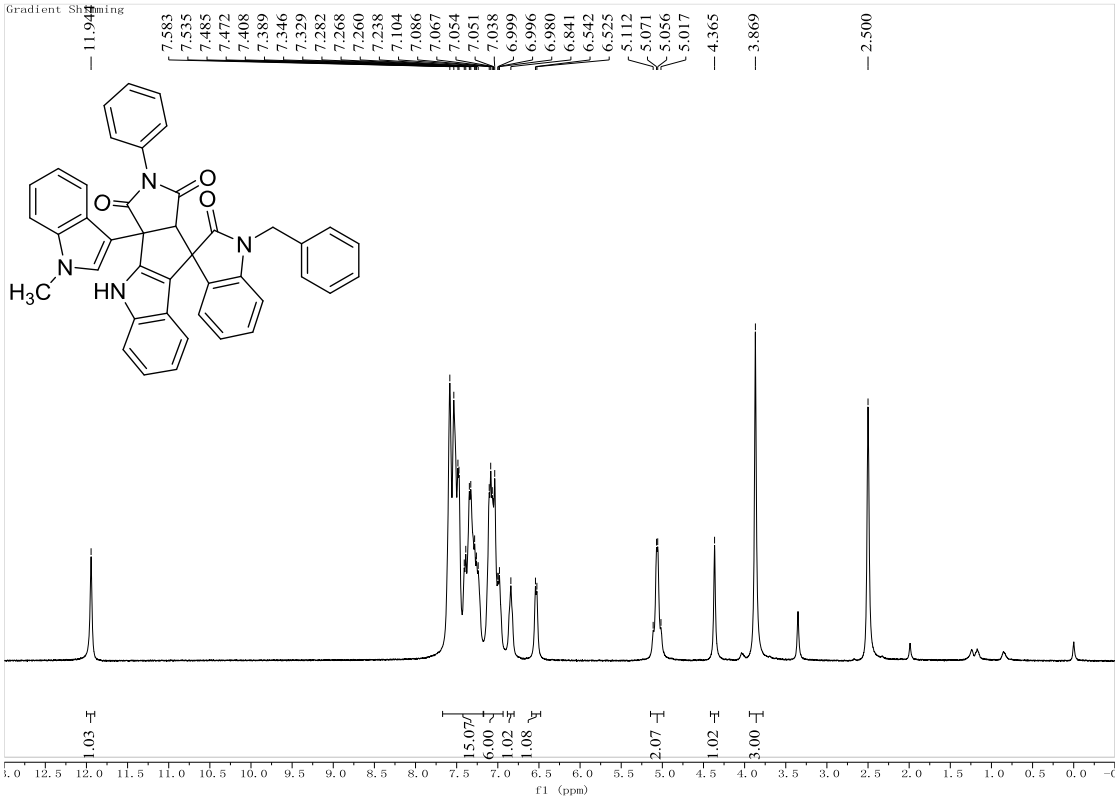
1-benzyl-3a'-(1-methyl-1*H*-indol-3-yl)-2'-phenyl-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,4'*H*)-trione (5h): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 421 mg, white solid, 66%, m.p. 231-233 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.50 (s, 1H, NH), 7.83-7.81 (m, 1H, ArH), 7.50-7.45 (m, 2H, ArH), 7.43-7.38 (m, 3H, ArH), 7.36-7.32 (m, 5H, ArH), 7.30-7.27 (m, 3H, ArH), 7.23-7.19 (m, 2H, ArH), 7.15-7.11 (m, 2H, ArH), 6.94-6.86 (m, 4H, ArH), 6.74-6.72 (m, 1H, ArH), 5.22 (d, J = 15.6 Hz, 1H, CH), 4.91 (d, J = 15.6 Hz, 1H, CH), 4.80 (s, 1H, CH), 3.77 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 177.9, 175.3, 172.8, 142.6, 142.4, 141.6, 137.8, 136.7, 132.1, 129.7, 129.6, 129.4, 129.1, 128.9, 128.0, 127.7, 127.3, 125.5, 124.6, 123.2, 122.4, 122.4, 121.4, 120.1, 119.5, 118.4, 117.9, 113.3, 110.7, 110.5, 110.1, 67.5, 56.8, 56.3, 43.7, 32.9; IR (KBr) υ: 3053, 2926, 2852, 2731, 2534, 2356, 1871, 1756, 1716, 1611, 1544, 1477, 1452, 1367, 1243, 1178, 1115, 830, 733 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{42}H_{30}NaN_4O_3$ ([M+Na]$^+$): 661.2210, Found: 661.2217.

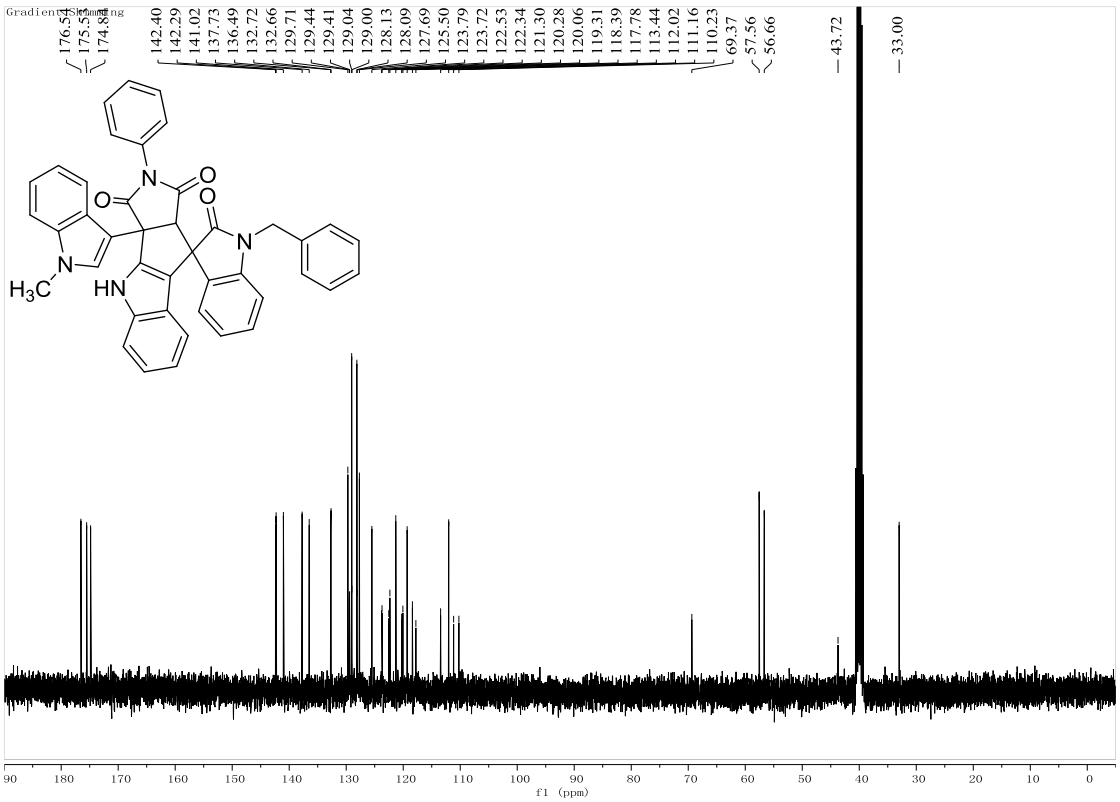


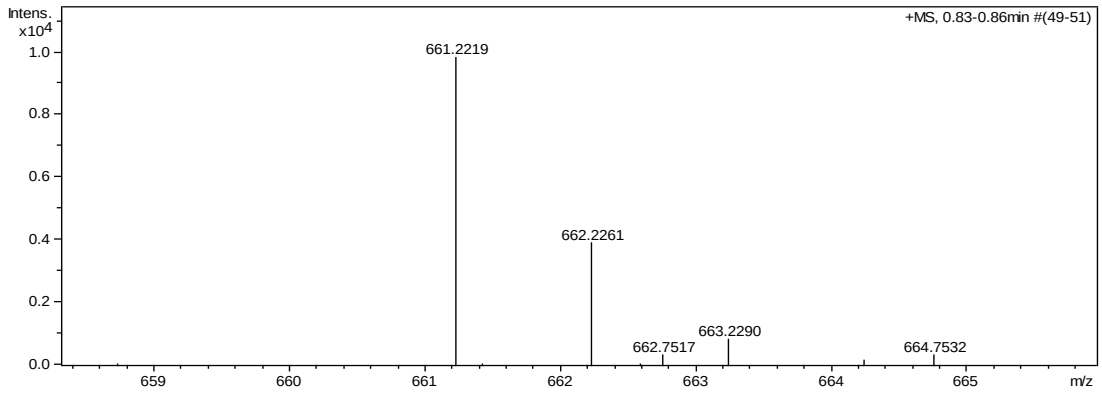

177.97
175.33
172.99
142.69
142.44
141.63
137.89
136.75
132.12
129.74
129.68
129.43
129.14
128.98
128.07
127.78
127.34
125.57
124.62
123.24
122.47
122.43
121.51
120.12
119.61
118.45
117.93
113.39
110.80
110.58
110.16
67.59
56.81
56.39
43.75
32.97
O
O
O
N
N
N
H3C
HN
90
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Intens.
x10^4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
+MS, 0.31-0.49min #(18-29)
661.2217
662.2255
663.2294
659
660
661
662
663
664
665
m/z

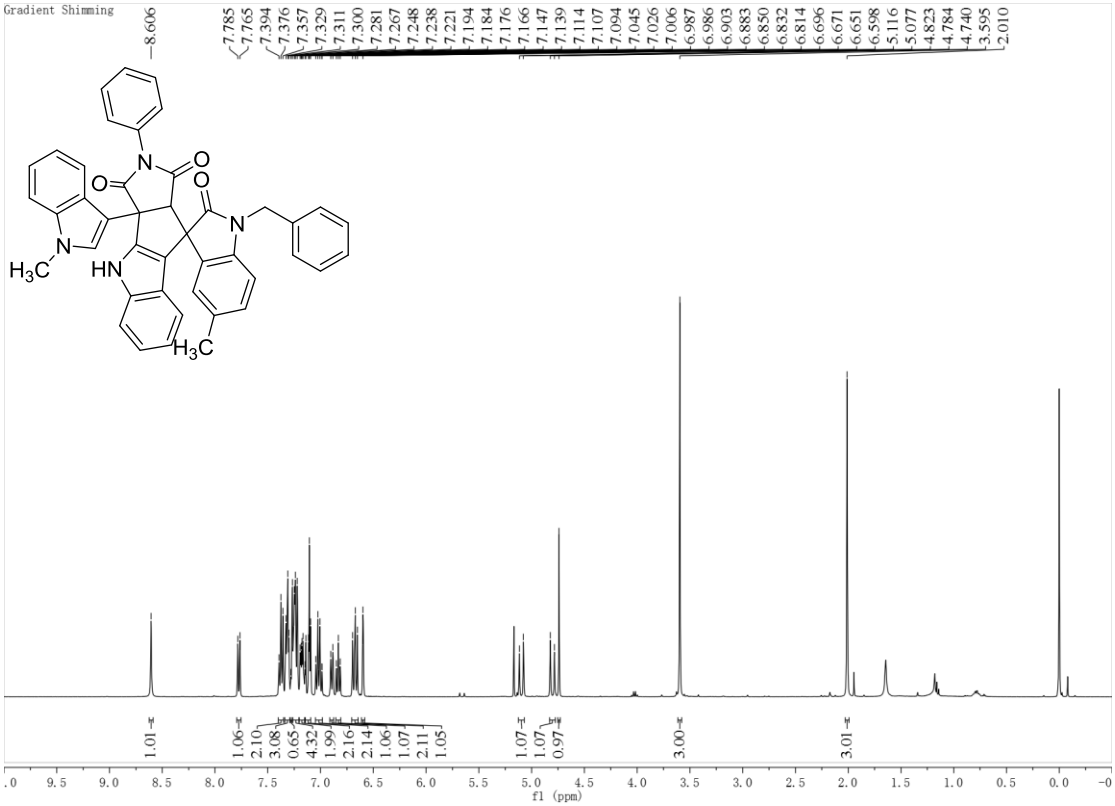
1-benzyl-3a'-(1-methyl-1*H*-indol-3-yl)-2'-phenyl-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',4':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,4'*H*)-trione (6h): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 109 mg, white solid, 17%, m.p. 236-238 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 11.94 (s, 1H, NH), 7.58-7.24 (m, 15H, ArH), 7.10-6.98 (m, 6H, ArH), 6.84 (s, 1H, ArH), 6.53 (d, J = 6.8 Hz, 1H, ArH), 5.11-5.02 (m, 2H, CH), 4.37 (s, 1H, CH), 3.87 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 176.5, 175.5, 174.8, 142.3, 142.2, 141.0, 137.7, 136.4, 132.7, 132.6, 129.6, 129.4, 129.3, 129.0, 128.9, 128.1, 128.0, 127.6, 125.4, 123.8, 123.7, 122.5, 122.3, 121.2, 120.2, 120.0, 119.2, 118.3, 117.7, 113.4, 112.0, 111.1, 110.2, 69.3, 57.5, 56.6, 43.7, 32.9; IR (KBr) υ: 3050, 2931, 2852, 2731, 2534, 2356, 1878, 1769, 1718, 1611, 1532, 1491, 1445, 1368, 1245, 1181, 1111, 821, 736 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{42}H_{30}NaN_4O_3$ ([M+Na]$^+$): 661.2210, Found: 661.2219.

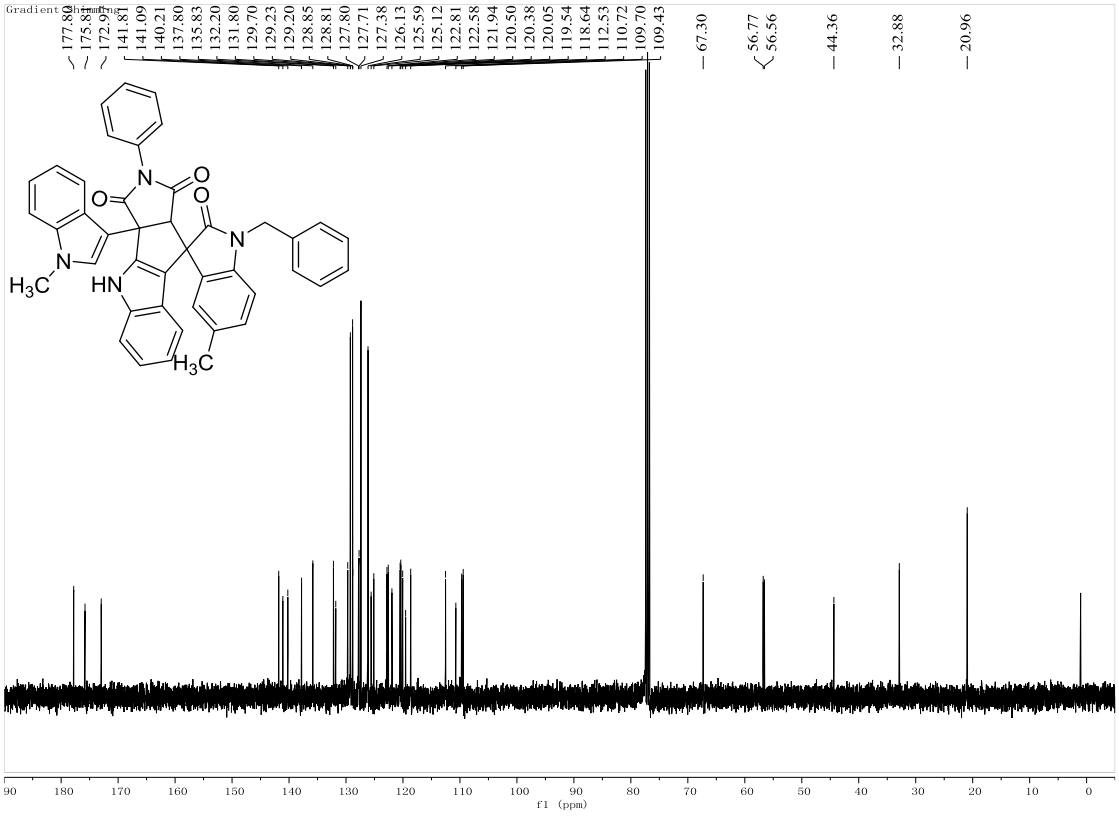


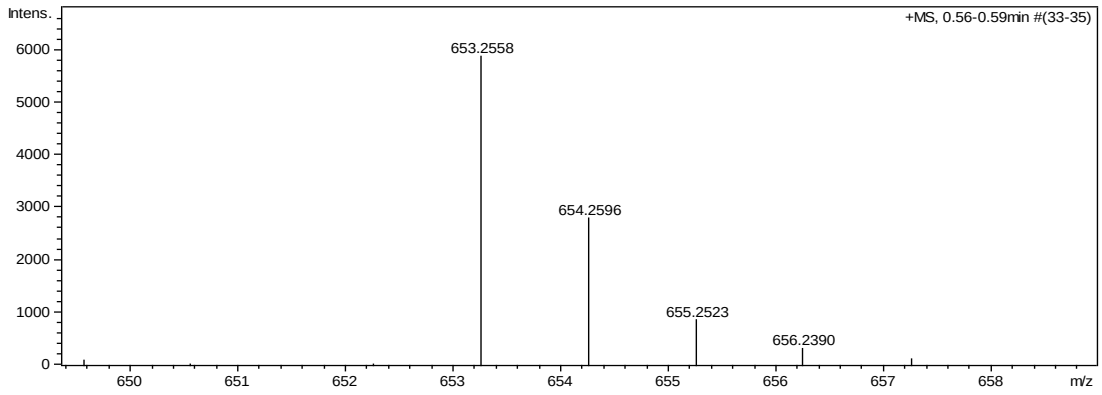

Gradient 3mmbang
176.54
175.53
174.88
142.40
142.29
141.02
137.73
136.49
132.72
132.66
129.71
129.44
129.41
129.04
129.00
128.13
128.09
127.69
125.50
123.79
123.72
122.53
122.34
121.30
120.28
120.06
119.31
118.39
117.78
113.44
112.02
111.16
110.23
69.37
57.56
56.66
43.72
33.00
O
O
O
N
N
N
HN
H3C
90 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


Intens.
x10^4
+MS, 0.83-0.86min #(49-51)
661.2219
662.2261
663.2290
662.7517
664.7532
1.0
0.8
0.6
0.4
0.2
0.0
659
660
661
662
663
664
665
m/z

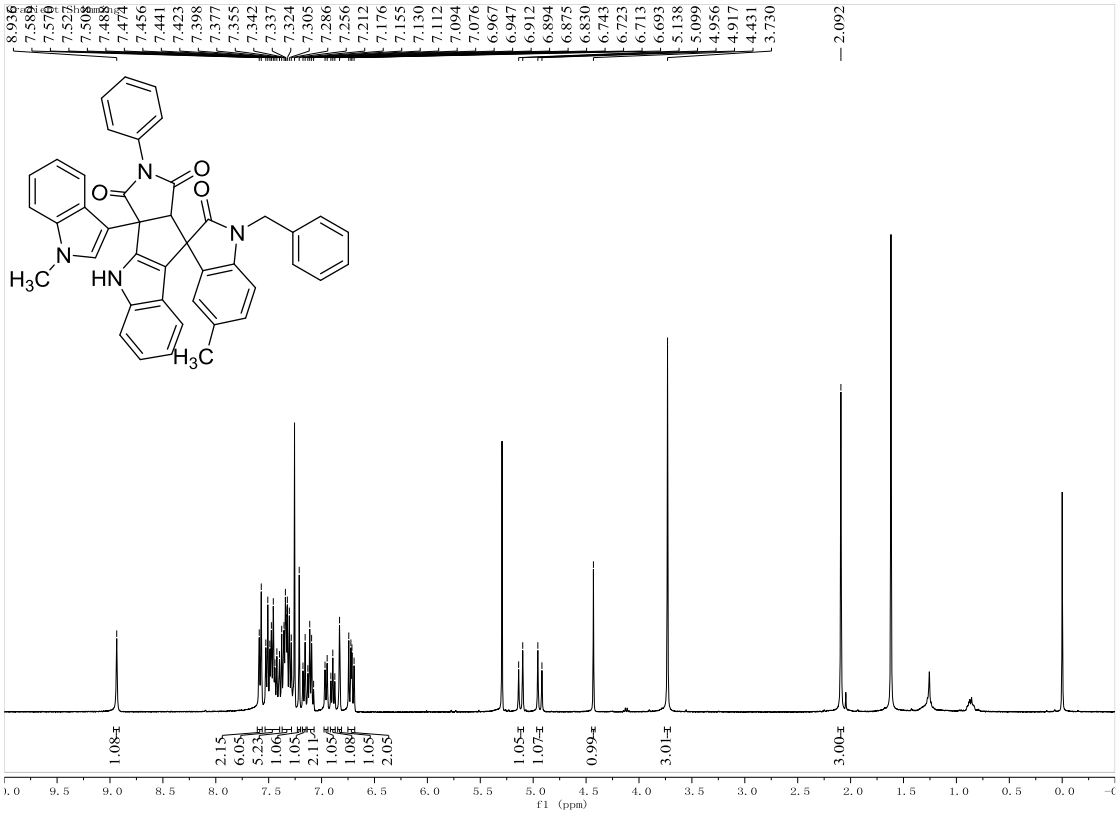
1-benzyl-5-methyl-3a'-(1-methyl-1*H*-indol-3-yl)-2'-phenyl-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,*4*'*H*)-trione (5i): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 437 mg, white solid, 67%, m.p. 215-217 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.61 (s, 1H, NH), 7.77 (d, *J* = 8.4 Hz, 1H, ArH), 7.38 (t, *J* = 8.0 Hz, 2H, ArH), 7.33-7.30 (m, 3H, ArH), 7.28-7.22 (m, 5H, ArH), 7.18 (dd, J_1 = 7.2 Hz, J_2 = 4.0 Hz, 2H, ArH), 7.15-7.09 (m, 2H, ArH), 7.05-6.99 (m, 2H, ArH), 6.89 (d, *J* = 8.0 Hz, 1H, ArH), 6.83 (t, *J* = 7.2 Hz, 1H, ArH), 6.70-6.65 (m, 2H, ArH), 6.60 (s, 1H, ArH), 5.10 (d, *J* = 15.6 Hz, 1H, CH), 4.80 (d, *J* = 15.6 Hz, 1H, CH), 4.74 (s, 1H, CH), 3.60 (s, 3H, CH_3), 2.01 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 177.7, 175.7, 172.9, 141.7, 141.0, 140.1, 137.7, 135.8, 132.1, 131.7, 129.6, 129.2, 129.1, 128.8, 128.7, 127.7, 127.6, 127.3, 126.1, 125.5, 125.1, 122.7, 122.5, 121.9, 120.4, 120.3, 120.0, 119.5, 118.6, 112.5, 110.7, 109.6, 109.4, 67.2, 56.7, 56.5, 44.3, 32.8, 20.9; IR (KBr) υ: 3048, 2931, 2853, 2731, 2532, 2363, 1875, 1765, 1718, 1611, 1538, 1485, 1445, 1368, 1234, 1179, 1118, 831, 733 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{43}H_{33}NaN_4O_3$ ([M+H]$^+$): 653.2547, Found: 653.2558.

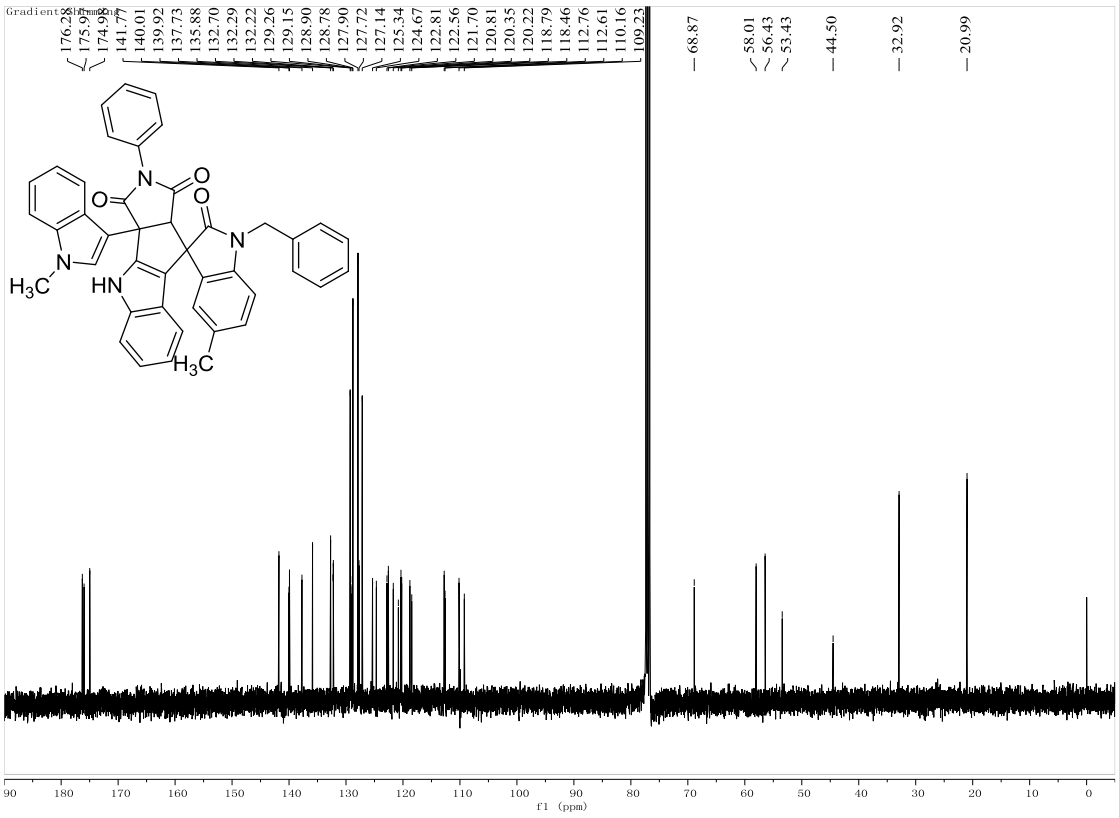


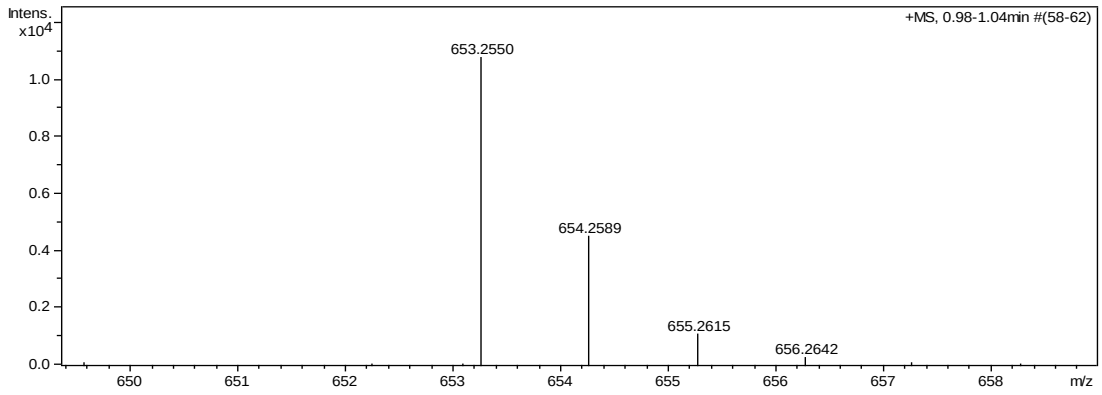

177.80
175.85
172.95
141.87
141.09
140.21
137.80
135.83
132.20
131.80
129.70
129.23
129.20
128.85
128.81
127.80
127.71
127.38
126.13
125.59
125.12
122.81
122.58
121.94
120.50
120.38
120.05
119.54
118.64
112.53
110.72
109.70
109.43
67.30
56.77
56.56
44.36
32.88
20.96
O
O
O
N
N
HN
H3C
H3C
90 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


Intens.
+MS, 0.56-0.59min #(33-35)
6000
5000
4000
3000
2000
1000
0
653.2558
654.2596
655.2523
656.2390
650 651 652 653 654 655 656 657 658 m/z

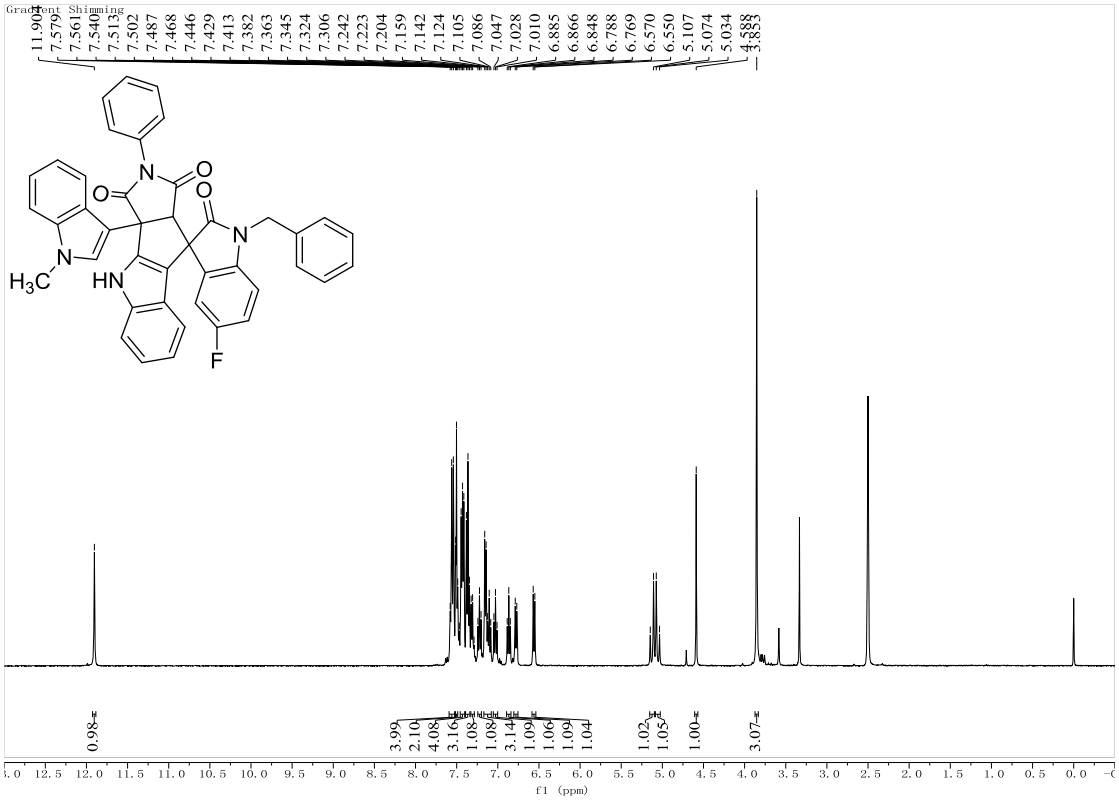
1-benzyl-5-methyl-3a'-(1-methyl-1*H*-indol-3-yl)-2'-phenyl-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,*4*'*H*)-trione (6i): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 104 mg, 16%, white solid, 16%, m.p. 222-225 ºC; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 8.94 (s, 1H, NH), 7.58 (d, *J* = 7.6 Hz, 2H, ArH), 7.53-7.40 (m, 6H, ArH), 7.38-7.29 (m, 5H, ArH), 7.21 (s, 1H, ArH), 7.17 (d, *J* = 8.4 Hz, 1H, ArH), 7.10 (dd, J_1 = 14.4 Hz, J_2 = 7.2 Hz, 2H, ArH), 6.96 (d, *J* = 8.0 Hz, 1H, ArH), 6.89 (t, *J* = 7.6 Hz, 1H, ArH), 6.83 (s, 1H, ArH), 6.72 (dd, J_1 = 12.0 Hz, J_2 = 8.0 Hz, 2H, ArH), 5.12 (d, *J* = 15.6 Hz, 1H, CH), 4.94 (d, *J* = 15.6 Hz, 1H, CH), 4.43 (s, 1H, CH), 3.73 (s, 3H, CH_3), 2.09 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $CDCl_3$) δ: 176.2, 175.9, 174.9, 141.7, 139.9, 139.8, 137.7, 135.8, 132.6, 132.3, 132.2, 129.2, 129.1, 128.8, 128.7, 127.8, 127.7, 127.1, 125.3, 124.6, 122.7, 122.5, 121.6, 120.7, 120.3, 120.2, 118.7, 118.4, 112.7, 112.6, 110.1, 109.2, 68.8, 58.0, 56.4, 53.4, 44.4, 32.9, 20.9; IR (KBr) υ: 3044, 2921, 2852, 2737, 2531, 2363, 1871, 1777, 1715, 1611, 1541, 1481, 1454, 1365, 1243, 1178, 1108, 834, 732 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{43}H_{33}NaN_4O_3$ ($[M+H]^+$): 653.2547, Found: 653.2550.

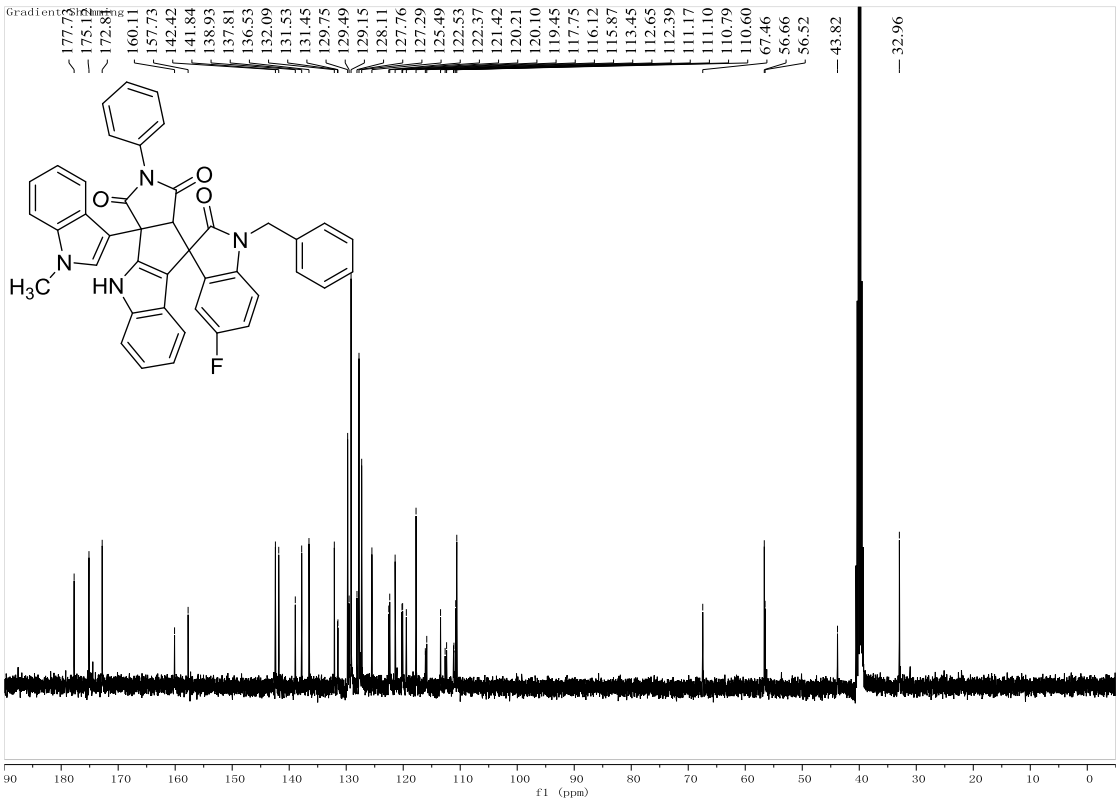


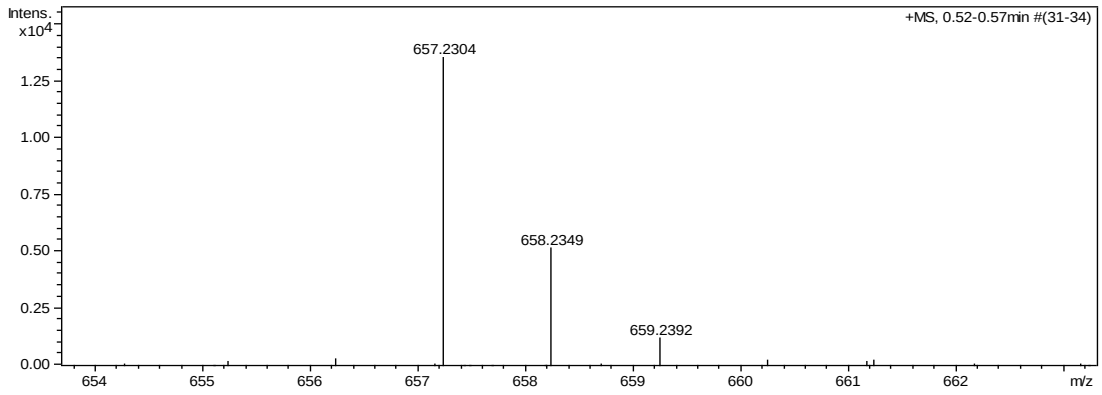

176.28
175.97
174.98
141.77
140.01
139.92
137.73
135.88
132.70
132.29
132.22
129.26
129.15
128.90
128.78
127.90
127.72
127.14
125.34
124.67
122.81
122.56
121.70
120.81
120.35
120.22
118.79
118.46
112.76
112.61
110.16
109.23
68.87
58.01
56.43
53.43
44.50
32.92
20.99
O
O
O
N
N
HN
H3C
H3C
90 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


Intens.
x10^4
+MS, 0.98-1.04min #(58-62)
653.2550
654.2589
655.2615
656.2642
1.0
0.8
0.6
0.4
0.2
0.0
650 651 652 653 654 655 656 657 658
m/z

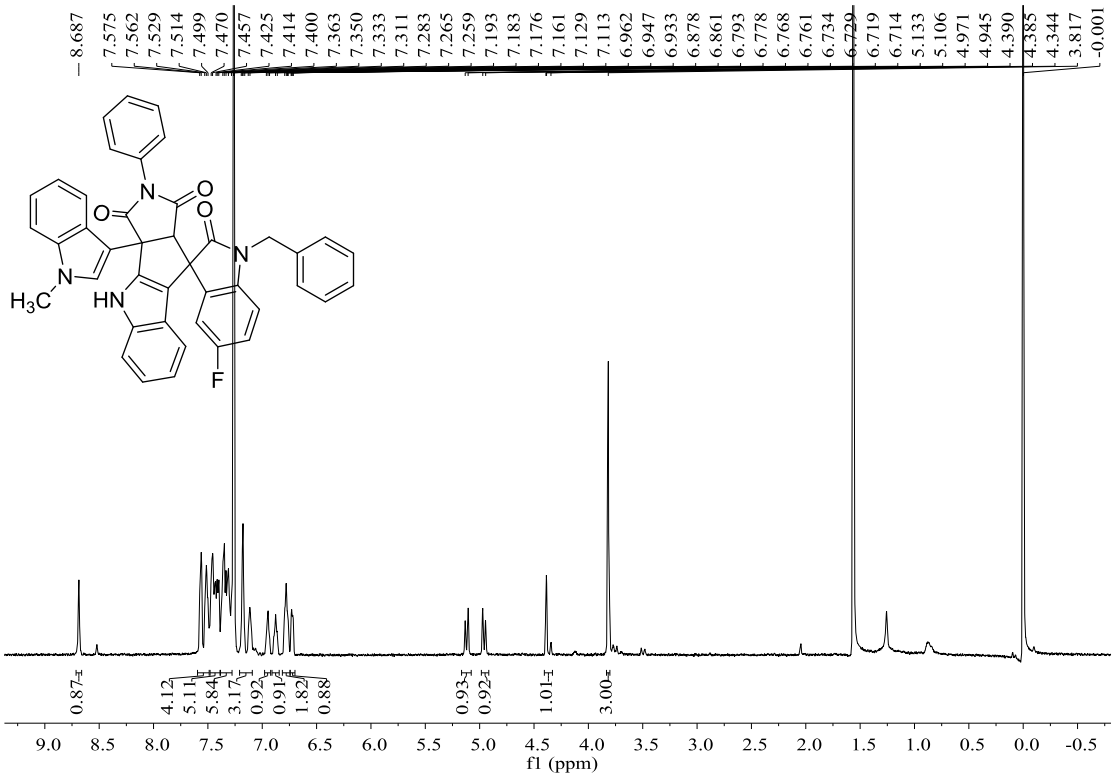
1-benzyl-5-fluoro-3a'-(1-methyl-1*H*-indol-3-yl)-2'-phenyl-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,4'*H*)-trione (5j): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 426 mg, white solid, 65%, m.p. 232-234 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 11.90 (s, 1H, NH), 7.56 (t, *J* = 8.0 Hz, 4H, ArH), 7.51-7.49 (m, 2H, ArH), 7.43 (t, *J* = 6.4 Hz, 4H, ArH), 7.36 (t, *J* = 7.6 Hz, 3H, ArH), 7.31 (t, *J* = 6.8 Hz, 1H, ArH), 7.22 (t, *J* = 7.6 Hz, 1H, ArH), 7.16-7.09 (m, 3H, ArH), 7.03 (t, *J* = 7.6 Hz, 1H, ArH), 6.87 (d, *J* = 7.6 Hz, 1H, ArH), 6.78 (d, *J* = 7.6 Hz, 1H, ArH), 6.56 (d, *J* = 8.0 Hz, 1H, ArH), 5.13 (d, *J* = 16.0 Hz, 1H, CH), 5.05 (d, *J* = 16.0 Hz, 1H, CH), 4.59 (s, 1H, CH), 3.85 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 177.7, 175.1, 172.7, 158.9 (d, *J* = 237.8 Hz), 142.4, 141.8, 138.9, 137.7, 136.5, 132.0, 131.5 (d, *J* = 7.5 Hz), 129.7, 129.4, 129.1, 128.0, 127.7, 127.2, 125.4, 122.5, 122.3, 121.4, 120.1 (d, *J* = 10.9 Hz), 119.4, 117.7, 116.0 (d, *J* = 24.6 Hz), 113.4, 112.5 (d, *J* = 25.6 Hz), 111.1 (d, *J* = 7.3 Hz), 110.7, 110.5, 67.4, 56.6, 56.5, 43.8, 32.9; IR (KBr) υ: 3052, 2923, 2867, 2737, 2521, 2368, 1871, 1767, 1732, 1609, 1532, 1485, 1445, 1369, 1234, 1178, 1123, 823, 744 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{42}H_{30}FN_4O_3$ ([M+H]$^+$): 657.2296, Found: 657.2304.

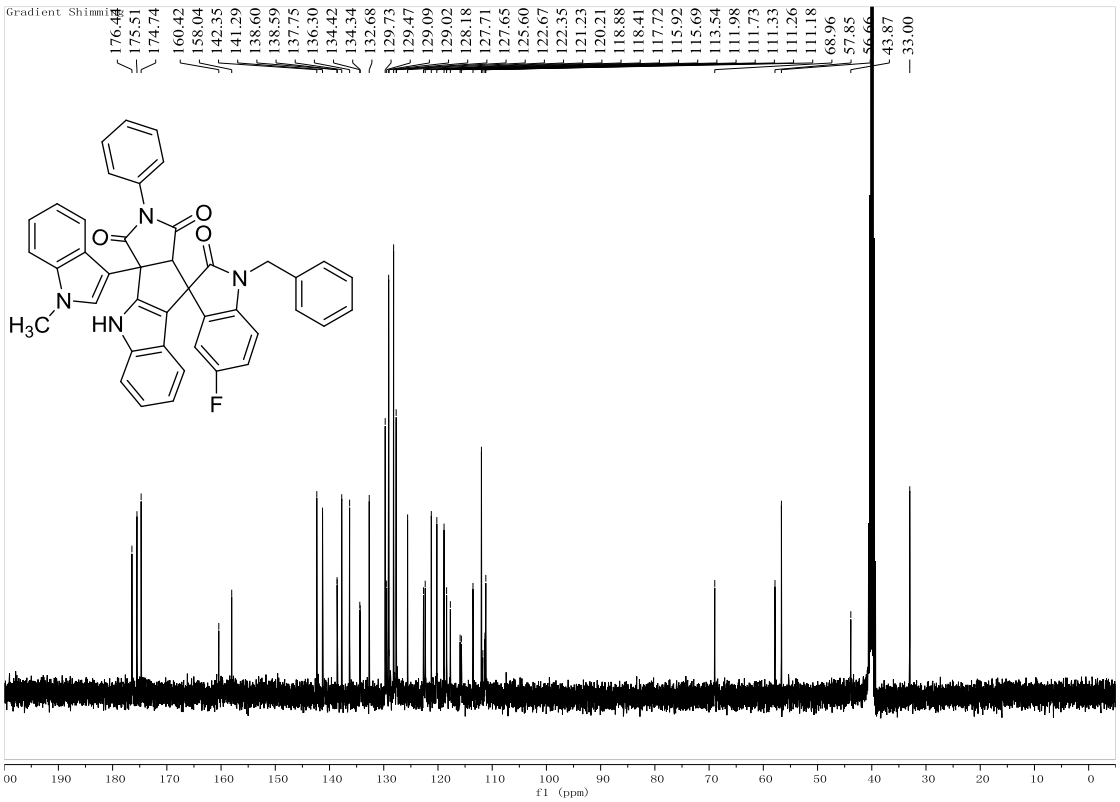


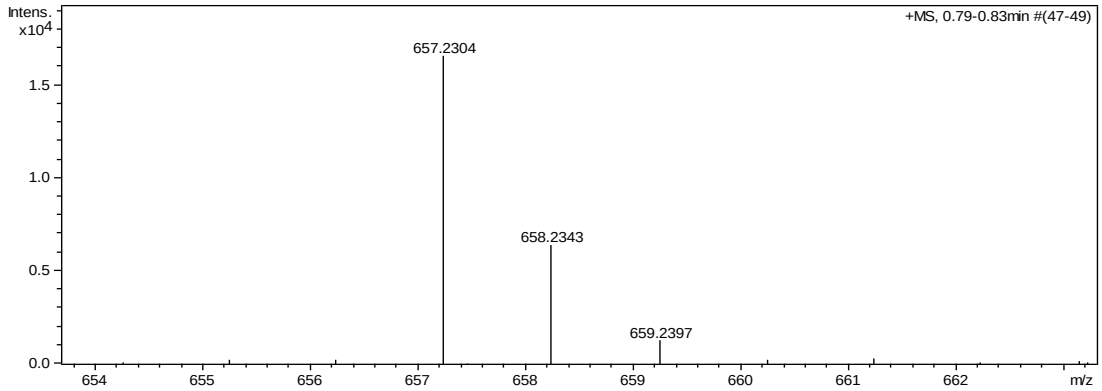

177.73
175.13
172.84
160.11
157.73
142.42
141.84
138.93
137.81
136.53
132.09
131.53
131.45
129.75
129.49
129.15
128.11
127.76
127.29
125.49
122.53
122.37
121.42
120.21
120.10
119.45
117.75
116.12
115.87
113.45
112.65
112.39
111.17
111.10
110.79
110.60
67.46
56.66
56.52
43.82
32.96
O
O
O
N
N
HN
H3C
F
f1 (ppm)


Intens.
x10^4
+MS, 0.52-0.57min #(31-34)
657.2304
658.2349
659.2392
m/z

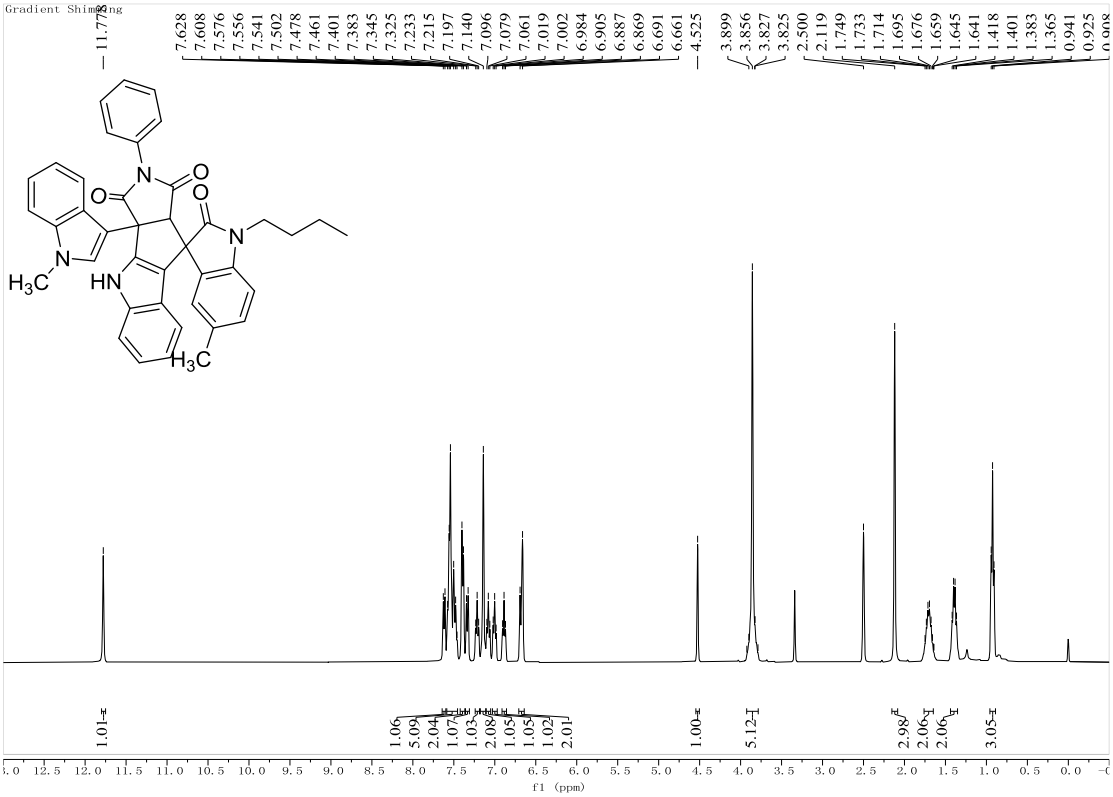
1-benzyl-5-fluoro-3a'-(1-methyl-1*H*-indol-3-yl)-2'-phenyl-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,*4*'*H*)-trione (6j): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 112 mg, white solid, 17%, m.p. 238-240 °C; ^{1}H NMR (600 MHz, $CDCl_3$) δ: 8.69 (s, 1H, NH), 7.58-7.50 (m, 4H, ArH), 7.47-7.40 (m, 5H, ArH), 7.36-7.31 (m, 6H, ArH), 7.19-7.11 (m, 3H, ArH), 6.96-6.93 (m, 1H, ArH), 6.88-6.86 (m, 1H, ArH), 6.79-6.76 (m, 2H, ArH), 6.73-6.71 (m, 1H, ArH), 5.12 (d, J = 16.0 Hz, 1H, CH), 4.96 (d, J = 16.0 Hz, 1H, CH), 4.39-4.34 (m, 1H, CH), 3.82 (s, 3H,CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 176.4, 175.4, 174.7, 159.2 (d, *J* = 238.3 Hz), 142.3, 141.2, 138.6 (d, *J* = 1.2 Hz), 137.7, 136.2, 134.4 (d, *J* = 7.8 Hz), 132.6, 129.7, 129.4, 129.0, 129.0, 128.1, 127.6, 127.6, 125.5, 122.6, 122.3, 121.2, 120.1, 118.8, 118.3, 117.7, 115.8 (d, *J* = 23.1 Hz), 113.5, 111.8 (d, *J* = 25.8 Hz), 111.3 (d, *J* = 7.4 Hz), 111.1, 68.9, 57.8, 56.6, 43.8, 32.9; IR (KBr) υ: 3045, 2922, 2852, 2731, 2532, 2368, 1885, 1765, 1722, 1614, 1535, 1477, 1445, 1367, 1248, 1178, 1122, 835, 721 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{42}H_{30}FN_4O_3$ ([M+H]$^+$): 657.2296, Found: 657.2304.

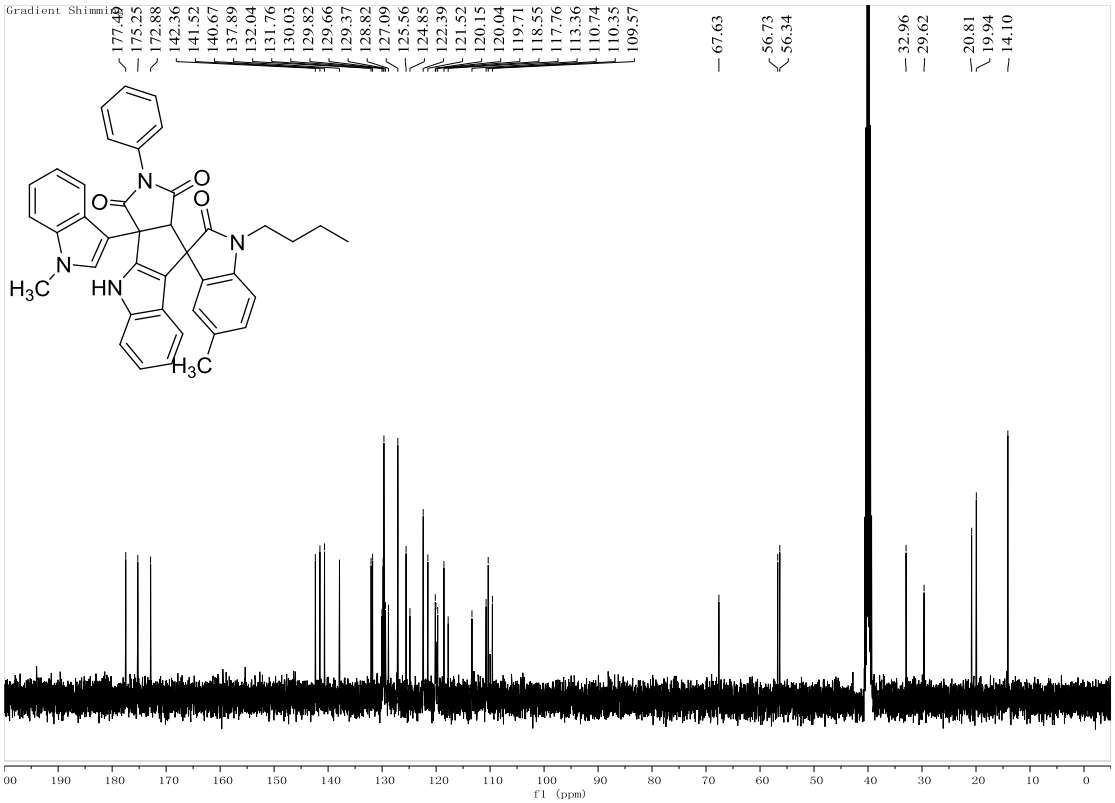


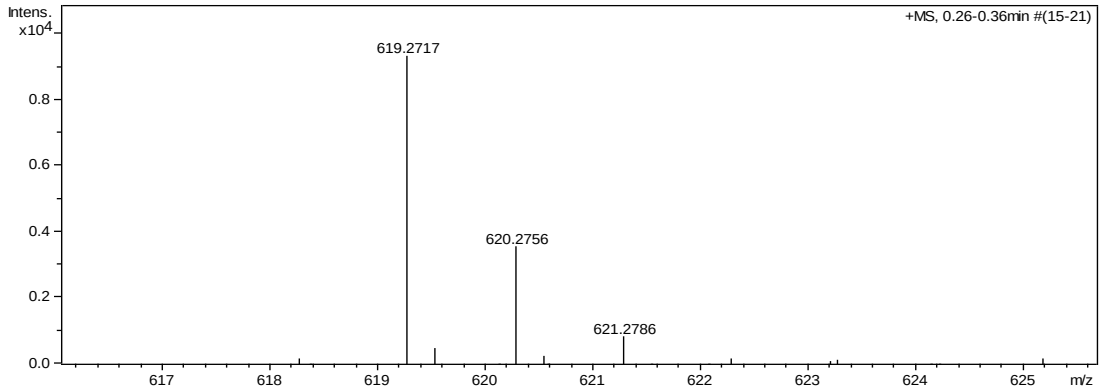

Gradient Shimming
176.44
175.51
174.74
160.42
158.04
142.35
141.29
138.60
138.59
137.75
136.30
134.42
134.34
132.68
129.73
129.47
129.09
129.02
128.18
127.71
127.65
125.60
122.67
122.35
121.23
120.21
118.88
118.41
117.72
115.92
115.69
113.54
111.98
111.73
111.33
111.26
111.18
68.96
57.85
56.66
43.87
33.00
O
O
O
N
N
HN
H3C
F
f1 (ppm)


Intens.
x10^4
+MS, 0.79-0.83min #(47-49)
657.2304
658.2343
659.2397
m/z

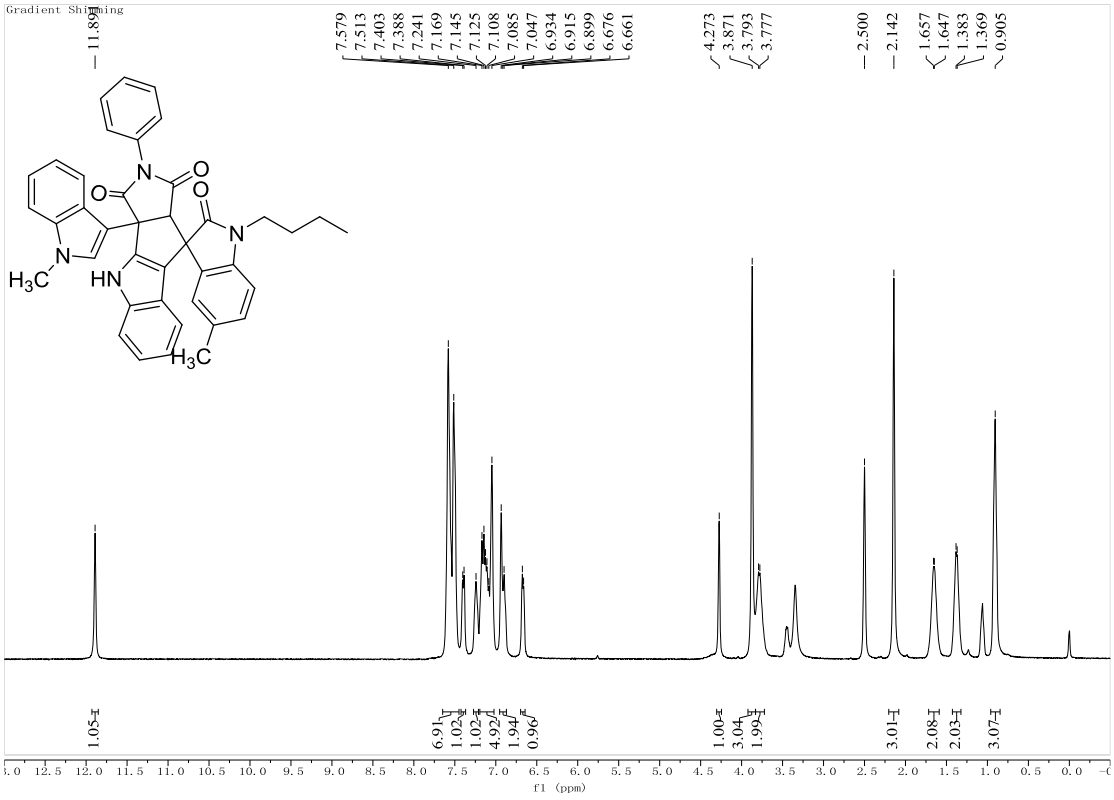
1-butyl-5-methyl-3a'-(1-methyl-1*H*-indol-3-yl)-2'-phenyl-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,4'*H*)-trione (5k): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 414 mg, white solid, 67%, m.p. 236-238 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 11.78 (s, 1H, NH), 7.62 (d, *J* = 8.0 Hz, 1H, ArH), 7.58-7.46 (m, 5H, ArH), 7.39 (d, *J* = 7.6 Hz, 2H, ArH), 7.34 (t, *J* = 8.0 Hz, 1H, ArH), 7.22 (t, *J* = 7.2 Hz, 1H, ArH), 7.14 (s, 2H, ArH), 7.08 (t, *J* = 7.2 Hz, 1H, ArH), 7.00 (t, *J* = 7.2 Hz, 1H, ArH), 6.89 (t, *J* = 7.2 Hz, 1H, ArH), 6.68 (d, *J* = 12.0 Hz, 2H, ArH), 4.53 (s, 1H, CH), 3.92-3.79 (m, 2H, CH), 3.86 (s, 3H, CH_3), 2.12 (s, 3H, CH_3), 1.75-1.64 (m, 2H, CH), 1.42-1.37 (m, 2H, CH), 0.93 (t, *J* = 6.8 Hz, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 177.4, 175.2, 172.8, 142.3, 141.5, 140.6, 137.8, 132.0, 131.7, 130.0, 129.8, 129.6, 129.3, 128.8, 127.0, 125.5, 124.8, 122.3, 121.5, 120.1, 120.0, 119.6, 118.5, 117.7, 113.3, 110.7, 110.3, 109.5, 67.6, 56.7, 56.3, 32.9, 29.6, 20.8, 19.9, 14.1; IR (KBr) υ: 3032, 2928, 2855, 2734, 2531, 2357, 1876, 1768, 1722, 1610, 1538, 1477, 1448, 1361, 1234, 1178, 1123, 830, 721 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{40}H_{35}N_4O_3$ ([M+H]$^+$): 619.2704, Found: 619.2717.

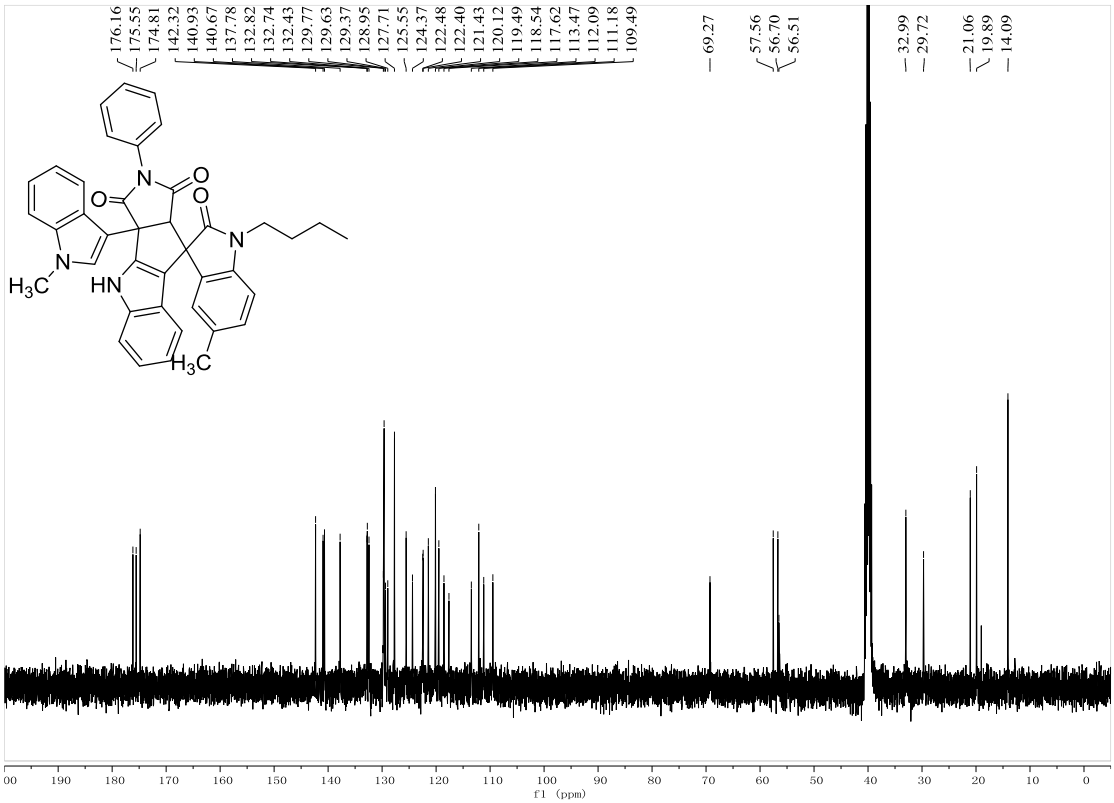


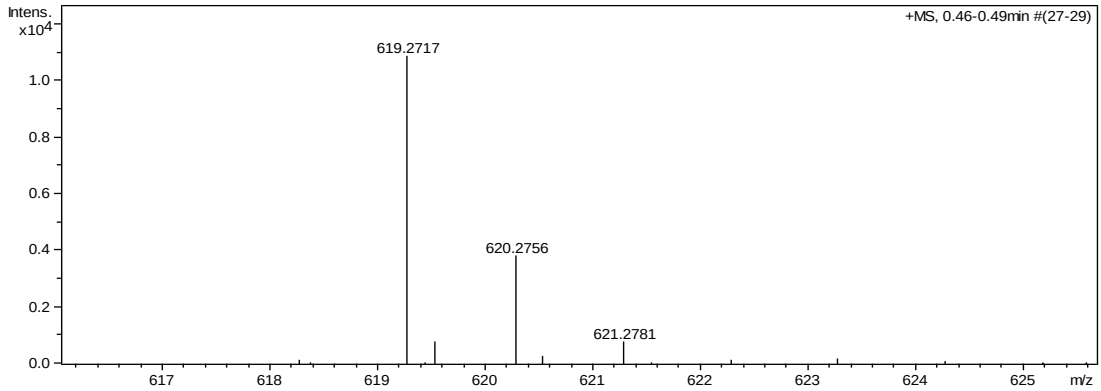

Gradient Shimming
177.49
175.25
172.88
142.36
141.52
140.67
137.89
132.04
131.76
130.03
129.82
129.66
129.37
128.82
127.09
125.56
124.85
122.39
121.52
120.15
120.04
119.71
118.55
117.76
113.36
110.74
110.35
109.57
67.63
56.73
56.34
32.96
29.62
20.81
19.94
14.10
O
O
O
N
N
HN
H3C
H3C
f1 (ppm)


Intens.
x10^4
+MS, 0.26-0.36min #(15-21)
619.2717
620.2756
621.2786
0.0
0.2
0.4
0.6
0.8
617
618
619
620
621
622
623
624
625
m/z

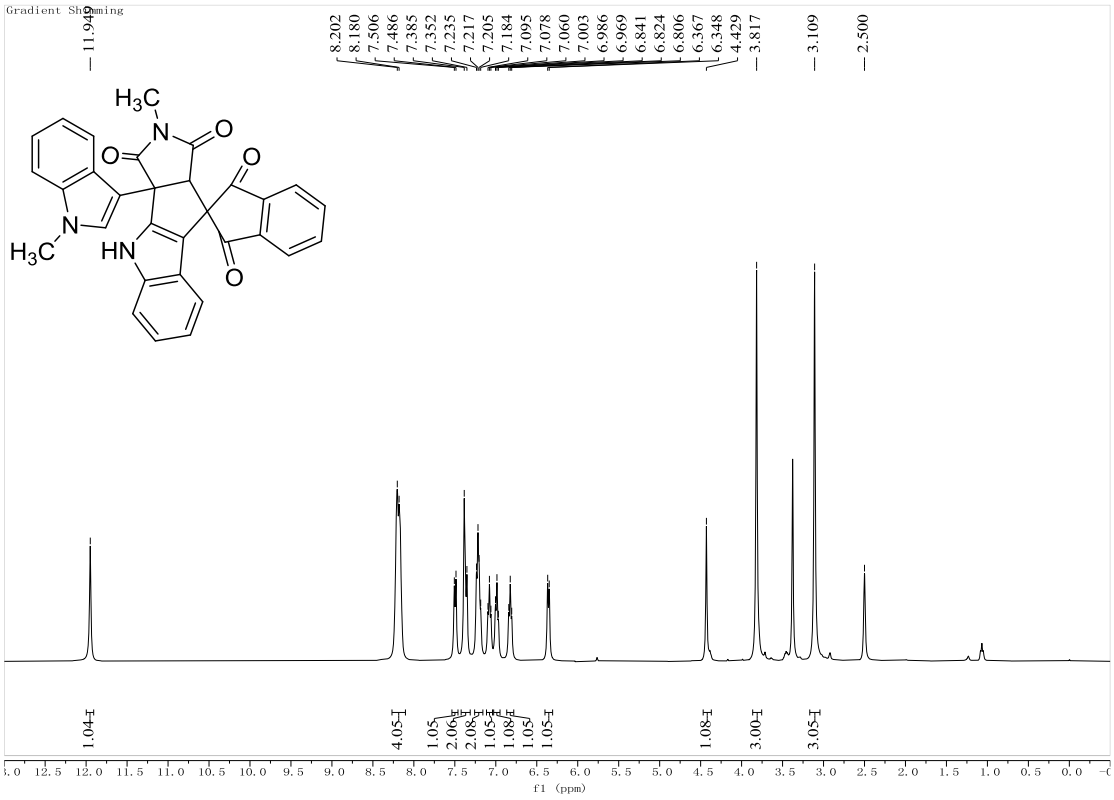
1-butyl-5-methyl-3a'-(1-methyl-1*H*-indol-3-yl)-2'-phenyl-3a',9a'-dihydrospiro[indoline-3,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1',2,3'(2'*H*,*4*'*H*)-trione (6k): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, white solid, 111 mg, white solid, 18%, m.p. 241-243 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 11.89 (s, 1H, NH), 7.55 (d, *J* = 25.2 Hz, 7H, ArH), 7.40 (d, *J* = 6.0 Hz, 1H, ArH), 7.24 (s, 1H, ArH), 7.17-7.05 (m, 5H, ArH), 6.93-6.90 (m, 2H, ArH), 6.67 (d, *J* = 6.0 Hz, 1H, ArH), 4.27 (s, 1H, CH), 3.87 (s, 3H, CH_3) , 3.79 (d, *J* = 6.4 Hz, 2H, CH), 2.14 (s, 3H, CH_3), 1.65 (d, *J* = 4.0 Hz, 1H, CH), 1.38 (d, *J* = 5.6 Hz, 1H, CH), 0.91 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 176.1, 175.5, 174.7, 142.3, 140.9, 140.6, 137.7, 132.8, 132.7, 132.4, 129.7, 129.6, 129.3, 128.9, 127.6, 125.5, 124.3, 122.4, 122.3, 121.4, 120.1, 119.4, 118.5, 117.6, 113.4, 112.0, 111.1, 109.4, 69.2, 57.5, 56.6, 56.4, 32.9, 29.7, 21.0, 19.8, 14.0; IR (KBr) υ: 3051, 2927, 2852, 2732, 2531, 2364, 1878, 1764, 1723, 1611, 1537, 1487, 1445, 1368, 1242, 1182, 1118, 827, 729 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{40}H_{35}N_4O_3$ ([M+H]$^+$): 619.2704, Found: 619.2717.

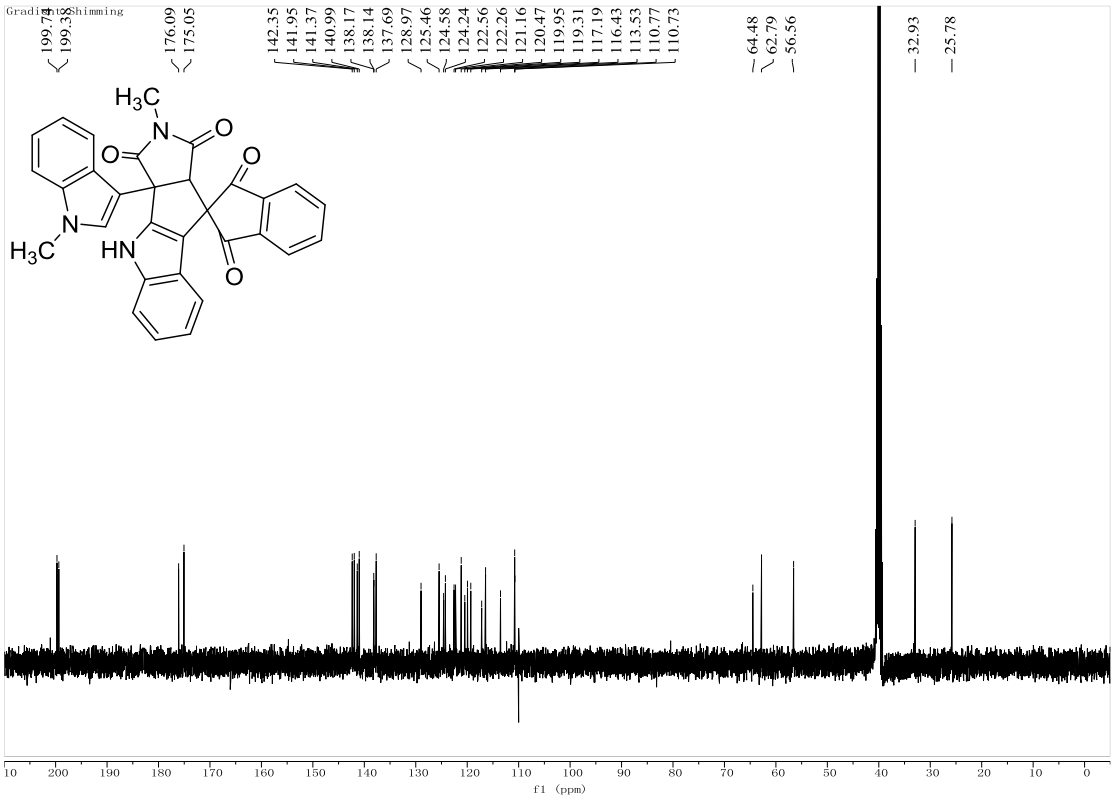


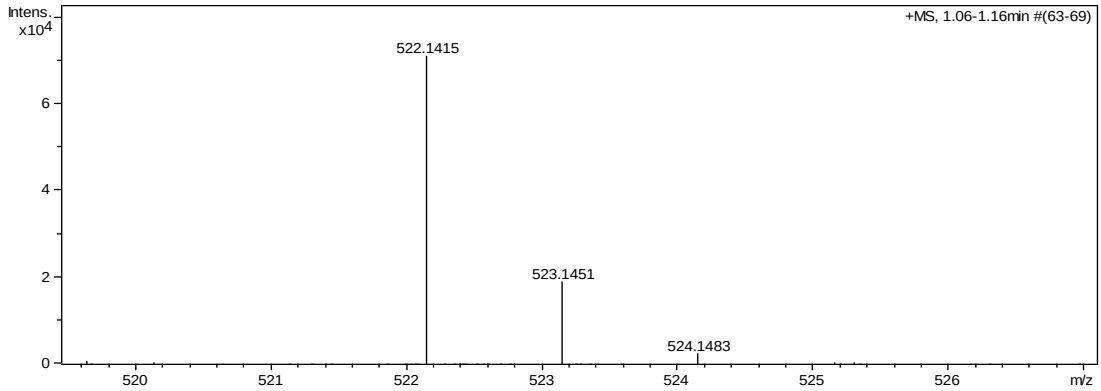

176.16
175.55
174.81
142.32
140.93
140.67
137.78
132.82
132.74
132.43
129.77
129.63
129.37
128.95
127.71
125.55
124.37
122.48
122.40
121.43
120.12
119.49
118.54
117.62
113.47
112.09
111.18
109.49
69.27
57.56
56.70
56.51
32.99
29.72
21.06
19.89
14.09
O
O
O
N
N
HN
H3C
H3C
f1 (ppm)


Intens.
x10^4
+MS, 0.46-0.49min #(27-29)
619.2717
620.2756
621.2781
m/z

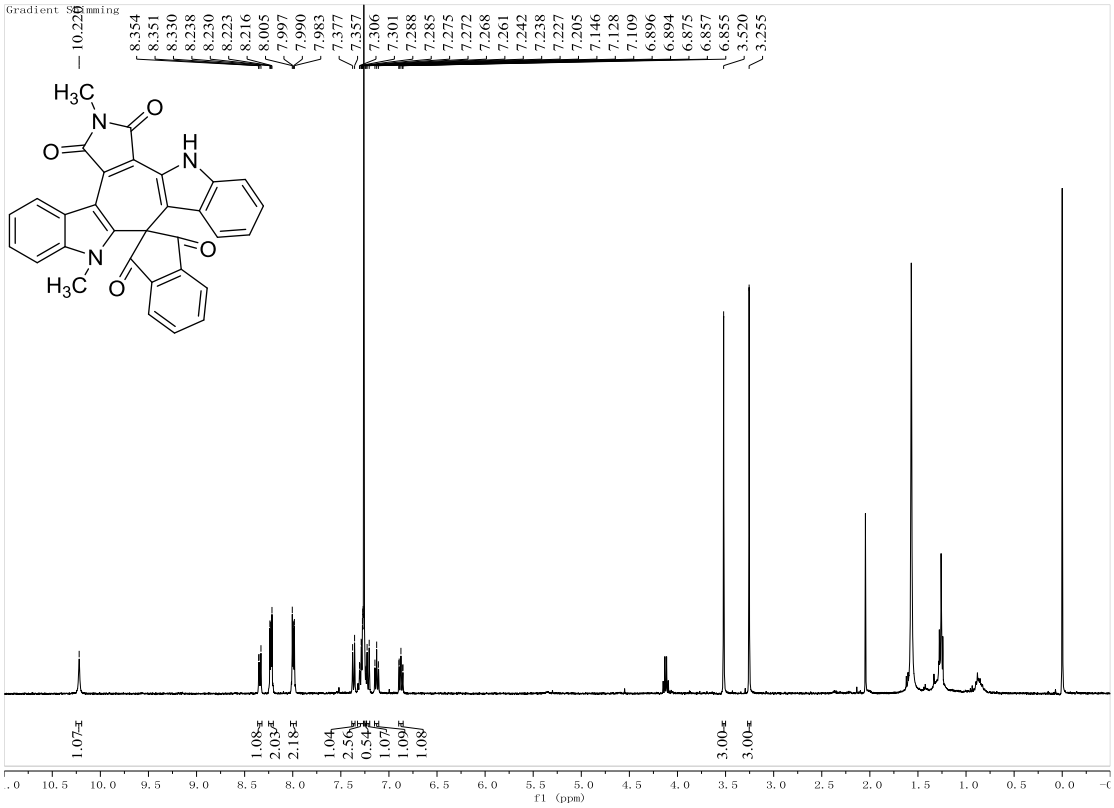
2'-methyl-3a'-(1-methyl-1*H*-indol-3-yl)-3a',9a'-dihydrospiro[indene-2,9'-pyrrolo[3',4':4,5]cyclopenta[1,2-*b*]indole]-1,1',3,3'(2'*H*,4'*H*)-tetraone (8a): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, yellow solid, 379 mg, yellow solid, 76%, m.p. 212-215 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 11.95 (s, 1H, NH), 8.19 (d, *J* = 8.8 Hz, 4H, ArH), 7.50 (d, *J* = 8.0 Hz, 1H, ArH), 7.39-7.35 (m, 2H, ArH), 7.24-7.18 (m, 2H, ArH), 7.08 (t, *J* = 6.8 Hz, 1H, ArH), 6.99 (t, *J* = 6.8 Hz, 1H, ArH), 6.82 (t, *J* = 6.8 Hz, 1H, ArH), 6.36 (d, *J* = 7.6 Hz, 1H, ArH), 4.43 (s, 1H, CH), 3.82 (s, 3H, CH_3), 3.11 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 199.7, 199.3, 176.0, 175.0, 142.3, 141.9, 141.3, 140.9, 138.2, 138.1, 137.6, 128.9, 125.4, 124.5, 124.2, 122.5, 122.2, 121.1, 120.4, 119.9, 119.2, 117.1, 116.4, 113.5, 110.7, 110.7, 64.4, 62.7, 56.5, 32.9, 25.7; IR (KBr) υ: 3056, 2933, 2823, 2675, 1929, 1709, 1600, 1548, 1473, 1427, 1396, 1185, 1340, 1248, 1133, 1049, 982, 926, 857, 760 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{31}H_{21}NaN_3O_4$ ([M+Na]$^{+}$): 522.1424, Found: 522.1415.

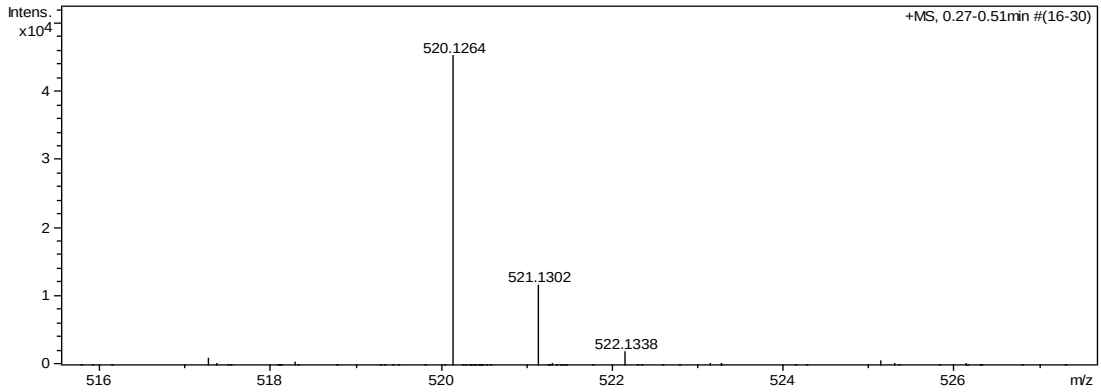



Gradient Shimming
199.74
199.38
176.09
175.05
142.35
141.95
141.37
140.99
138.17
138.14
137.69
128.97
125.46
124.58
124.24
122.56
122.26
121.16
120.47
119.95
119.31
117.19
116.43
113.53
110.77
110.73
64.48
62.79
56.56
32.93
25.78
H3C
N
O
O
O
N
H3C
HN
O
f1 (ppm)

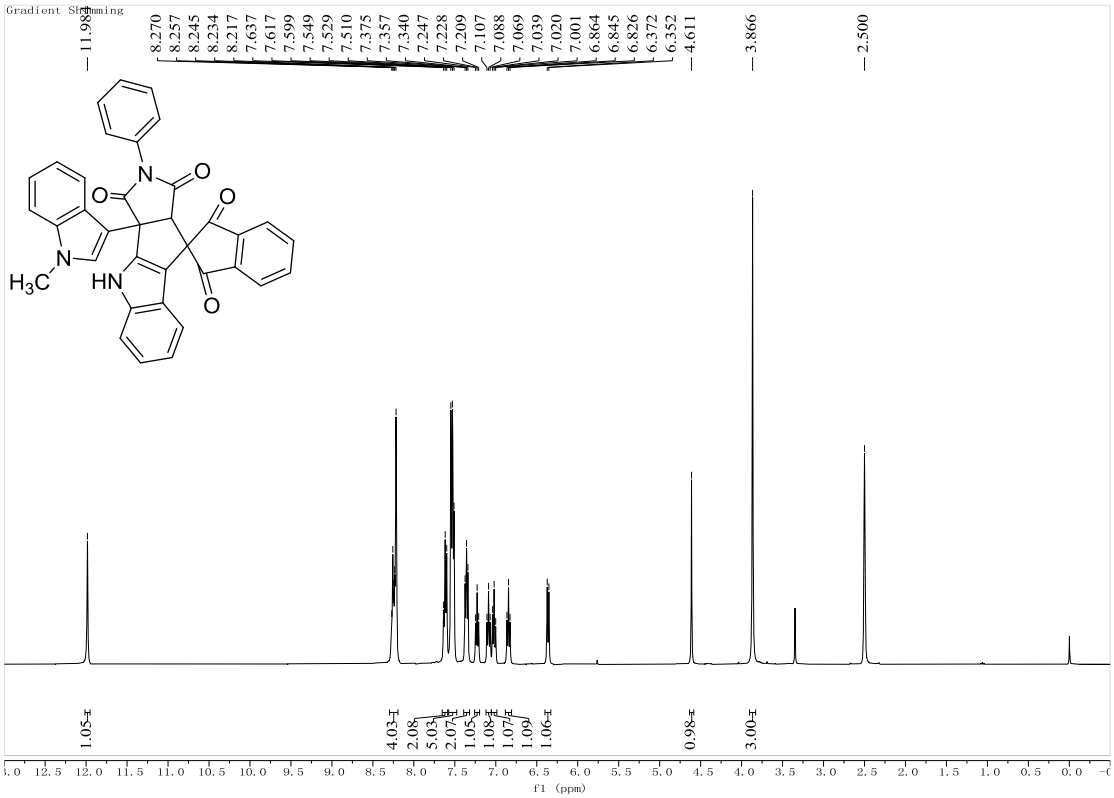

Intens.
x10^4
+MS, 1.06-1.16min #(63-69)
522.1415
523.1451
524.1483
m/z

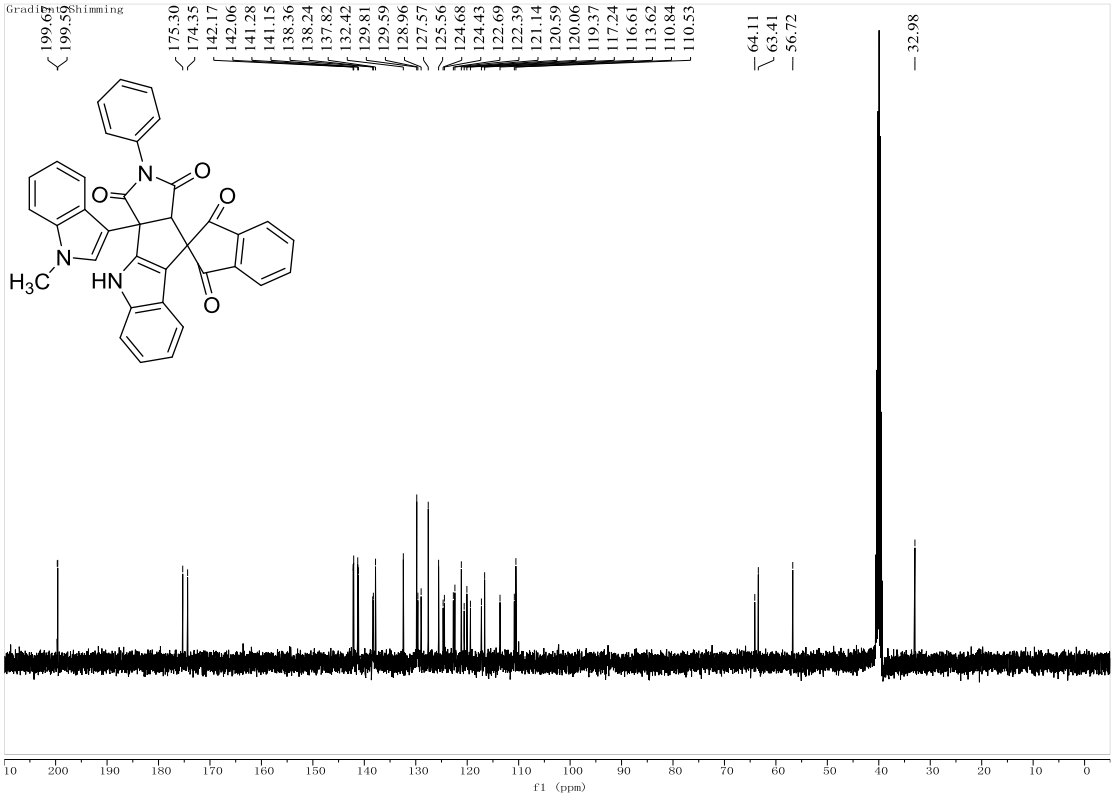
2',8'-dimethyl-8',14'-dihydro-1'*H*-spiro[indene-2,9'-pyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole]-1,1',3,3'(2'*H*)-tetraone (9a): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, red solid, 90 mg, red solid, 18%, m.p. 238-240 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 10.22 (s, 1H, NH), 8.35-8.33 (m, 1H, ArH), 8.23 (q, *J* = 2.8 Hz, 2H, ArH), 8.00 (q, *J* = 2.8 Hz, 2H, ArH), 7.37 (q, *J* = 8.0 Hz, 1H, ArH), 7.31-7.24 (m, 3H, ArH), 7.22 (d, *J* = 8.8 Hz, 1H, ArH), 7.13 (t, *J* = 7.2 Hz, 1H, ArH), 6.90-6.85 (m, 1H, ArH), 3.52 (s, 3H, CH_3), 3.26 (s, 3H, CH_3); IR (KBr) υ: 3051, 2939, 2849, 2352, 1751, 1694, 1612, 1542, 1485, 1394, 1384, 1246, 1321, 1168, 1098, 1060, 1016, 998, 852, 798, 736 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{31}H_{19}NaN_3O_4$ ([M+Na]$^+$): 520.1268, Found: 520.1264.

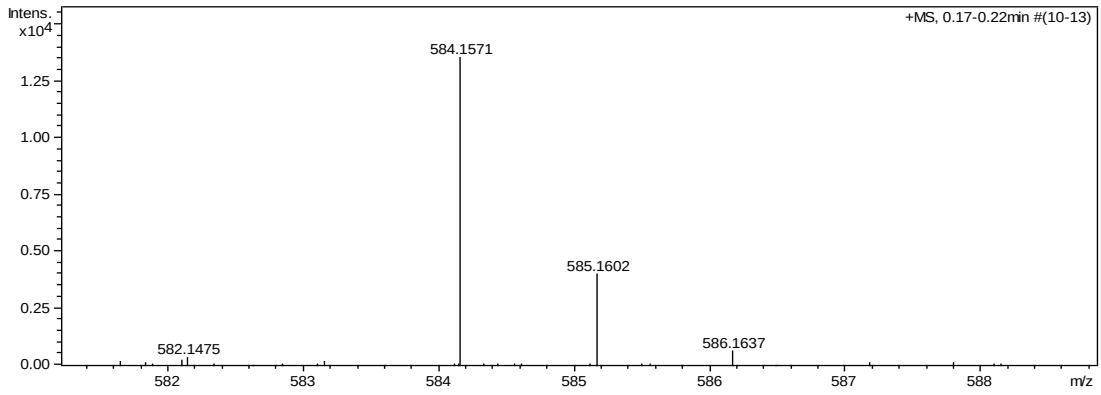




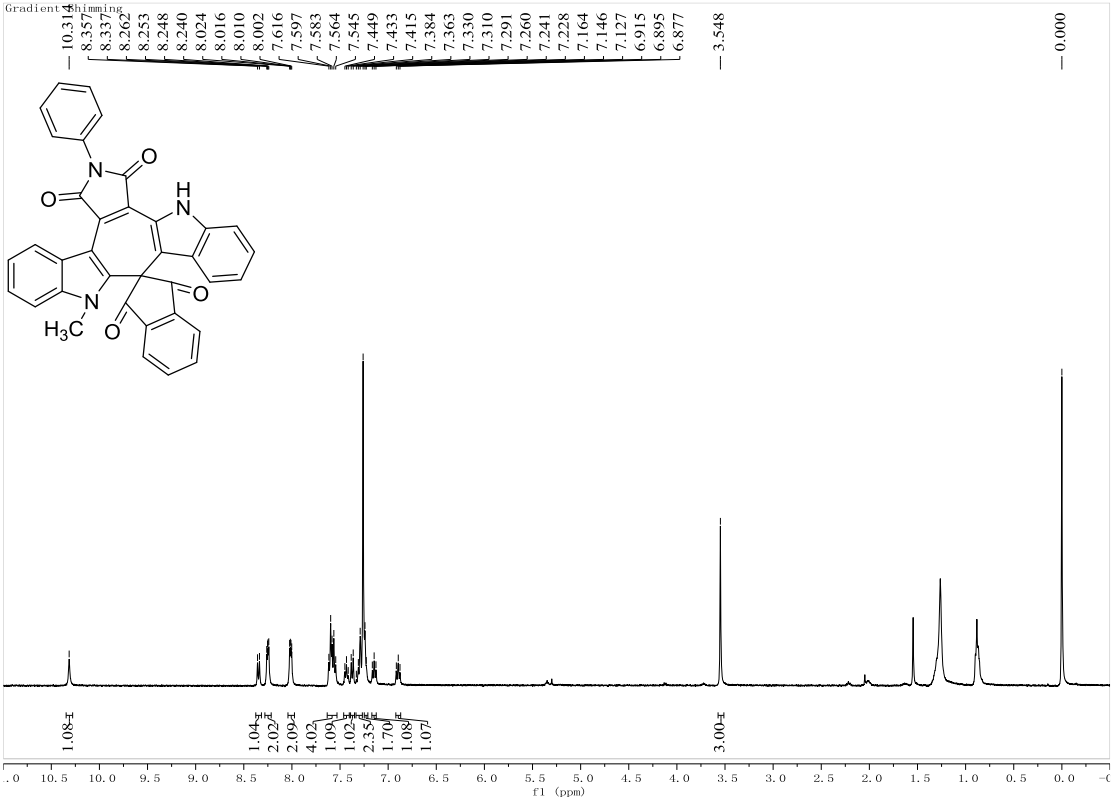
3a'-(1-methyl-1*H*-indol-3-yl)-2'-phenyl-3a',9a'-dihydrospiro[indene-2,9'-pyrrolo[3',4':4,5]cyclopenta[1,2-*b*]indole]-1,1',3,3'(2'*H*,4'*H*)-tetraone (8b): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, yellow solid, 438 mg, yellow solid, 78%, m.p. 218-221 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 11.98 (s, 1H, NH), 8.27-8.22 (m, 4H, ArH), 7.64-7.60 (m, 2H, ArH), 7.53 (t, *J* = 8.0 Hz, 5H, ArH), 7.36 (t, *J* = 6.8 Hz, 2H, ArH), 7.23 (t, *J* = 7.6 Hz, 1H, ArH), 7.09 (t, *J* = 7.6 Hz, 1H, ArH), 7.02 (t, *J* = 7.6 Hz, 1H, ArH), 6.85 (t, *J* = 7.6 Hz, 1H, ArH), 6.36 (d, *J* = 8.0 Hz, 1H, ArH), 4.61 (s, 1H, CH), 3.87 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 199.6, 199.5, 175.2, 174.3, 142.1, 142.0, 141.2, 141.1, 138.3, 138.2, 137.8, 132.4, 129.7, 129.5, 128.9, 127.5, 125.5, 124.6, 124.4, 122.6, 122.3, 121.1, 120.5, 120.0, 119.3, 117.2, 116.6, 113.6, 110.8, 110.5, 64.0, 63.4, 56.7, 32.9; IR (KBr) υ: 3055, 2931, 2825, 2676, 1934, 1711, 1611, 1551, 1456, 1432, 1388, 1184, 1348, 1250, 1132, 1041, 972, 927, 843, 756 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{36}H_{23}NaN_3O_4$ ([M+Na]$^+$): 584.1581, Found: 584.1571.

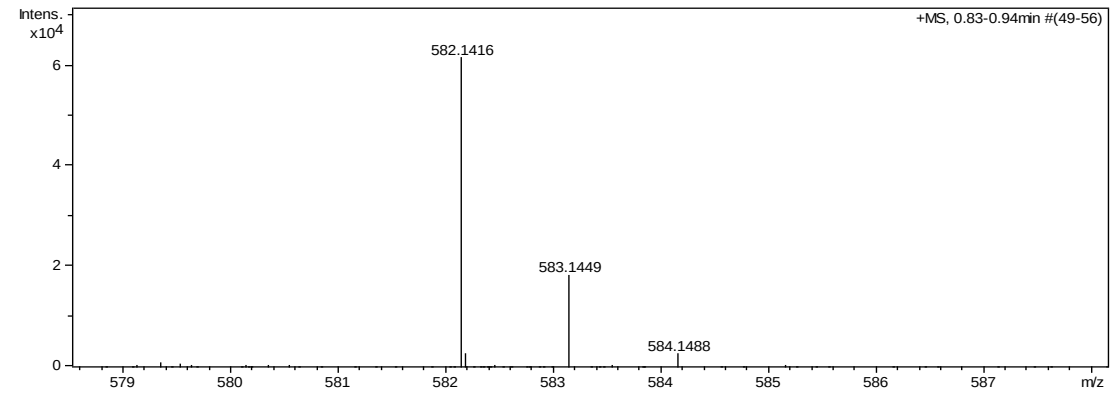



Gradient Shimming
199.67
199.59
175.30
174.35
142.17
142.06
141.28
141.15
138.36
138.24
137.82
132.42
129.81
129.59
128.96
127.57
125.56
124.68
124.43
122.69
122.39
121.14
120.59
120.06
119.37
117.24
116.61
113.62
110.84
110.53
64.11
63.41
56.72
32.98
O
N
O
O
O
N
HN
H3C
f1 (ppm)

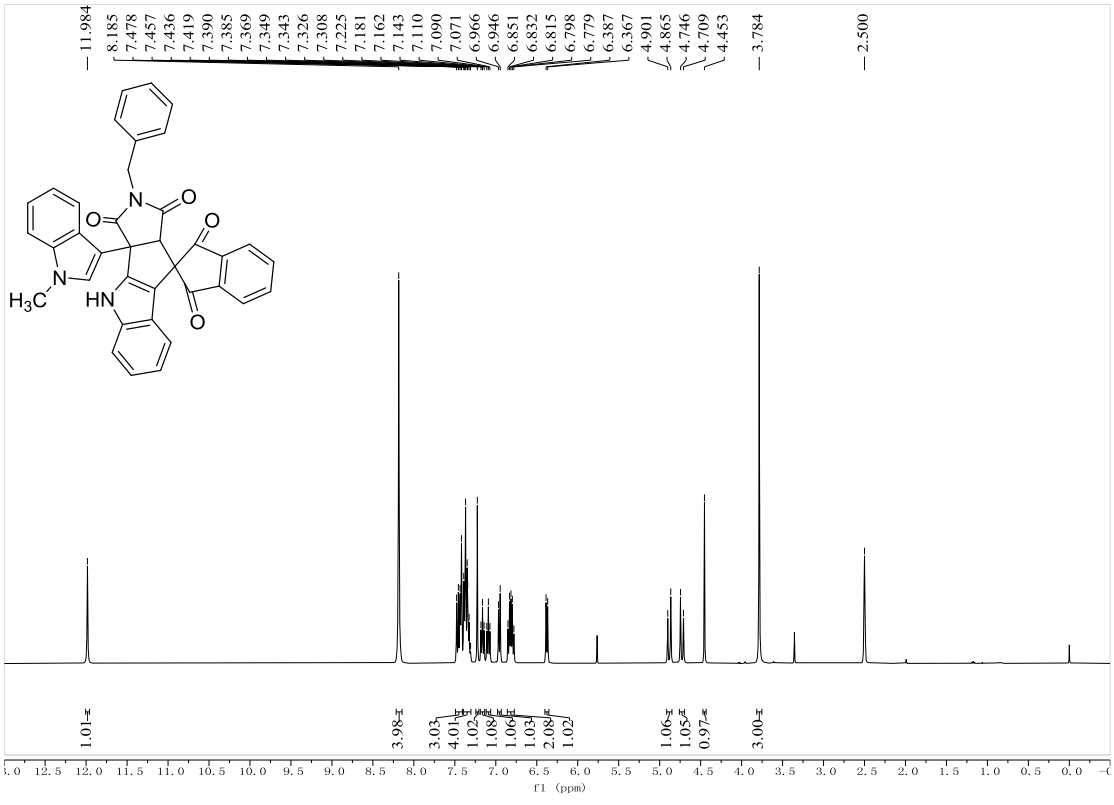

Intens.
x10^4
+MS, 0.17-0.22min #(10-13)
584.1571
585.1602
582.1475
586.1637
m/z

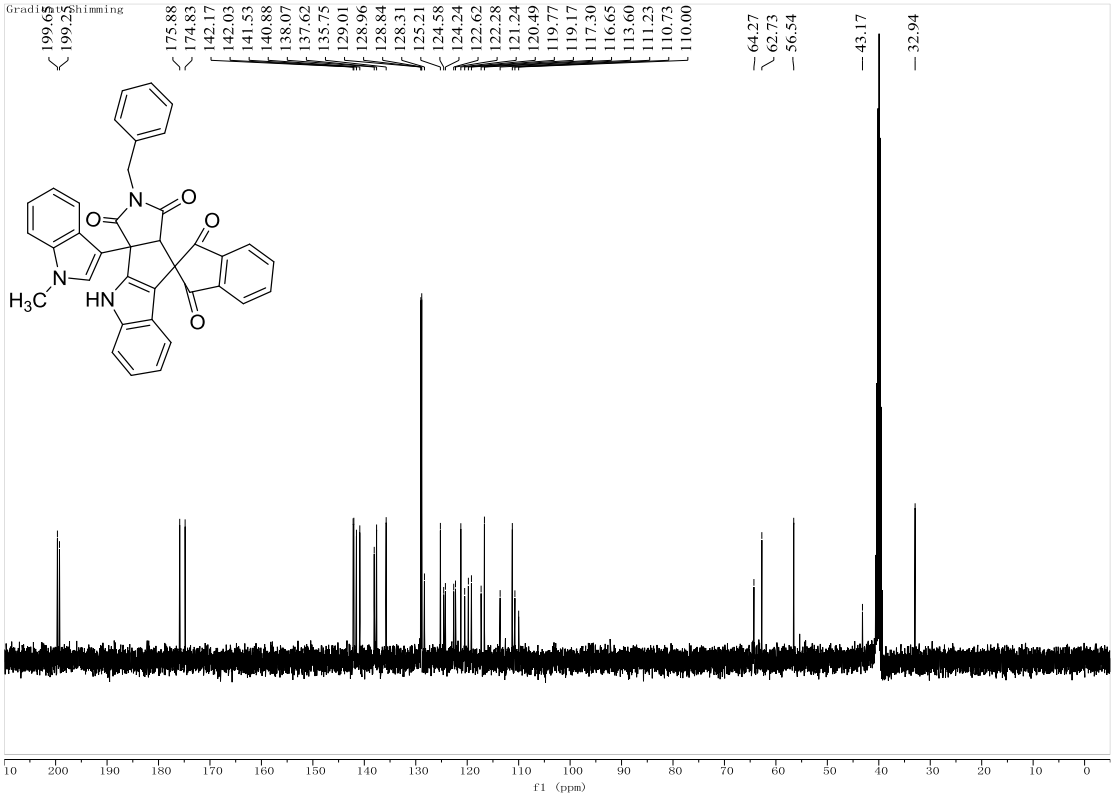
8'-methyl-2'-phenyl-8',14'-dihydro-1'*H*-spiro[indene-2,9'-pyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole]-1,1',3,3'(2'*H*)-tetraone (9b): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, red solid, 95 mg, red solid, 17%, m.p. 235-238 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 10.31 (s, 1H, NH), 8.35 (d, J = 8.0 Hz, 1H, ArH), 8.25 (dd, J_1 = 5.2 Hz, J_2 = 3.2 Hz, 2H, ArH), 8.01 (dd, J_1 = 5.6 Hz, J_2 = 3.2 Hz, 2H, ArH), 7.62-7.55 (m, 4H, ArH), 7.45-7.42 (m, 1H, ArH), 7.37 (d, J = 8.4 Hz, 1H, ArH), 7.33-7.23 (m, 4H, ArH), 7.15 (d, J = 7.6 Hz, 1H, ArH), 6.90 (d, J = 8.0 Hz, 1H, ArH), 3.55 (s, 3H, CH_3); IR (KBr) υ: 3055, 2941, 2845, 2348, 1745, 1685, 1622, 1535, 1478, 1384, 1367, 1250, 1325, 1178, 1088, 1056, 1022, 989, 845, 775, 745 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{36}H_{21}NaN_3O_4$ ([M+Na]$^{+}$): 5582.1424, Found: 582.1416.

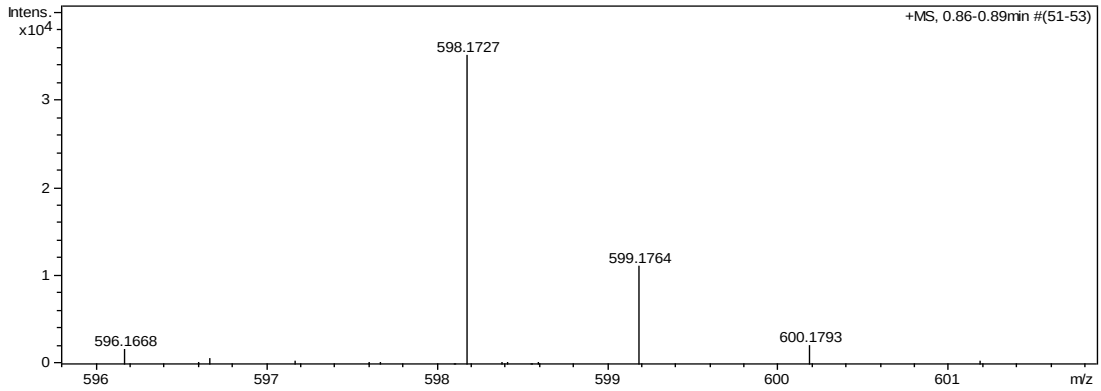




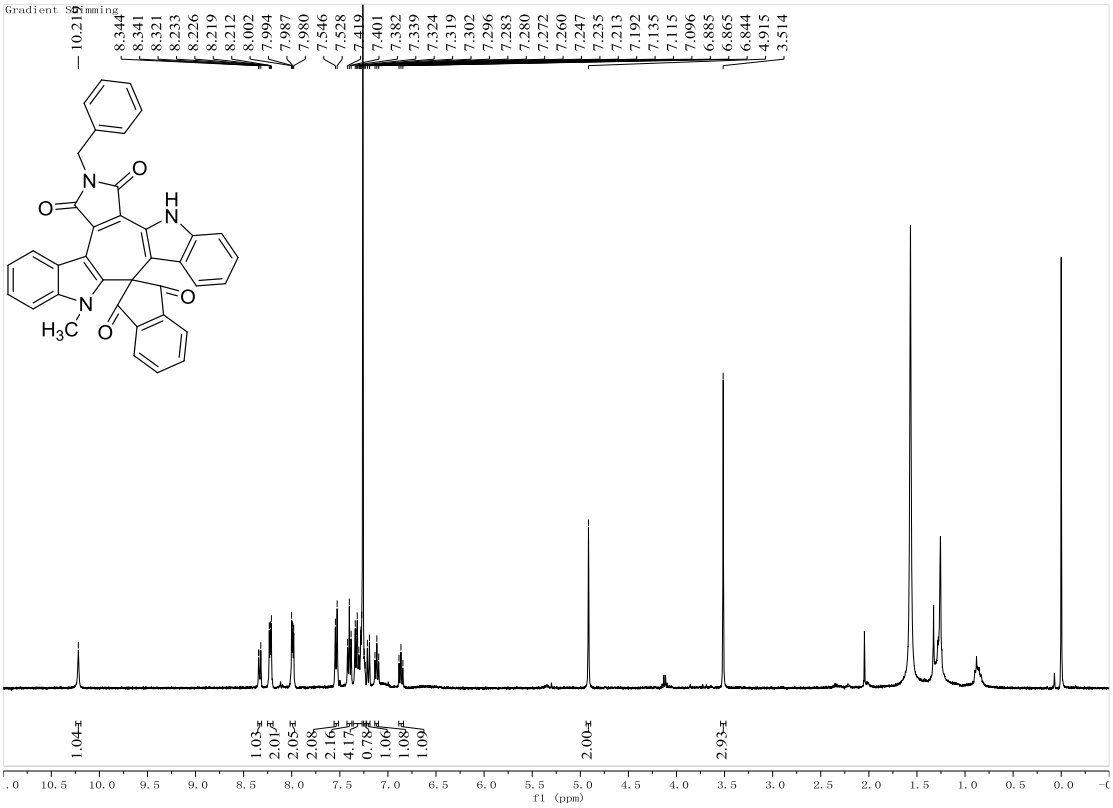
2'-benzyl-3a'-(1-methyl-1*H*-indol-3-yl)-3a',9a'-dihydrospiro[indene-2,9'-pyrrolo[3',*4*':4,5]cyclopenta[1,2-*b*]indole]-1,1',3,3'(2'*H*,4'*H*)-tetraone (8c): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, yellow solid, 431 mg, yellow solid, 75%, m.p. 222-224 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 11.98 (s, 1H, NH), 8.19 (s, 4H, ArH), 7.48-7.42 (m, 3H, ArH), 7.39-7.31 (m, 4H, ArH), 7.23 (s, 1H, ArH), 7.16 (t, *J* = 7.6 Hz, 1H, ArH), 7.09 (t, *J* = 7.6 Hz, 1H, ArH), 6.96 (t, *J* = 8.0 Hz, 1H, ArH), 6.85-6.78 (m, 2H, ArH), 6.38 (d, *J* = 7.6 Hz, 1H, ArH), 4.88 (d, *J* = 14.4 Hz, 1H, CH), 4.73 (d, *J* = 14.4 Hz, 1H, CH), 4.45 (s, 1H, CH), 3.78 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 199.6, 199.2, 175.8, 174.8, 142.1, 142.0, 141.5, 140.8, 138.0, 137.6, 135.7, 129.0, 128.9, 128.8, 128.2, 125.1, 124.5, 124.2, 122.6, 122.2, 121.2, 120.4, 119.7, 119.1, 117.2, 116.6, 113.5, 111.2, 110.7, 109.9, 64.2, 62.7, 56.5, 43.1, 32.9; IR (KBr) υ: 3055, 2931, 2831, 2678, 1931, 1711, 1615, 1550, 1468, 1431, 1386, 1184, 1343, 1250, 1132, 1048, 979, 931, 854, 756 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{37}H_{25}NaN_3O_4$ ([M+Na]$^+$): 598.1737, Found: 598.1727.

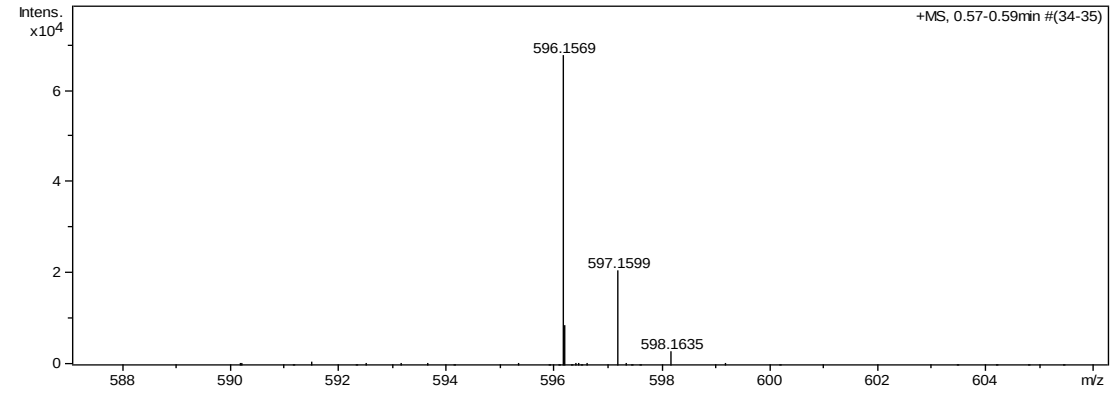



Gradient Shimming
199.65
199.25
175.88
174.83
142.17
142.03
141.53
140.88
138.07
137.62
135.75
129.01
128.96
128.84
128.31
125.21
124.58
124.24
122.62
122.28
121.24
120.49
119.77
119.17
117.30
116.65
113.60
111.23
110.73
110.00
64.27
62.73
56.54
43.17
32.94
O
O
O
O
N
N
HN
H3C
f1 (ppm)

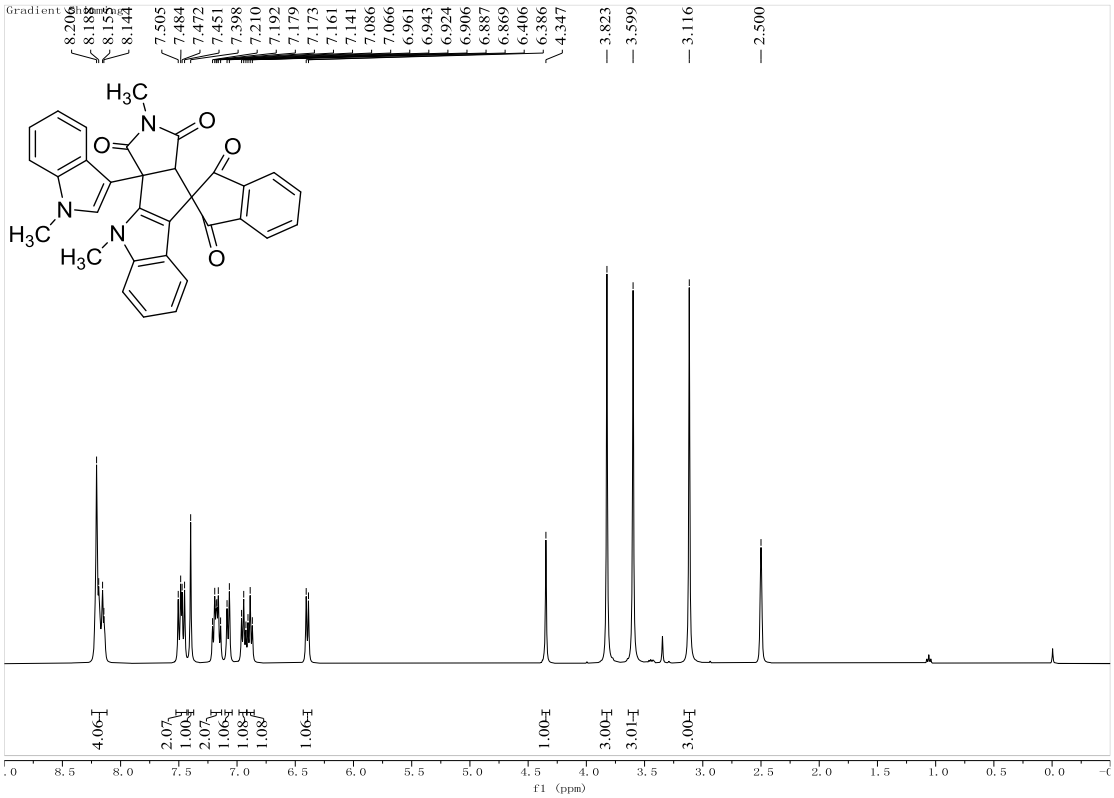

Intens.
x10^4
+MS, 0.86-0.89min #(51-53)
598.1727
599.1764
596.1668
600.1793
m/z

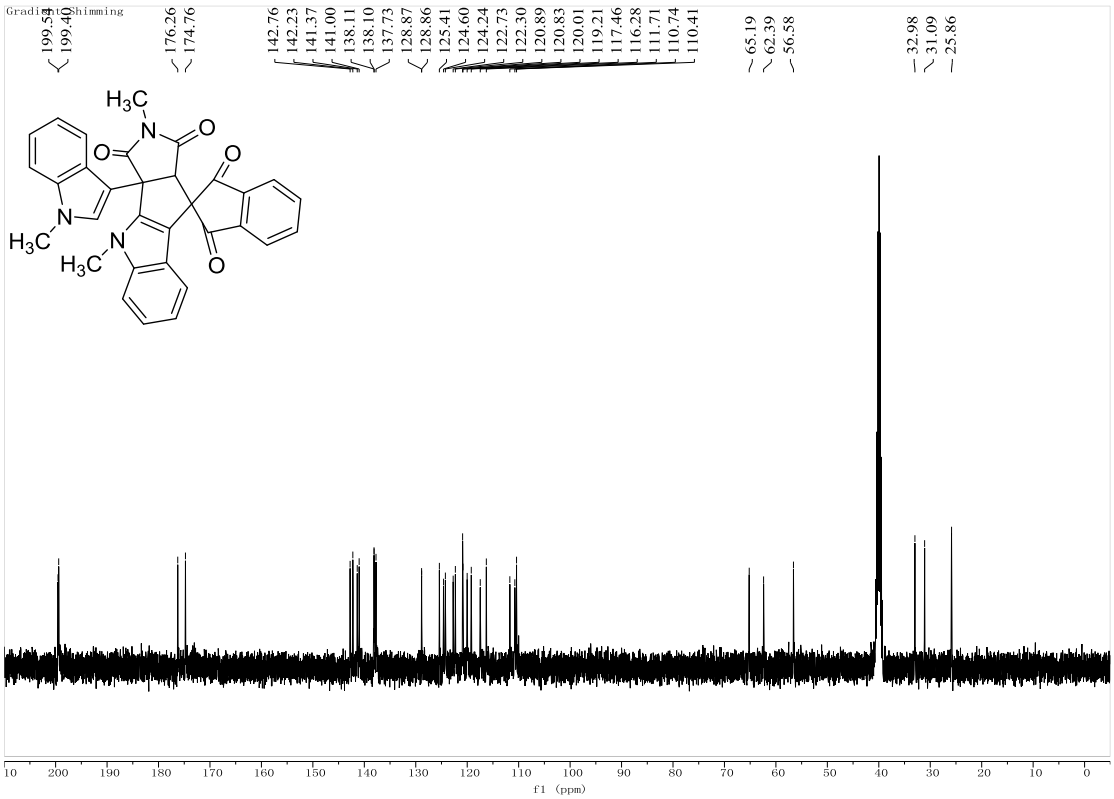
2'-benzyl-8'-methyl-8',14'-dihydro-1'*H*-spiro[indene-2,9'-pyrrolo[3',4':6,7]cyclohepta[1,2-*b*:4,5-*b*']diindole]-1,1',3,3'(2'*H*)-tetraone (9c): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, red solid, 120 mg, red solid, 21%, m.p. 240-242 ^{o}C; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 10.22 (s, 1H, NH), 8.34-8.32 (m, 1H, ArH), 8.22 (dd, J_1 = 6.4 Hz, J_2 = 2.8 Hz, 2H, ArH), 7.99 (dd, J_1 = 6.4 Hz, J_2 = 2.8 Hz, 2H, ArH), 7.54 (d, J = 7.2 Hz, 2H, ArH), 7.40 (t, J = 7.6 Hz, 2H, ArH), 7.34-7.24 (m, 5H, ArH), 7.20 (d, J = 8.4 Hz, 1H, ArH), 7.12 (t, J = 8.0 Hz, 1H, ArH), 6.86 (d, J = 8.0 Hz, 1H, ArH), 4.92 (s, 2H, CH), 3.51 (s, 3H, CH_3); IR (KBr) υ: 3045, 2941, 2838, 2348, 1748, 1686, 1622, 1548, 1478, 1389, 1378, 1251, 1323, 1178, 1088, 1065, 1021, 989, 851, 788, 745 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{37}H_{23}NaN_3O_4$ ([M+Na]$^{+}$): 596.1581, Found: 596.1569.

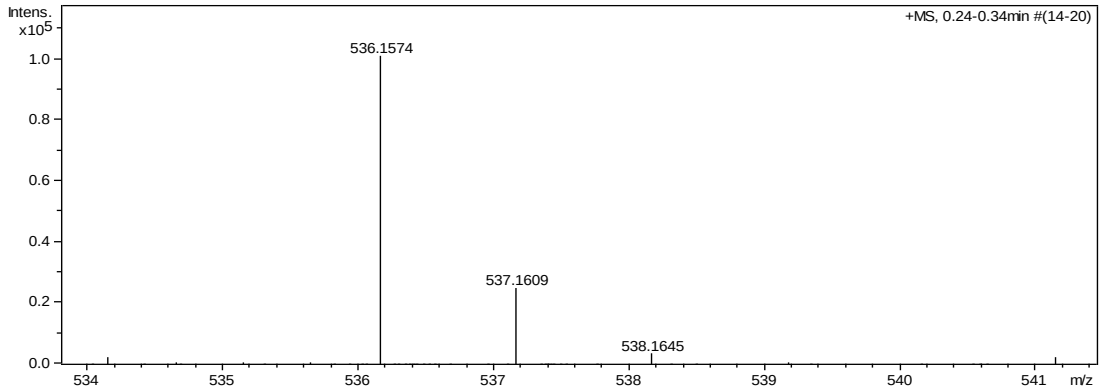




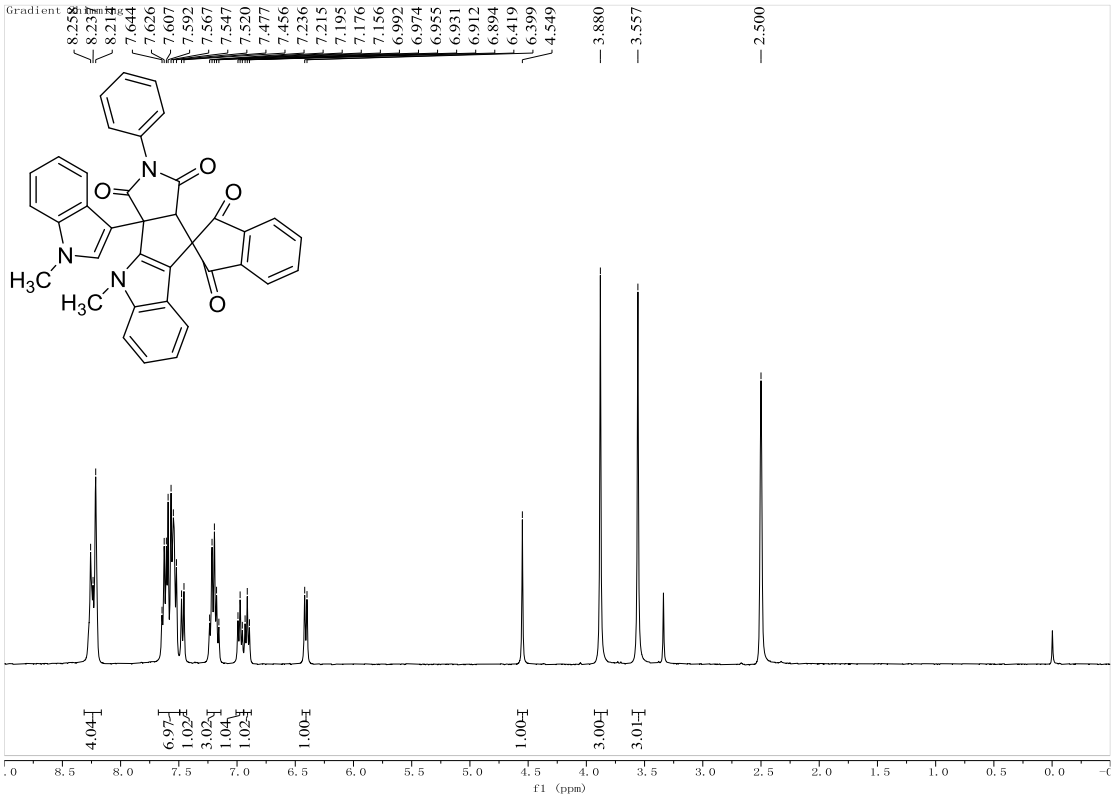
2',4'-dimethyl-3a'-(1-methyl-1*H*-indol-3-yl)-3a',9a'-dihydrospiro[indene-2,9'-pyrrolo[3',4':4,5]cyclopenta[1,2-*b*]indole]-1,1',3,3'(2'*H*,4'*H*)-tetraone (8d): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, yellow solid, 482 mg, yellow solid, 94%, m.p. 224-226 ^{o}C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 8.21-8.14 (m, 4H, ArH), 7.48 (dd, J_1 = 13.2 Hz, J_2 = 8.4 Hz, 2H, ArH), 7.40 (s, 1H, ArH) , 7.21-7.14 (m, 2H, ArH), 7.08 (d, J = 8.0 Hz, 1H, ArH), 6.94 (t, J = 7.2 Hz, 1H, ArH), 6.89 (t, J = 7.2 Hz, 1H, ArH), 6.40 (d, J = 8.0 Hz, 1H, ArH), 4.35 (s, 1H, CH), 3.82 (s, 3H, CH_3), 3.60 (s, 3H, CH_3), 3.12 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 199.5, 199.3, 176.2, 174.7, 142.7, 142.2, 141.3, 140.9, 138.1, 138.0, 137.7, 128.8, 128.7, 125.3, 124.5, 124.2, 122.7, 122.2, 120.9, 120.8, 119.9, 119.1, 117.4, 116.2, 111.6, 110.7, 110.4, 65.1, 62.3, 56.5, 32.9, 31.0, 25.8; IR (KBr) υ: 3048, 2924, 2819, 2642, 1931, 1711, 1611, 1551, 1478, 1431, 1389, 1179, 1350, 1250, 1132, 1051, 978, 924, 855, 756 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{32}H_{23}NaN_3O_4$ ([M+Na]$^{+}$): 536.1581, Found: 536.1574.

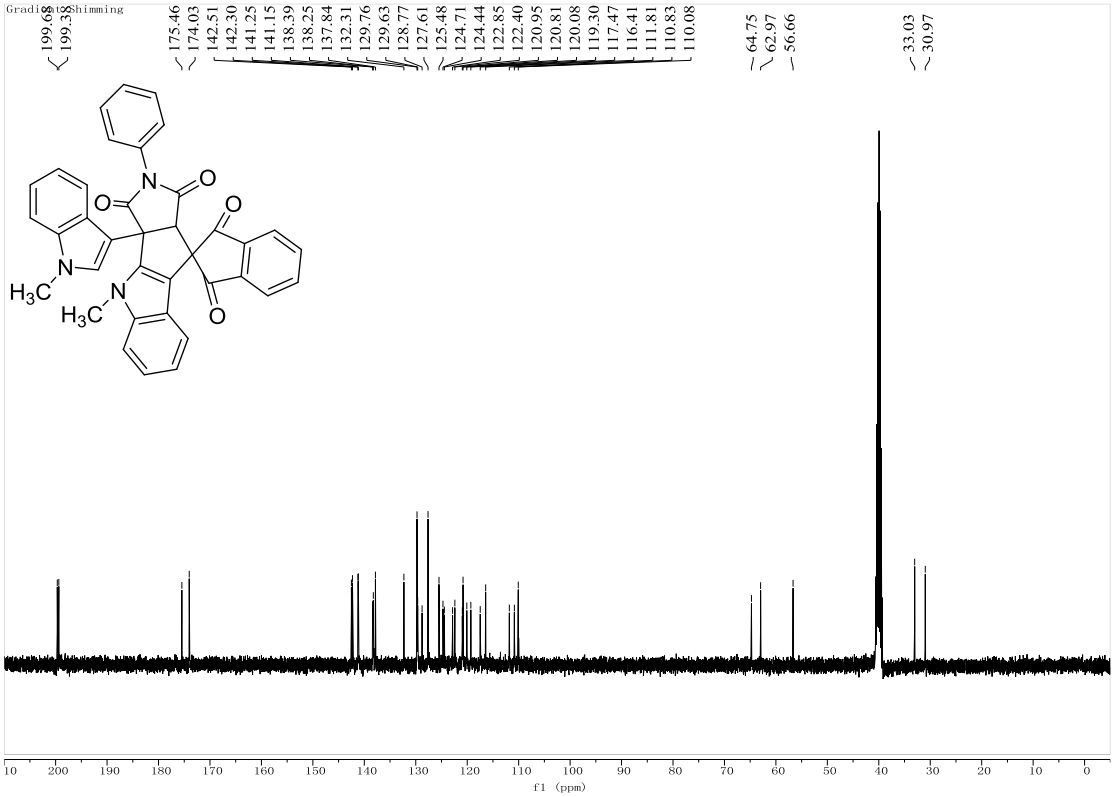


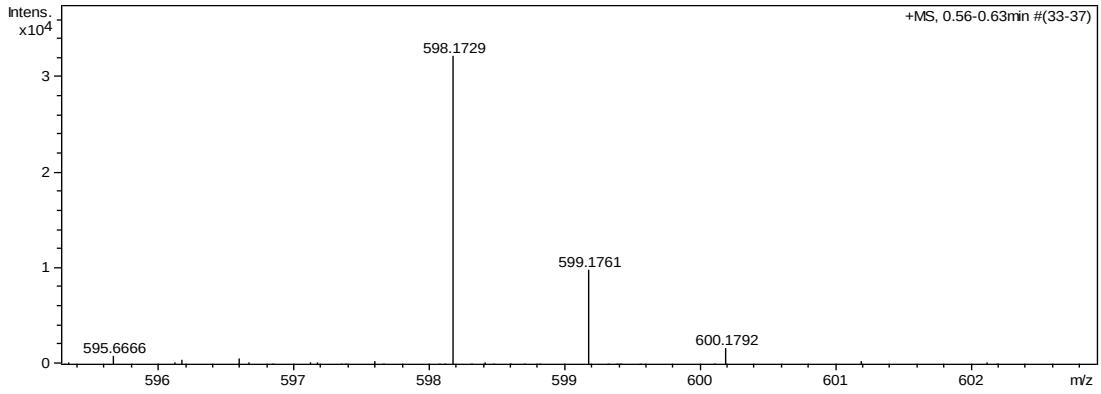

Gradient Shimming
199.58
199.40
176.26
174.76
142.76
142.23
141.37
141.00
138.11
138.10
137.73
128.87
128.86
125.41
124.60
124.24
122.73
122.30
120.89
120.83
120.01
119.21
117.46
116.28
111.71
110.74
110.41
65.19
62.39
56.58
32.98
31.09
25.86
H3C
N
O
O
O
N
H3C
N
H3C
O
f1 (ppm)


Intens.
x10^5
+MS, 0.24-0.34min #(14-20)
536.1574
537.1609
538.1645
m/z

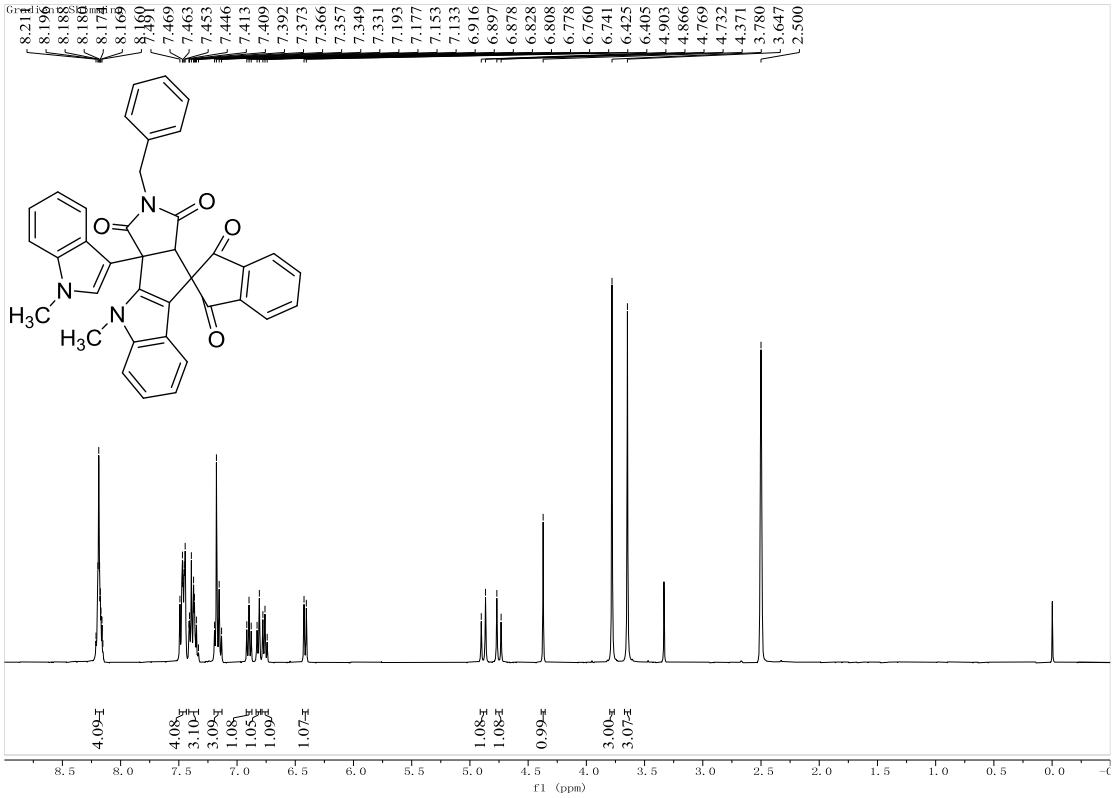
4'-methyl-3a'-(1-methyl-1*H*-indol-3-yl)-2'-phenyl-3a',9a'-dihydrospiro[indene-2,9'-pyrrolo[3',4':4,5]cyclopenta[1,2-*b*]indole]-1,1',3,3'(2'*H*,4'*H*)-tetraone (8e): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, yellow solid, 552 mg, yellow solid, 96%, m.p. 221-223 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 8.26-8.21 (m, 4H, ArH), 7.64-7.52 (m, 7H, ArH), 7.47 (d, *J* = 8.4 Hz, 1H, ArH), 7.24-7.16 (m, 3H, ArH), 6.97 (t, *J* = 7.2 Hz, 1H, ArH), 6.91 (t, *J* = 7.2 Hz, 1H, ArH), 6.41 (t, *J* = 8.0 Hz, 1H, ArH), 4.55 (s, 1H, CH), 3.88 (s, 3H, CH_3), 3.56 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 199.6, 199.3, 175.4, 174.0, 142.4, 142.3, 141.2, 141.1, 138.3, 138.2, 137.8, 132.2, 129.7, 129.6, 128.7, 127.5, 125.4, 124.6, 124.4, 122.8, 122.3, 120.9, 120.7, 120.0, 119.2, 117.4, 116.4, 111.7, 110.8, 110.0, 64.7, 62.9, 56.6, 33.0, 30.9; IR (KBr) υ: 3045, 2934, 2821, 2669, 1939, 1714, 1611, 1545, 1469, 1431, 1394, 1175, 1342, 1245, 1133, 1051, 976, 931, 857, 756 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{37}H_{25}NaN_3O_4$ ([M+Na]$^+$): 5498.1737, Found: 598.1729.

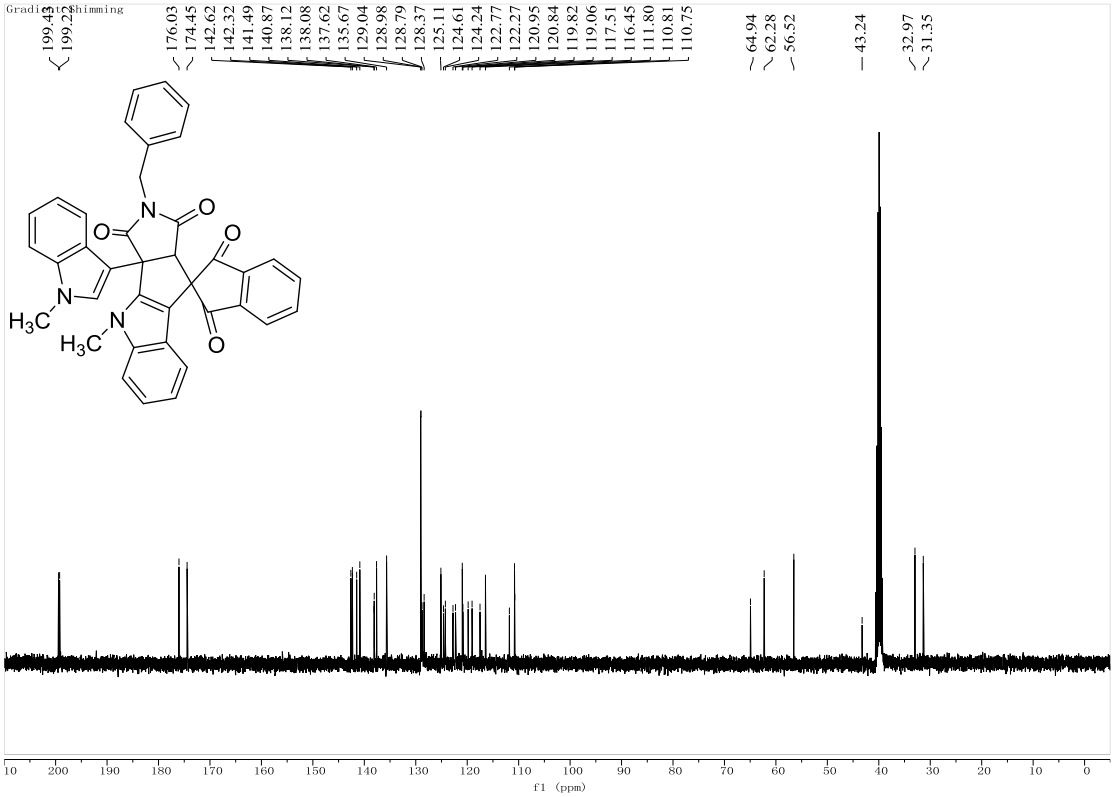


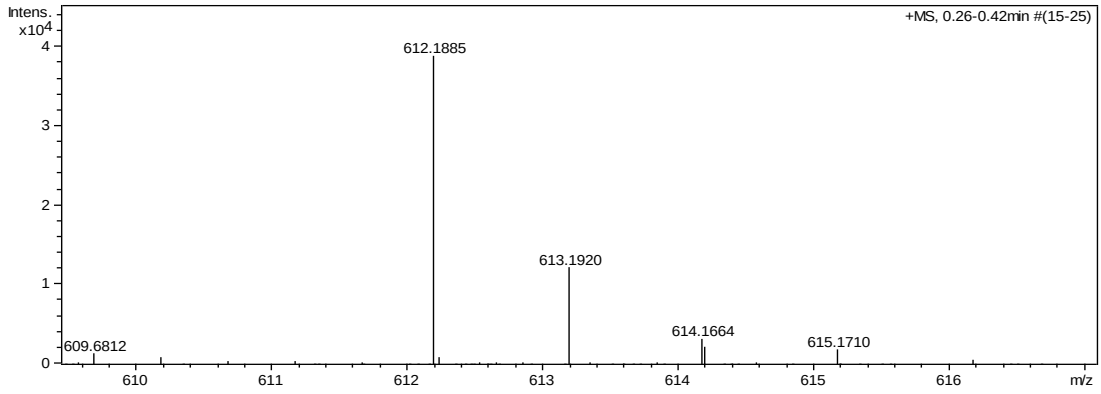

Gradient Shimming
199.68
199.38
175.46
174.03
142.51
142.30
141.25
141.15
138.39
138.25
137.84
132.31
129.76
129.63
128.77
127.61
125.48
124.71
124.44
122.85
122.40
120.95
120.81
120.08
119.30
117.47
116.41
111.81
110.83
110.08
64.75
62.97
56.66
33.03
30.97
O
O
O
O
N
N
H3C
H3C
10 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


Intens.
x10^4
+MS, 0.56-0.63min #(33-37)
598.1729
599.1761
600.1792
595.6666
0
1
2
3
596
597
598
599
600
601
602
m/z

2'-benzyl-4'-methyl-3a'-(1-methyl-1*H*-indol-3-yl)-3a',9a'-dihydrospiro[indene-2,9'-pyrrolo[3',4':4,5]cyclopenta[1,2-*b*]indole]-1,1',3,3'(2'*H*,4'*H*)-tetraone (8f): petroleum ether and methylene dichloride (V/V = 4:1) as the eluent, yellow solid, 560 mg, yellow solid, 95%, m.p. 226-228 °C; ^{1}H NMR (400 MHz, d_6-DMSO) δ: 8.21-8.15 (m, 4H, ArH), 7.49-7.45 (m, 4H, ArH), 7.41-7.33 (m, 3H, ArH), 7.19-7.13 (m, 3H, ArH), 6.90 (t, *J* = 7.6 Hz, 1H, ArH), 6.82 (d, *J* = 8.0 Hz, 1H, ArH), 6.76 (d, *J* = 7.6 Hz, 1H, ArH), 6.42 (d, *J* = 8.0 Hz, 1H, ArH), 4.88 (d, *J* = 14.8 Hz, 1H, CH), 4.75 (d, *J* = 14.8 Hz, 1H, CH), 4.37 (s, 1H, CH), 3.78 (s, 3H, CH_3), 3.65 (s, 3H, CH_3); ^{13}C NMR (100 MHz, d_6-DMSO) δ: 199.4, 199.2, 176.0, 174.4, 142.6, 142.3, 141.4, 140.8, 138.1, 138.0, 137.6, 135.6, 129.0, 128.9, 128.7, 128.3, 125.1, 124.5, 124.2, 122.7, 122.2, 120.9, 120.8, 119.8, 119.0, 117.4, 116.4, 111.7, 110.8, 110.7, 64.9, 62.2, 56.5, 43.2, 32.9, 31.3; IR (KBr) υ: 3052, 2934, 2825, 2668, 1934, 1711, 1614, 1545, 1475, 1431, 1389, 1175, 1345, 1245, 1135, 1051, 978, 933, 852, 756 cm^{-1}; MS (*m*/*z*): HRMS (ESI-TOF) Calcd. for $C_{38}H_{27}NaN_3O_4$ ([M+Na]$^+$): 612.1894, Found: 612.1885.




Gradient Shimming
199.43
199.22
176.03
174.45
142.62
142.32
141.49
140.87
138.12
138.08
137.62
135.67
129.04
128.98
128.79
128.37
125.11
124.61
124.24
122.77
122.27
120.95
120.84
119.82
119.06
117.51
116.45
111.80
110.81
110.75
64.94
62.28
56.52
43.24
32.97
31.35
O
O
O
O
N
N
H3C
H3C
10 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


Intens.
x10^4
4
3
2
1
0
+MS, 0.26-0.42min #(15-25)
612.1885
613.1920
614.1664
615.1710
609.6812
610
611
612
613
614
615
616
m/z