Organocatalytic Enantioselective Michael/Cyclization Domino Reaction between 3-Amideoxindoles and *α,β*-Unsaturated Aldehydes: One-Pot Preparation of Chiral Spirocyclic Oxindole-*γ*-Lactams

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Supporting Information

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1. Crystallographic Information for Spirocyclic Oxindole-γ-Lactams 4d

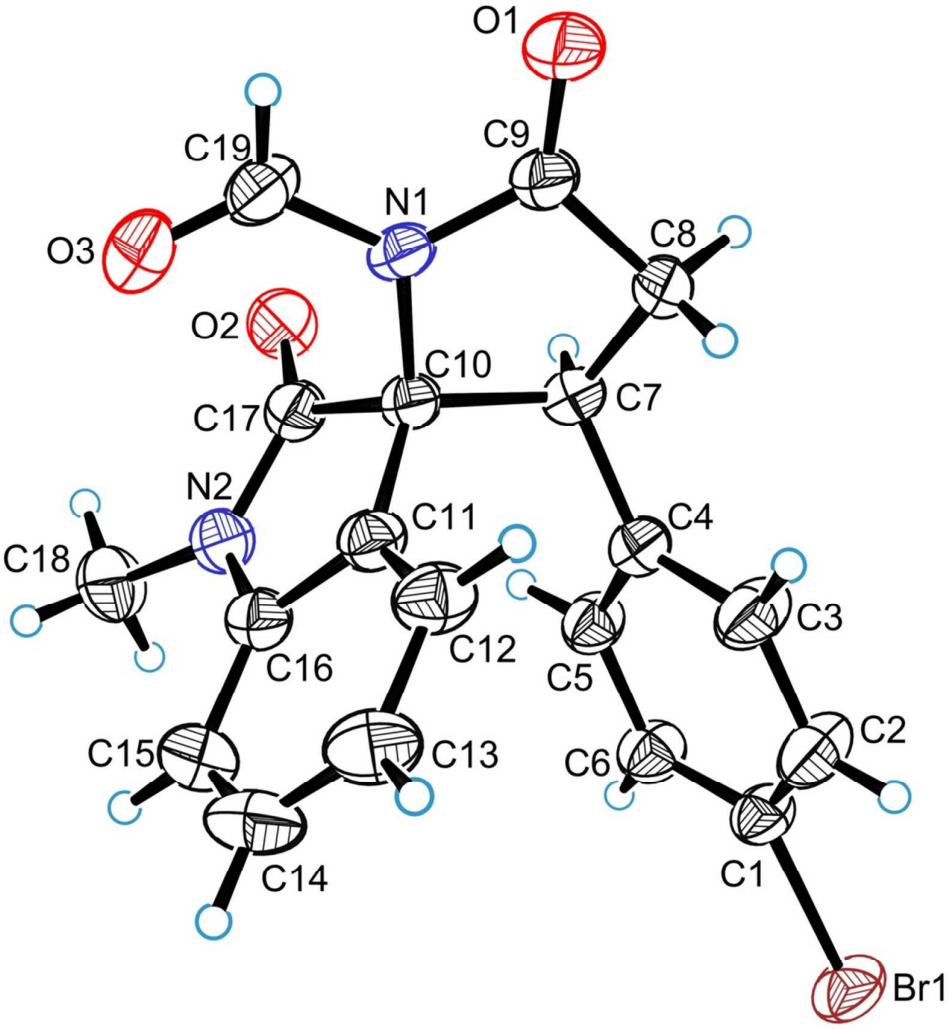


Figure S1. ORTEP plot of compound **4d** (CCDC 1497000)
Thermal ellipsoids are drawn at 30% probability level.

Table S1: Important crystal data of compound **4d**

Empirical formula	$C_{19}H_{15}BrN_2O_3$
Formula weight	399.24
Temperature/K	290(2)
Crystal system	trigonal
Space group	R3
a/Å	20.8688(6)
b/Å	20.8688(6)
c/Å	12.2249(4)
α/°	90
β/°	90
γ/°	120
Volume/Å^3	4610.7(3)
Z	9
ρ_{calc}g/cm^3	1.294
μ/mm^{-1}	2.884
F(000)	1818.0
Crystal size/mm^3	0.210 × 0.200 × 0.180
Radiation	CuKα (λ = 1.54184)
2Θ range for data collection/°	8.474 to 142.48
Index ranges	$-25 \leq h \leq 17$, $-16 \leq k \leq 24$, $-14 \leq l \leq 14$
Reflections collected	3434
Independent reflections	2362 [R_{int} = 0.0206, R_{sigma} = 0.0252]
Data/restraints/parameters	2362/1/227
Goodness-of-fit on F^2	1.048
Final R indexes [I>=2σ (I)]	R_1 = 0.0264, wR_2 = 0.0710
Final R indexes [all data]	R_1 = 0.0266, wR_2 = 0.0713
Largest diff. peak/hole / e Å^{-3}	0.26/-0.34
Flack parameter	-0.014(17)

2. NMR spectra of products 4 and 6

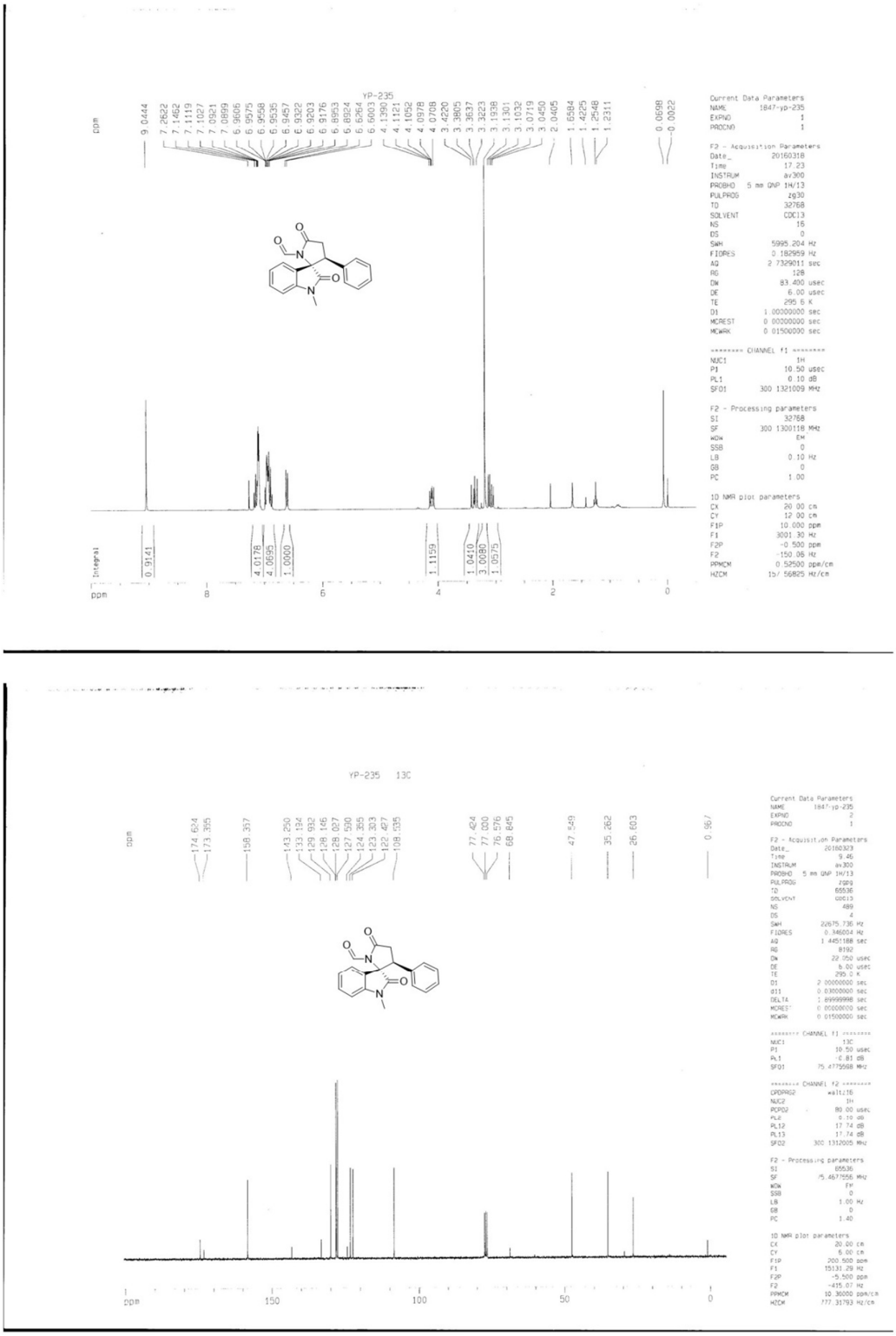
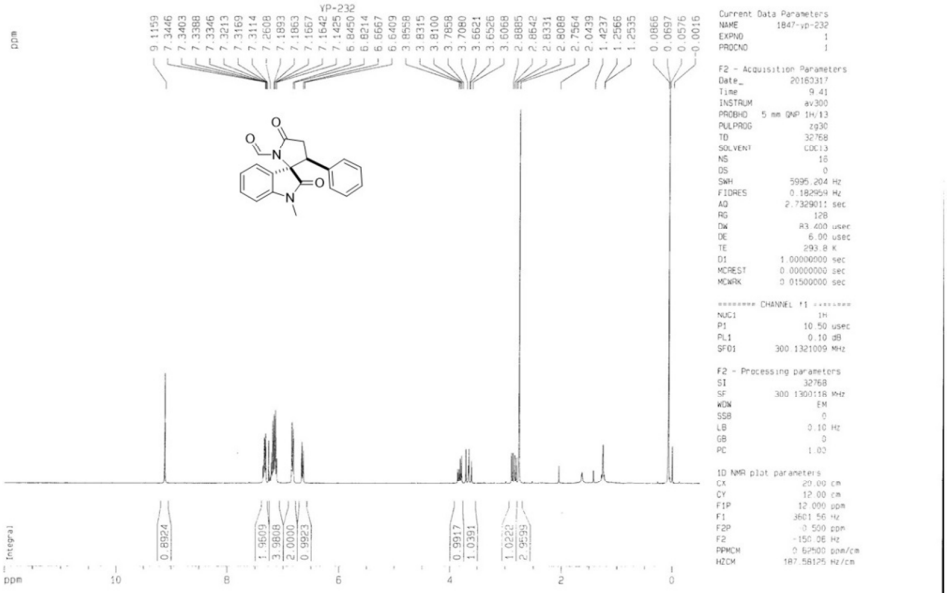


Figure S2: NMR spectra of compound **4a**



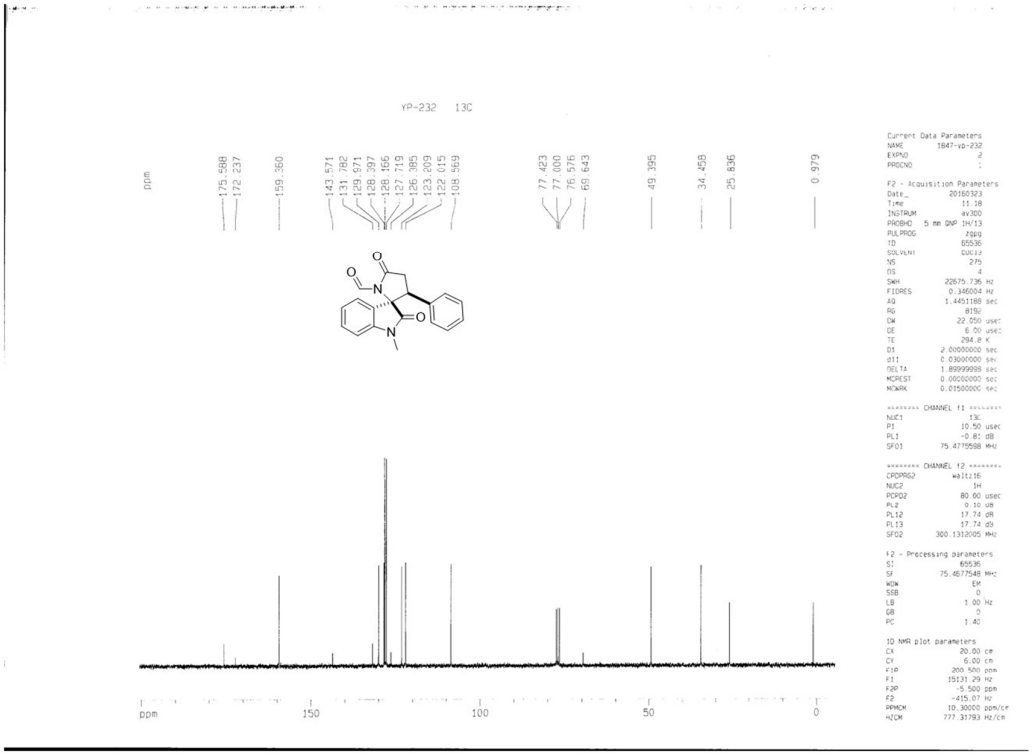


Figure S2': NMR spectra of compound **4a'**

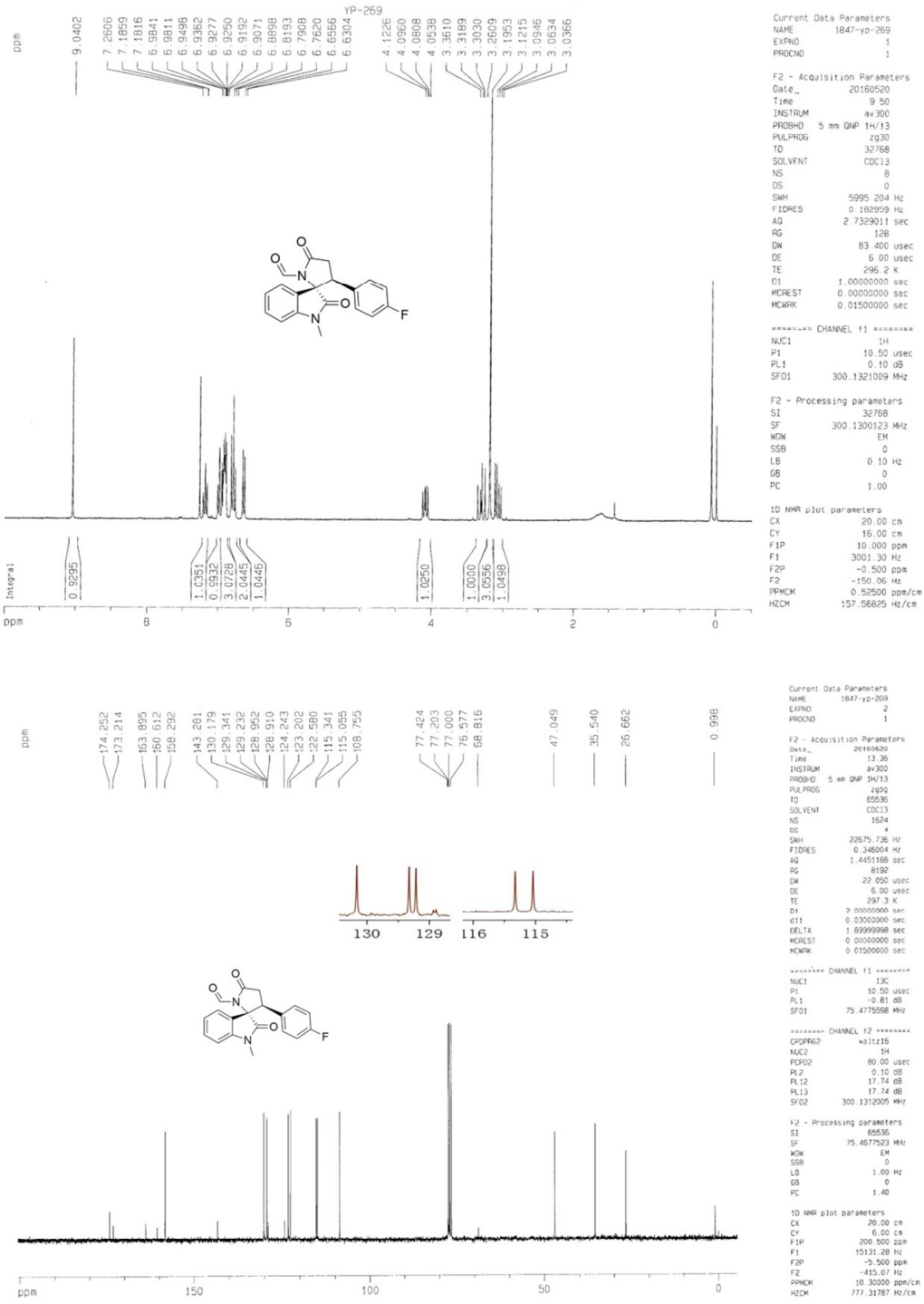
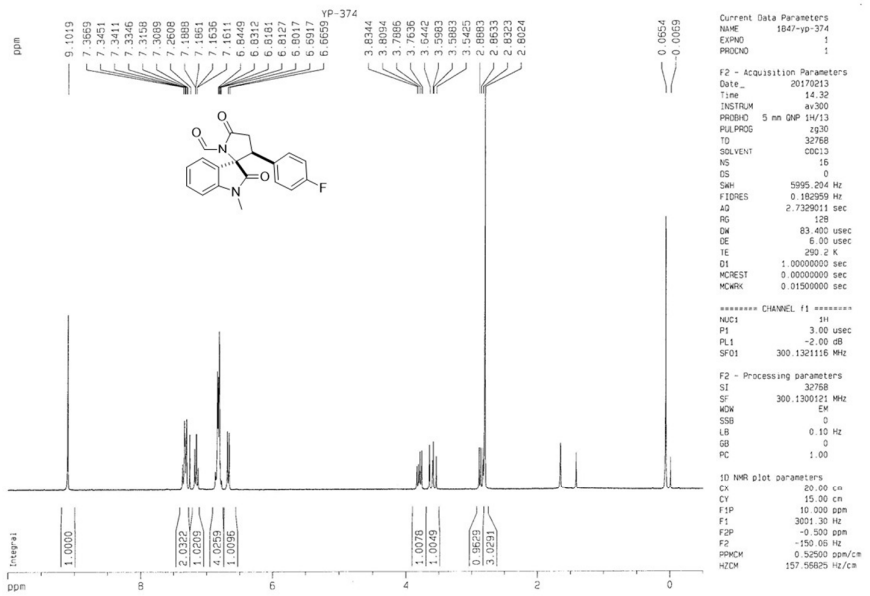


Figure S3: NMR spectra of compound **4b**



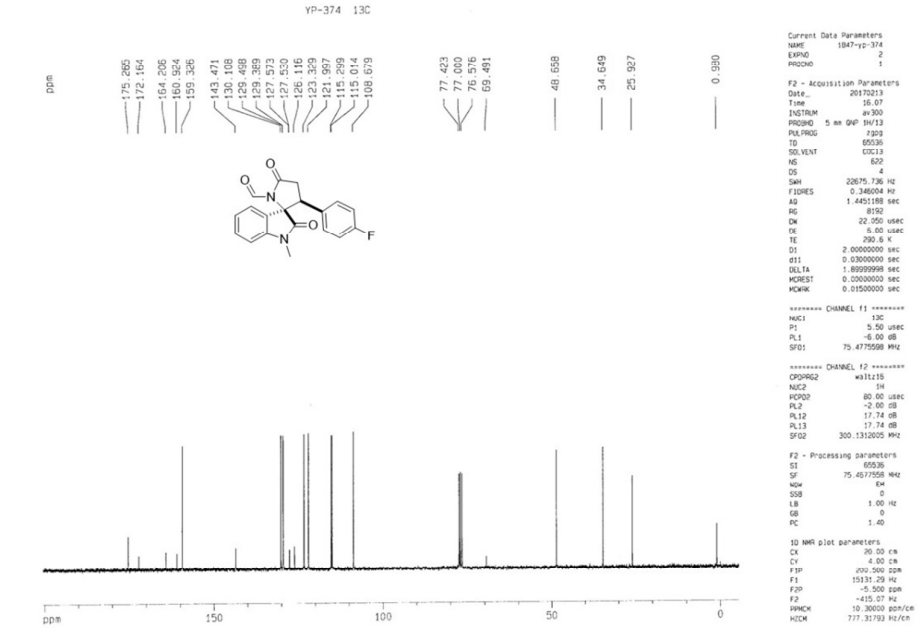
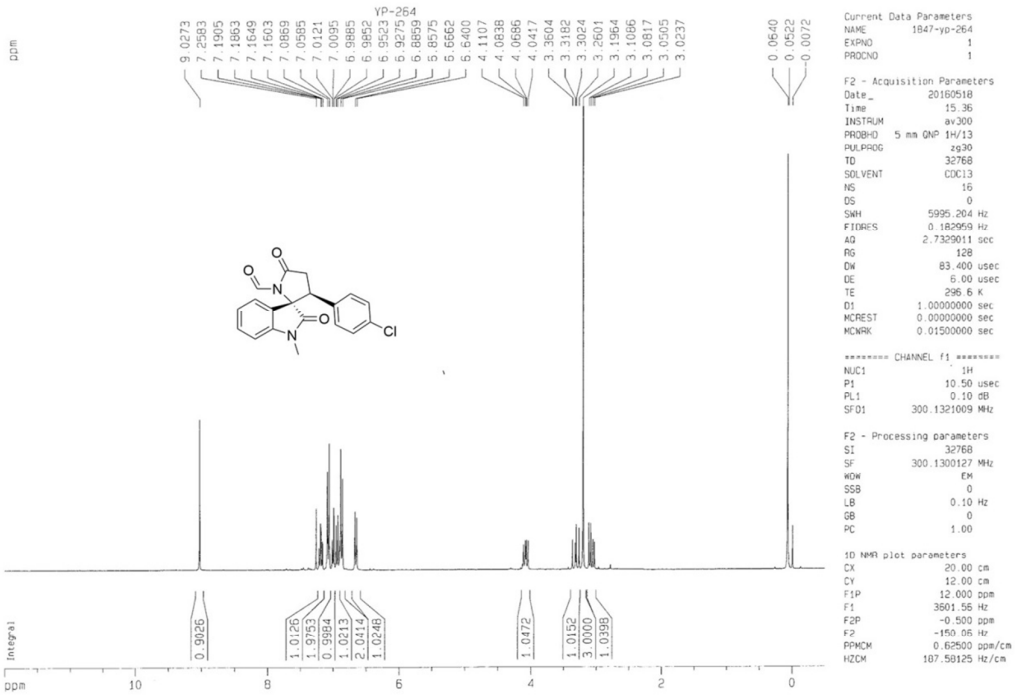


Figure S3': NMR spectra of compound **4b'**



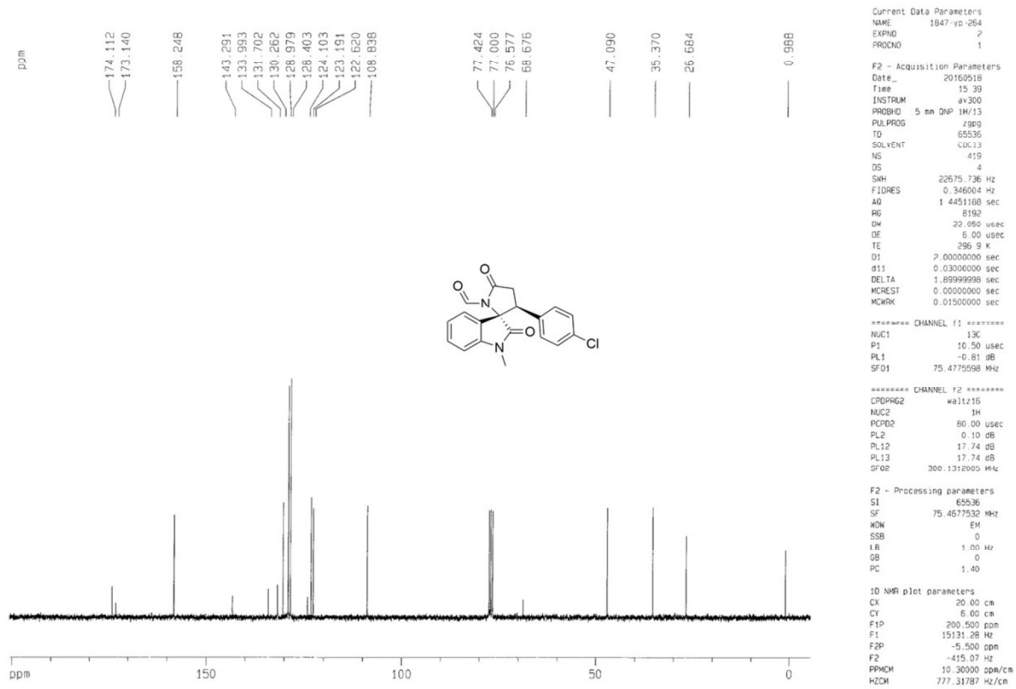
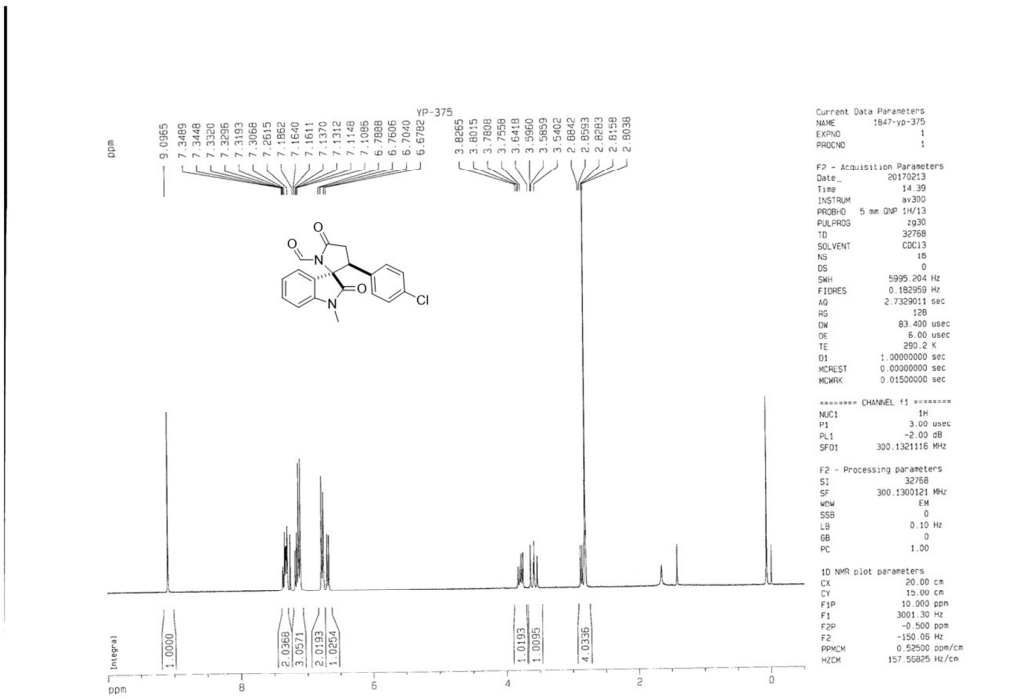


Figure S4: NMR spectra of compound **4c**



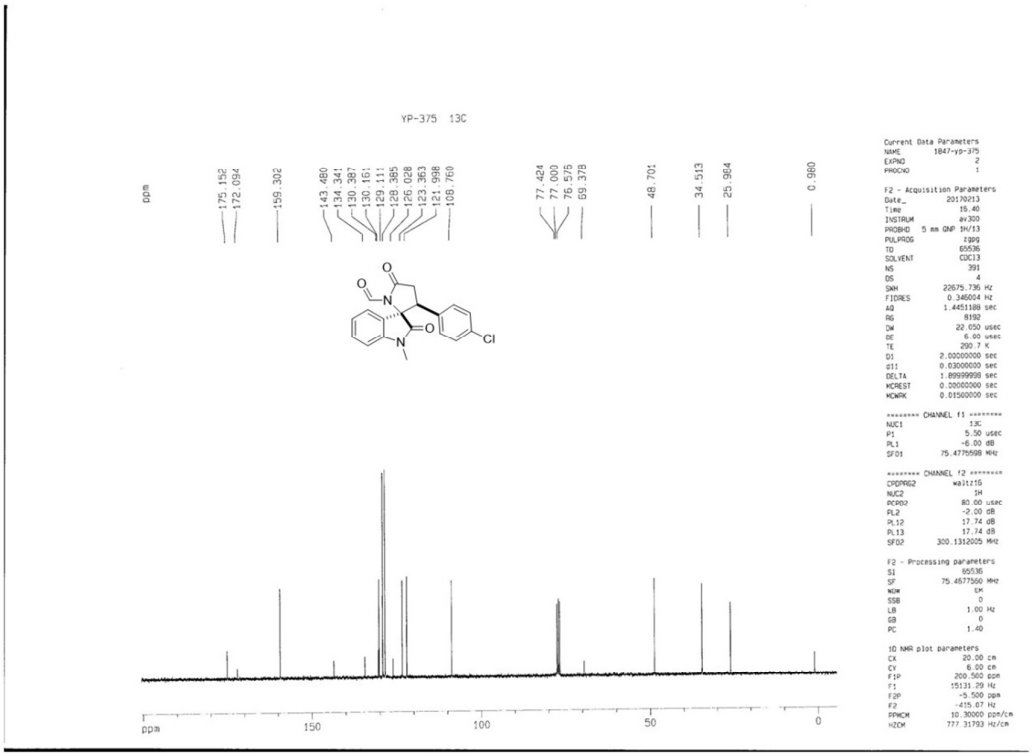
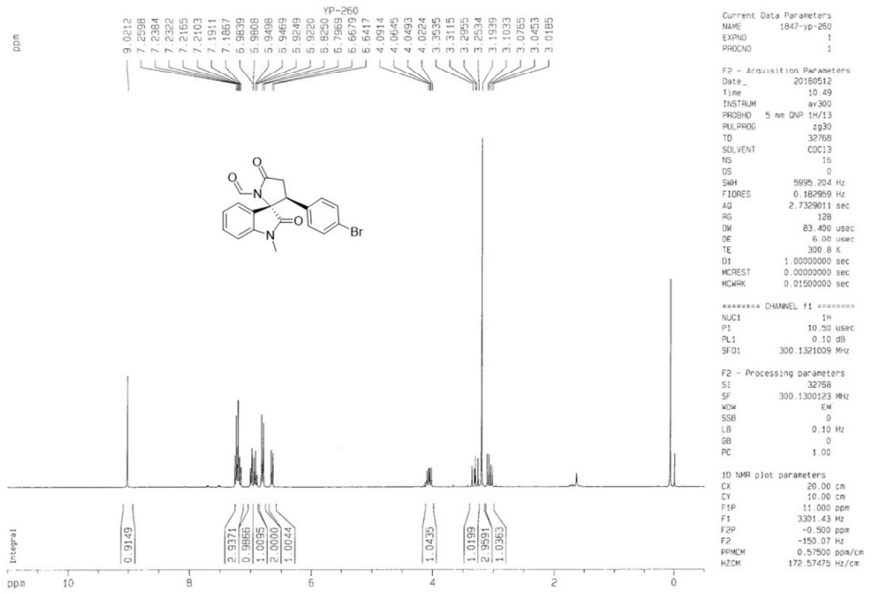


Figure S4': NMR spectra of compound **4c'**



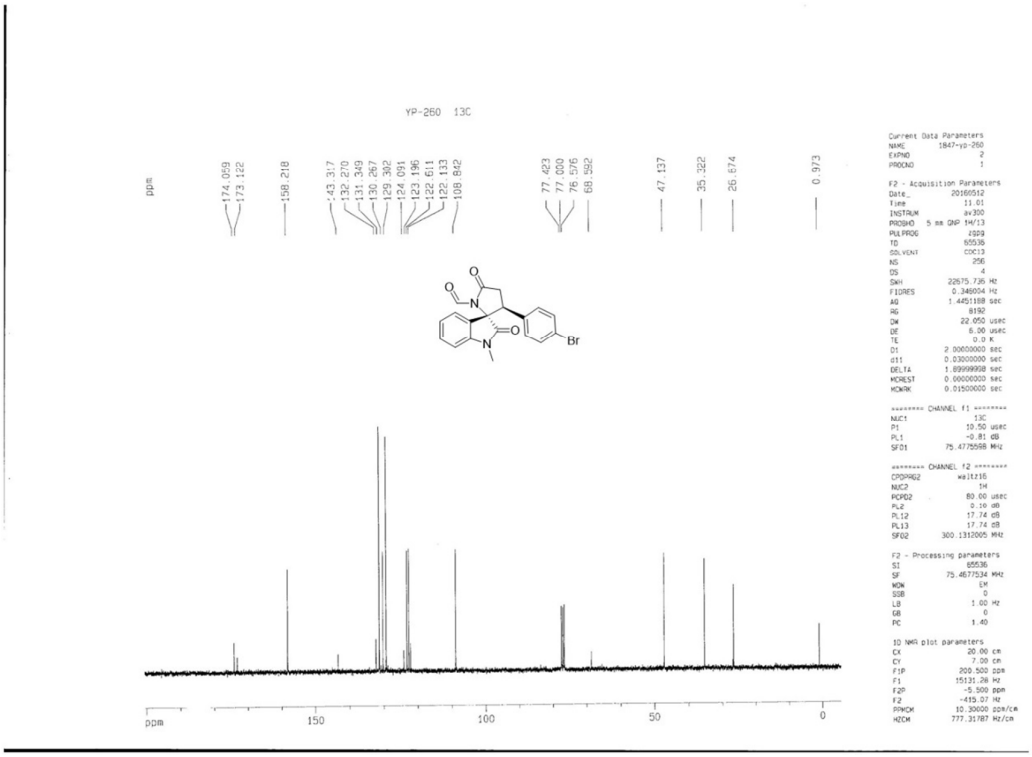
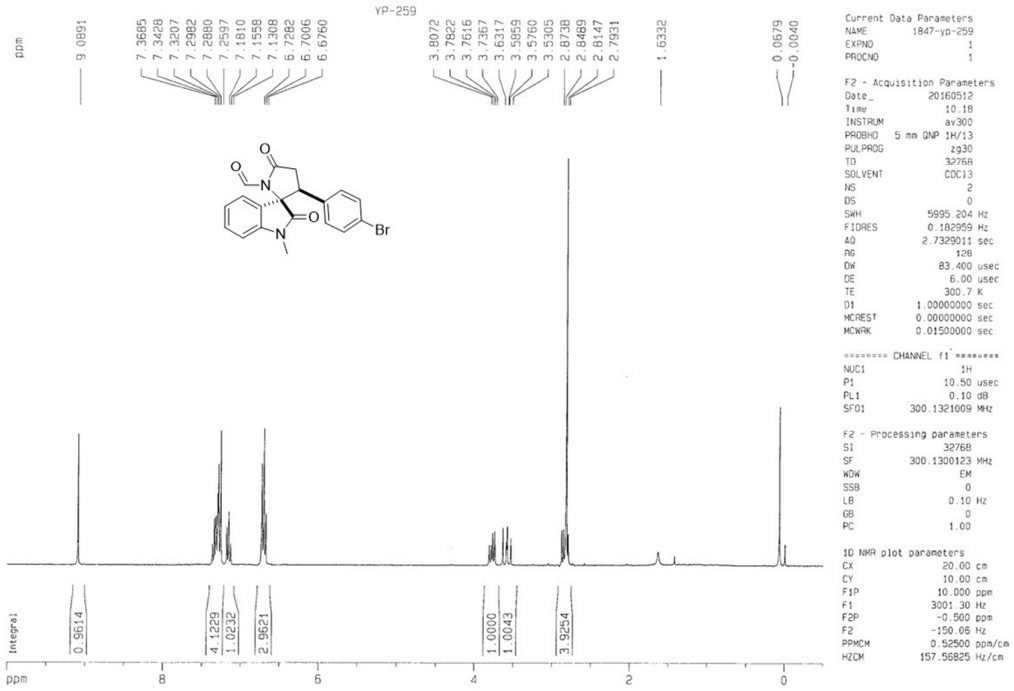


Figure S5: NMR spectra of compound **4d**

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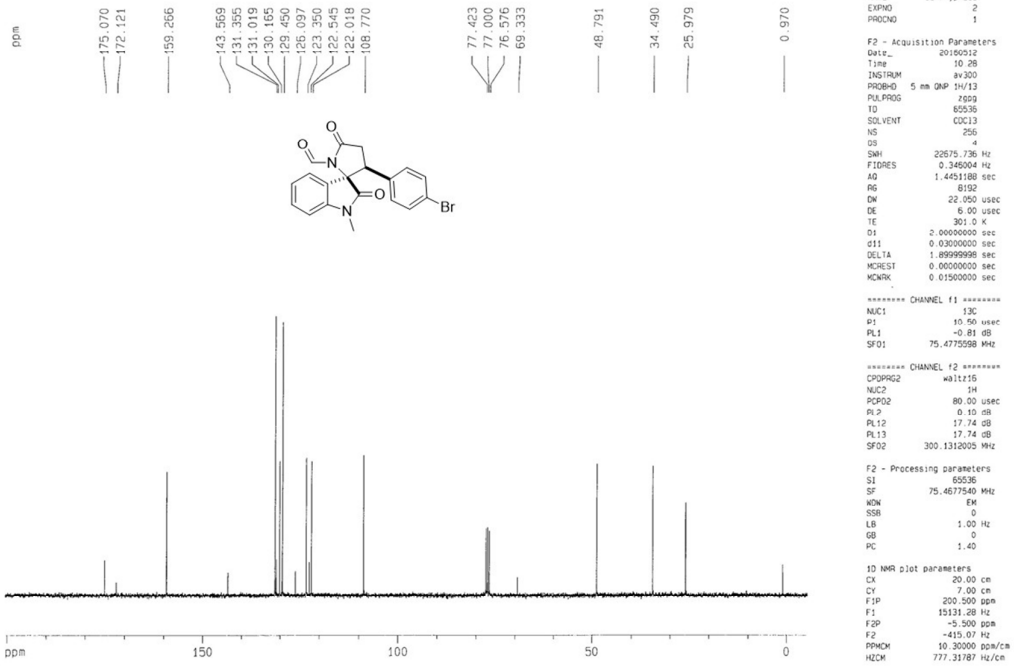


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MCWRK          0.01500000 sec

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P1                10.50 usec
PL1               -0.81 dB
SFO1         75.4775598 MHz

======== CHANNEL f2 ========
CPDPRG2         waltz16
NUC2                 1H
PCPD2            80.00 usec
PL2                0.10 dB
PL12              17.74 dB
PL13              17.74 dB
SFO2         300.1312005 MHz

F2 - Processing parameters
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SSB                   0
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PC                 1.40

1D NMR plot parameters
CX                20.00 cm
CY                 7.00 cm
F1P              200.500 ppm
F1             15131.28 Hz
F2P              -5.500 ppm
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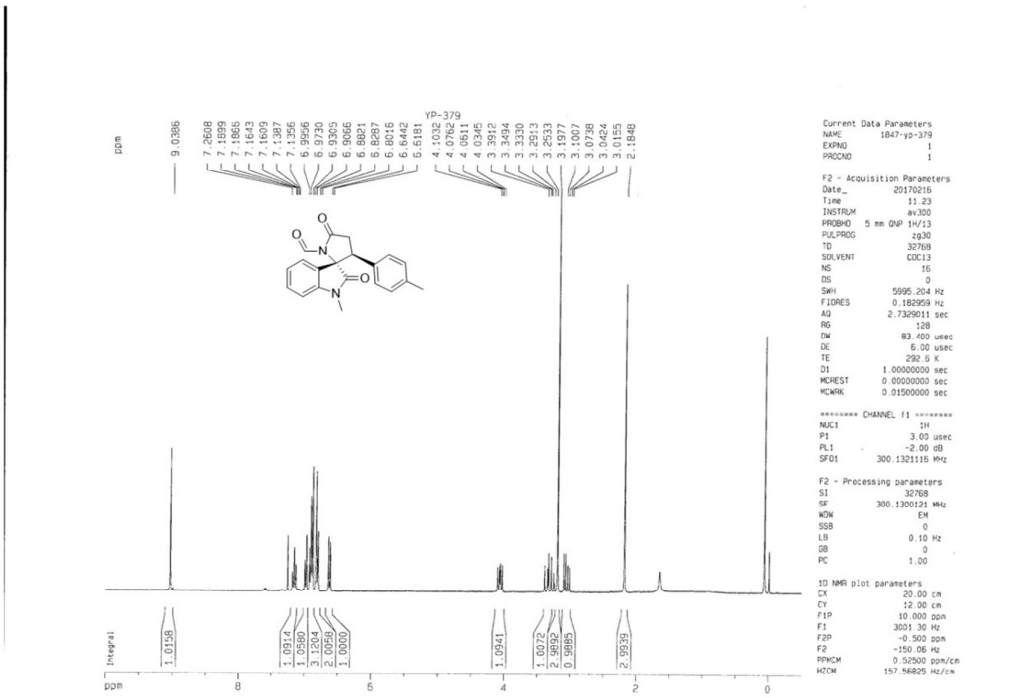
Figure S5': NMR spectra of compound **4d'**



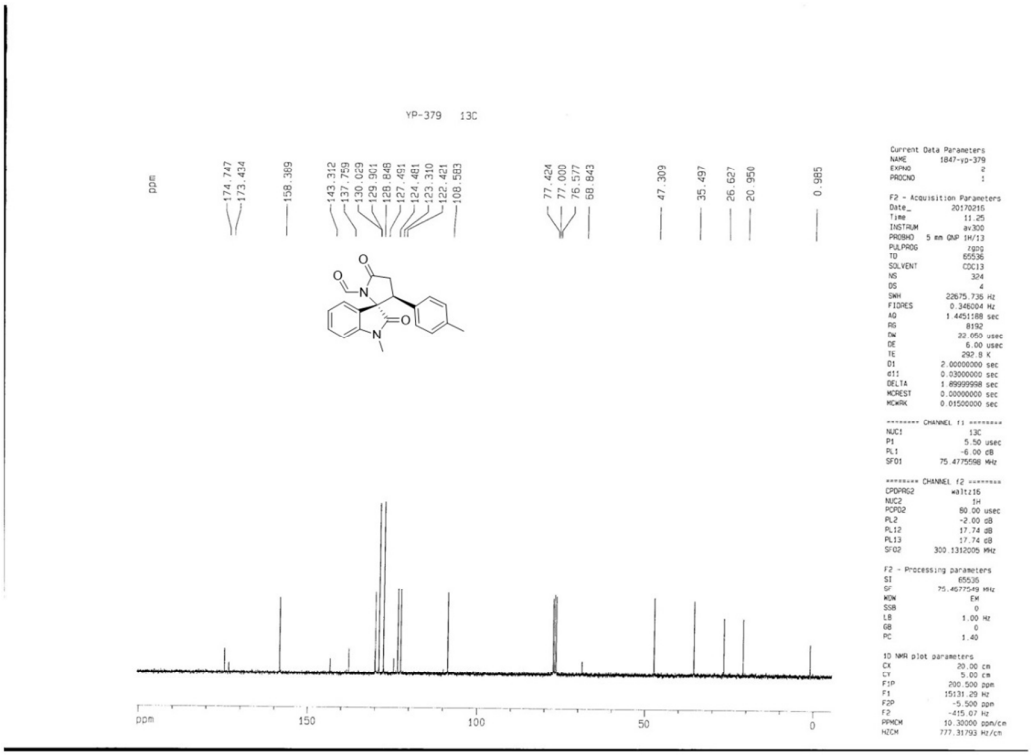
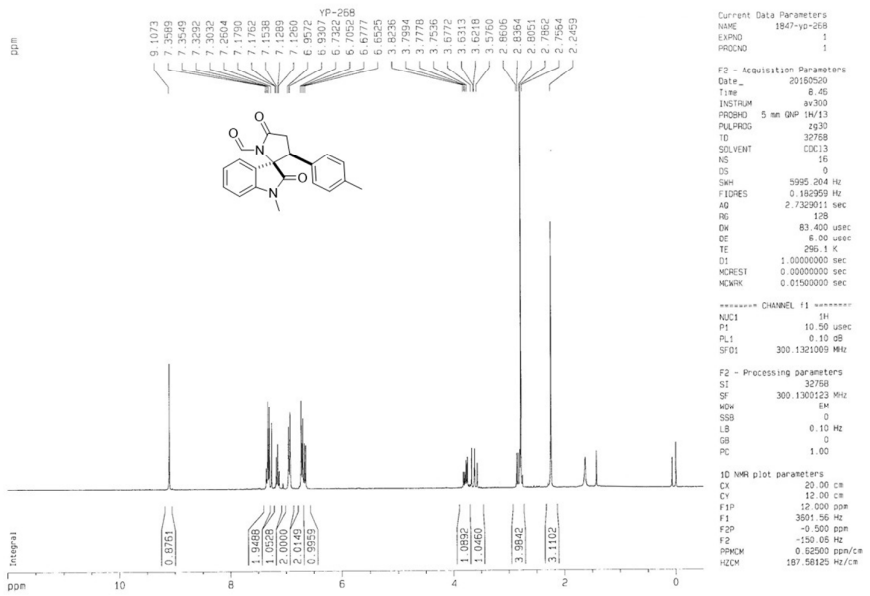


Figure S6: NMR spectra of compound **4e**



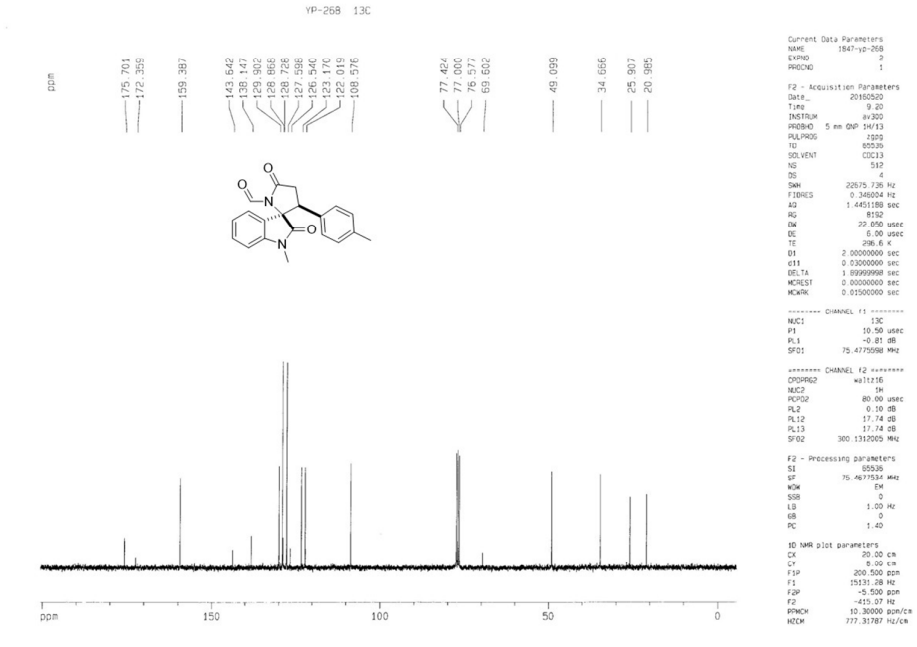
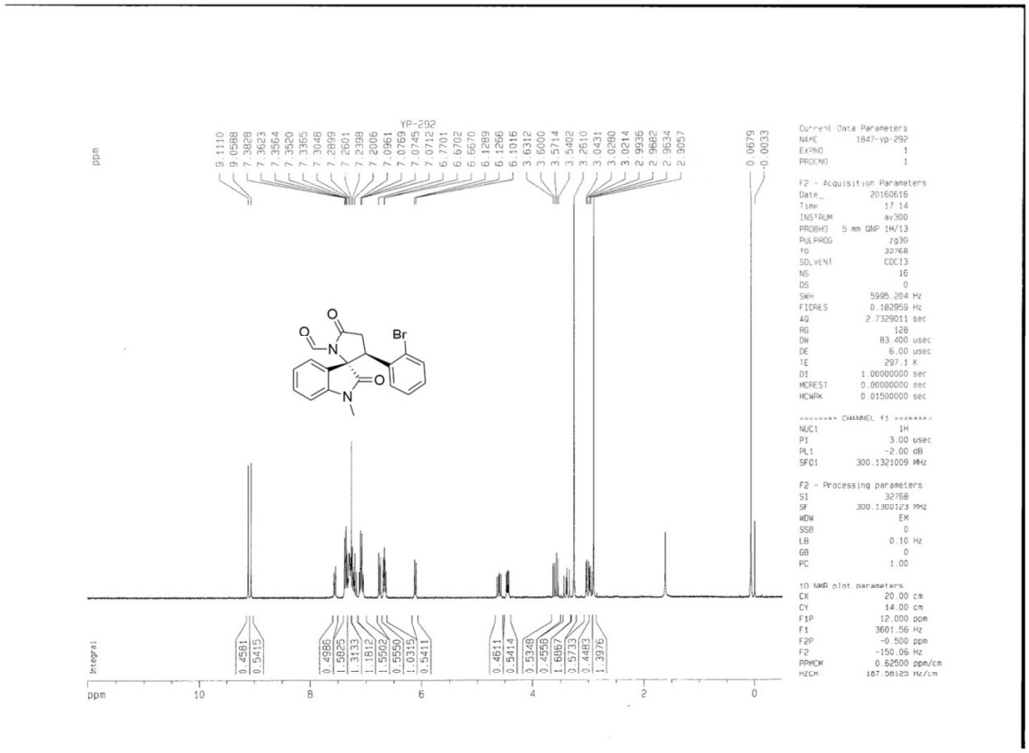


Figure S6': NMR spectra of compound 4e'



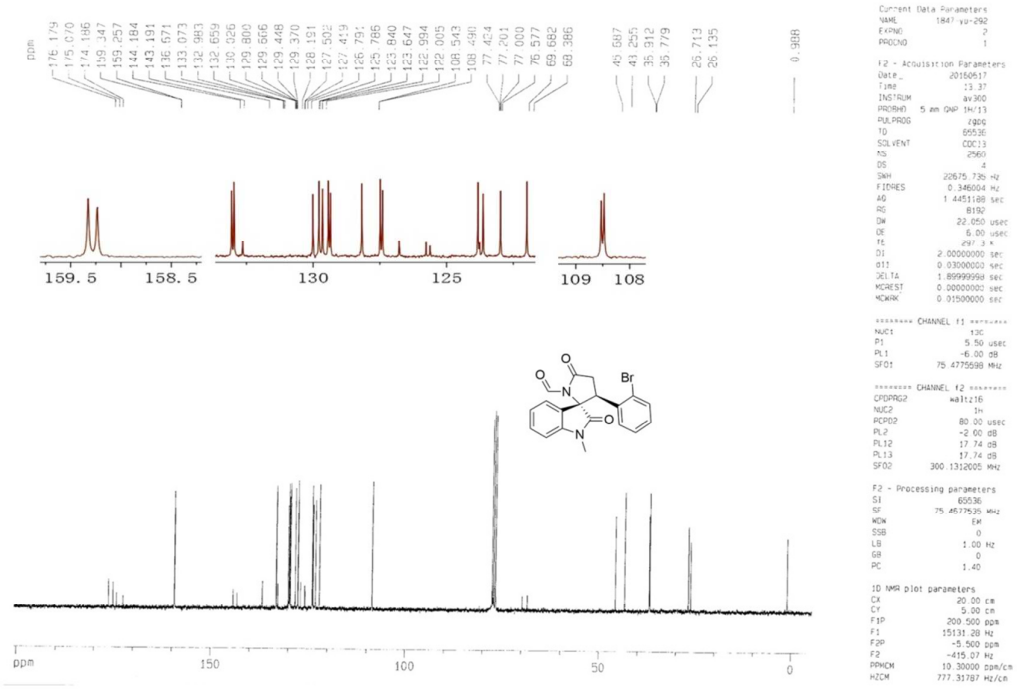
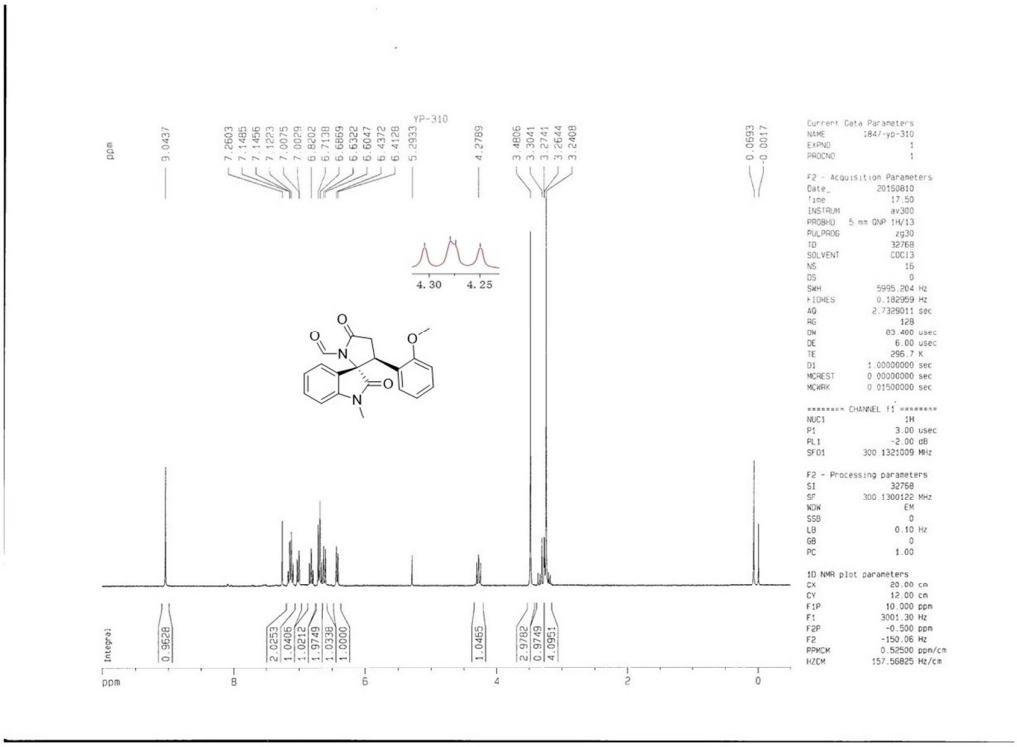


Figure S7: NMR spectra of compound **4f**



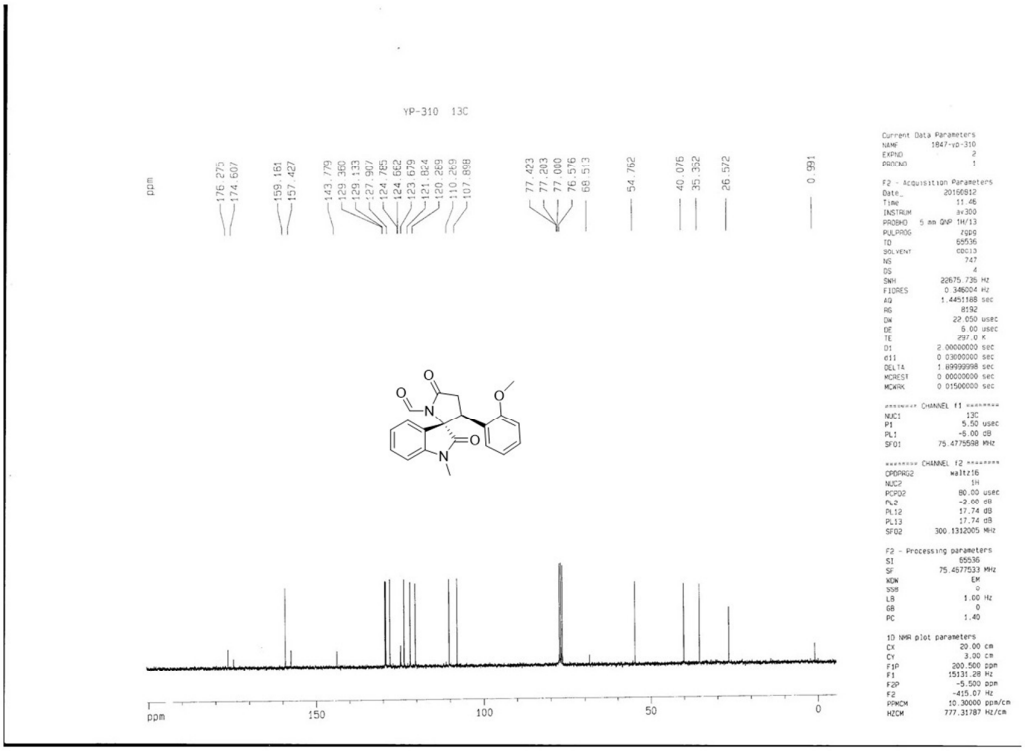
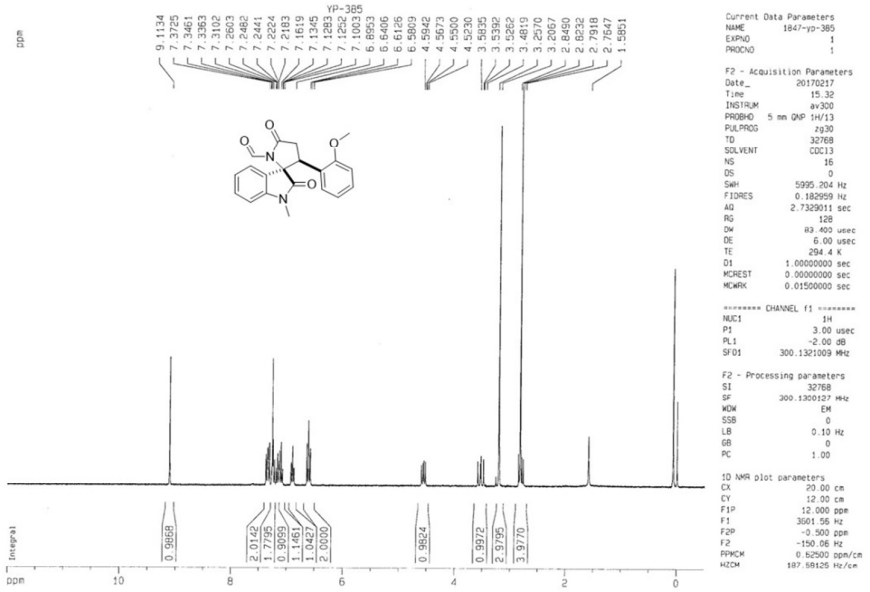


Figure S8: NMR spectra of compound **4g**



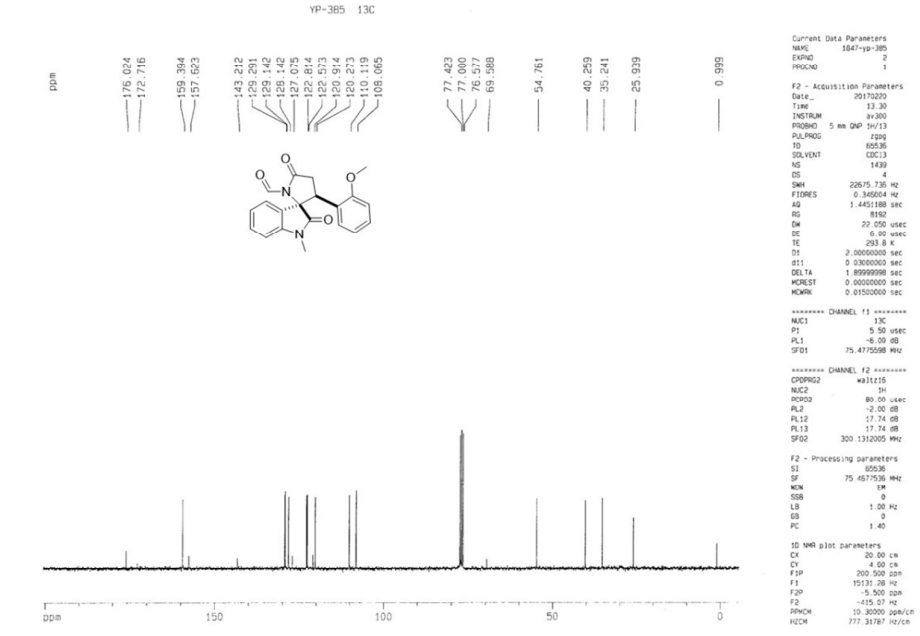


Figure S8': NMR spectra of compound 4g'

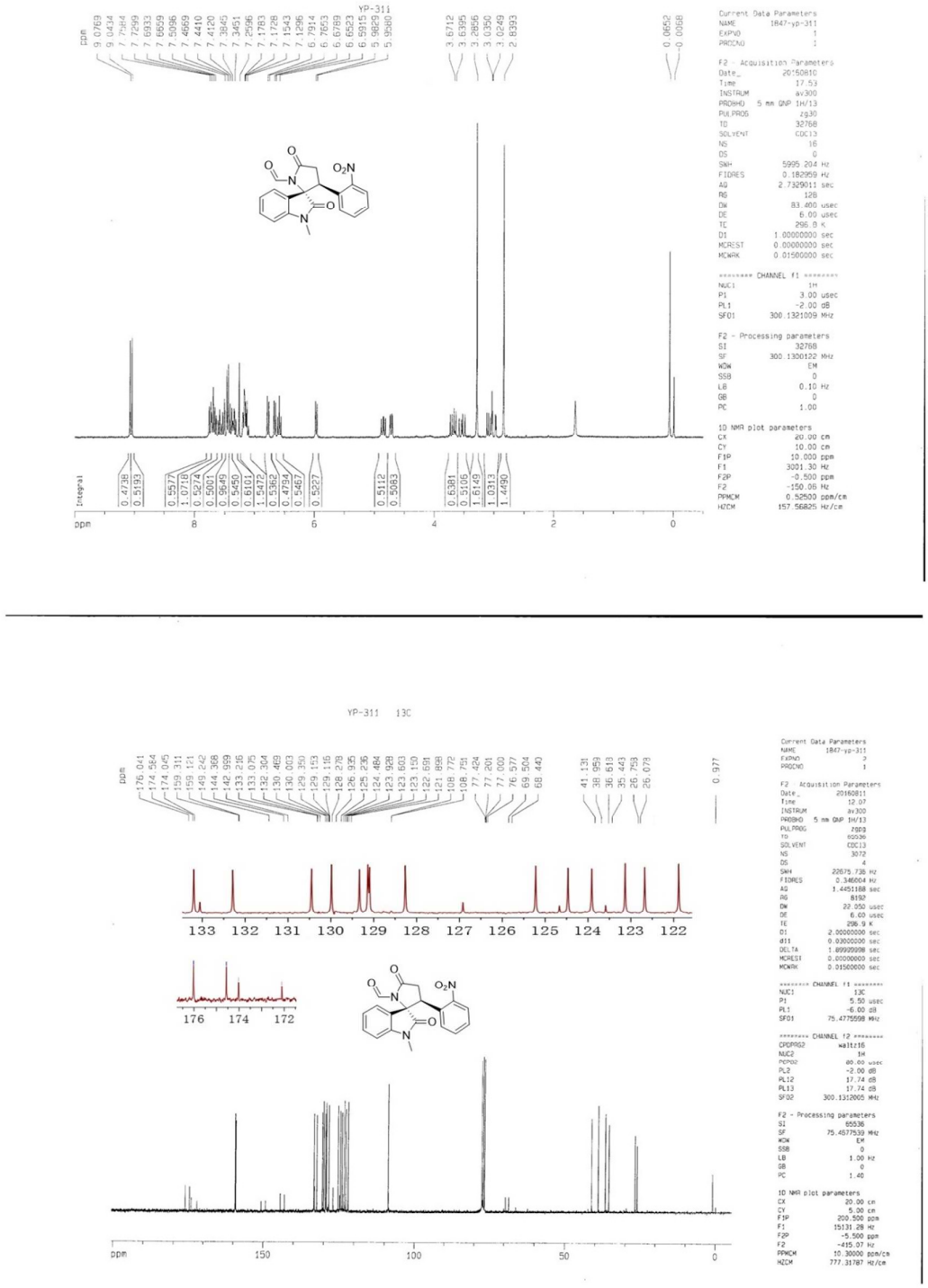
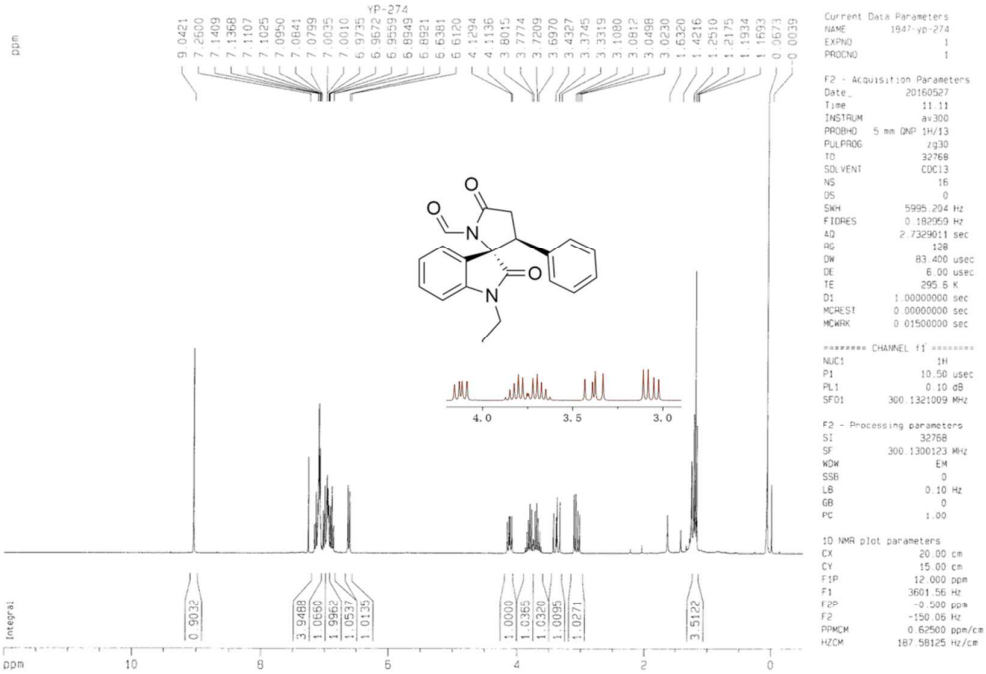


Figure S9: NMR spectra of compound **4h**



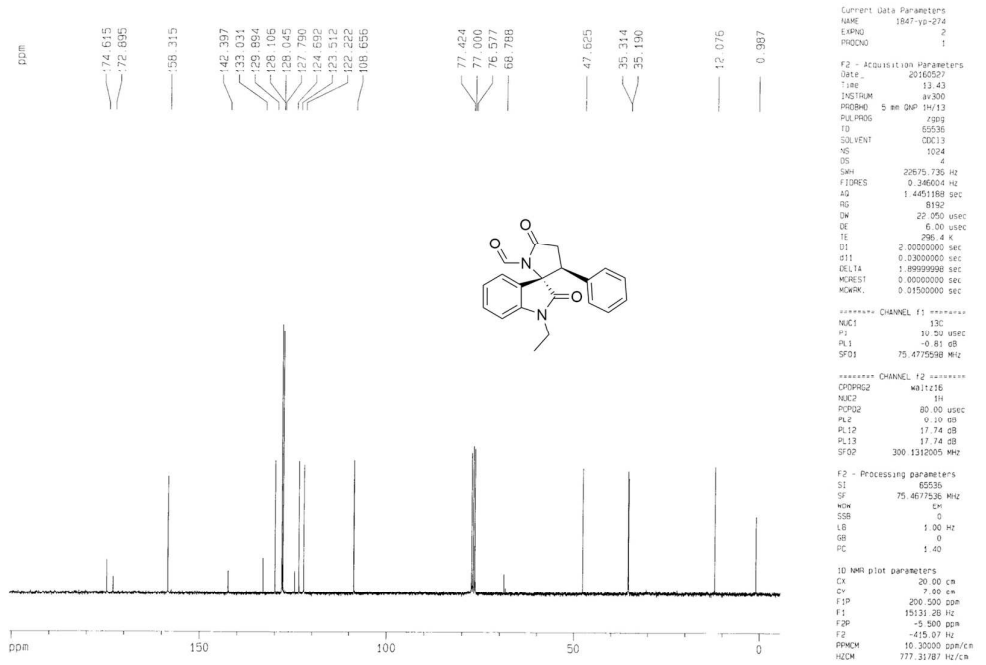
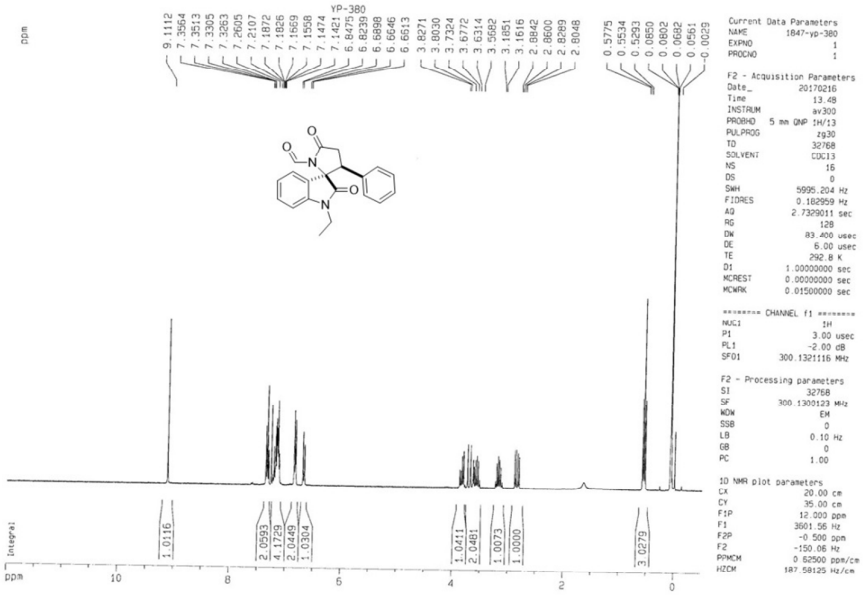


Figure S10: NMR spectra of compound **4i**



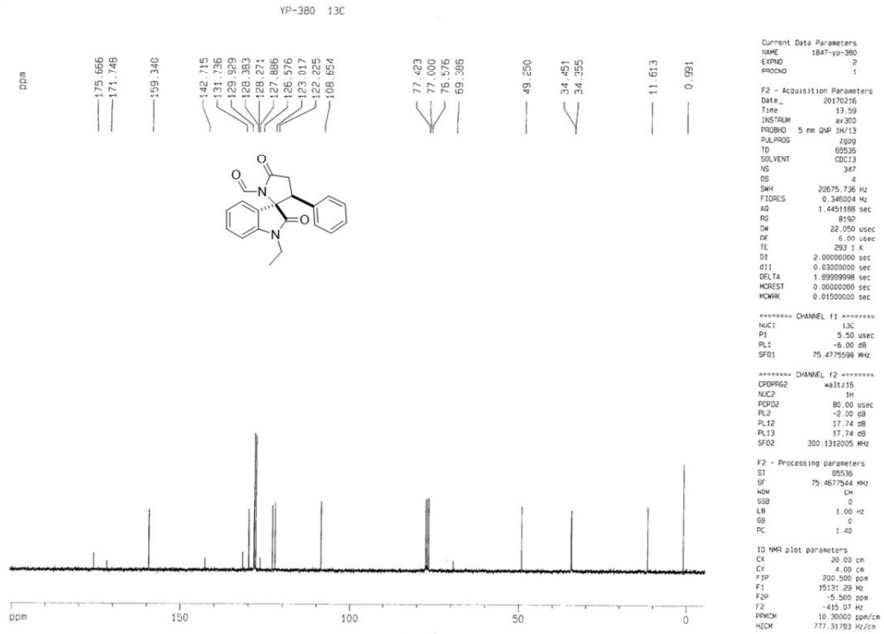
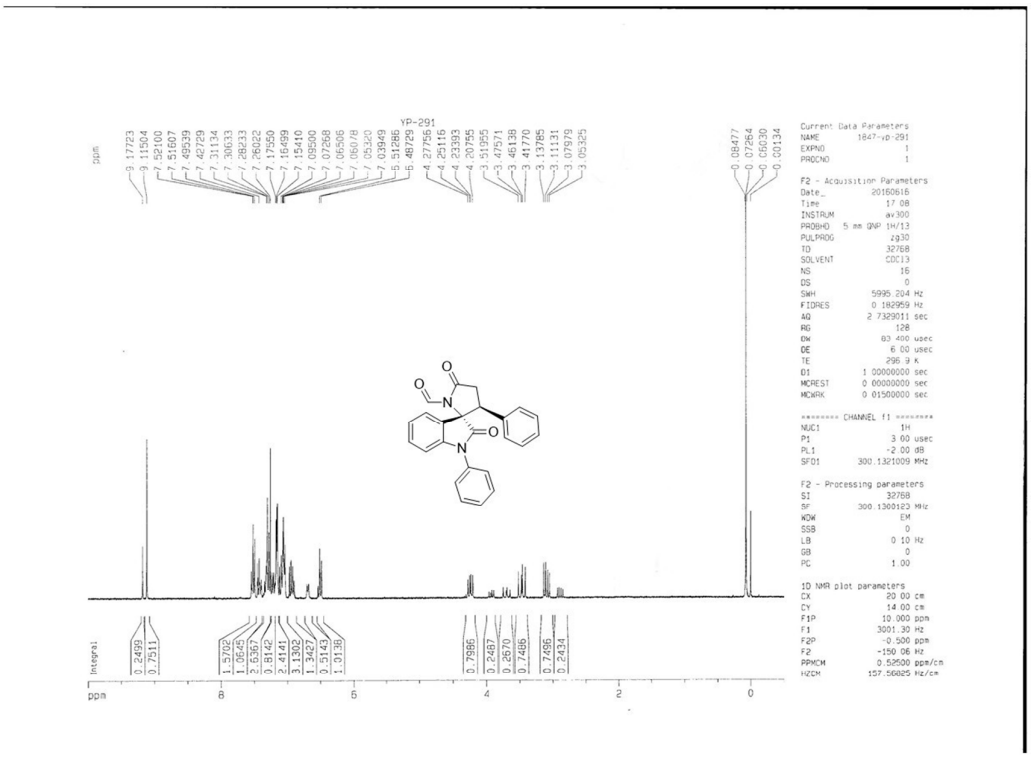
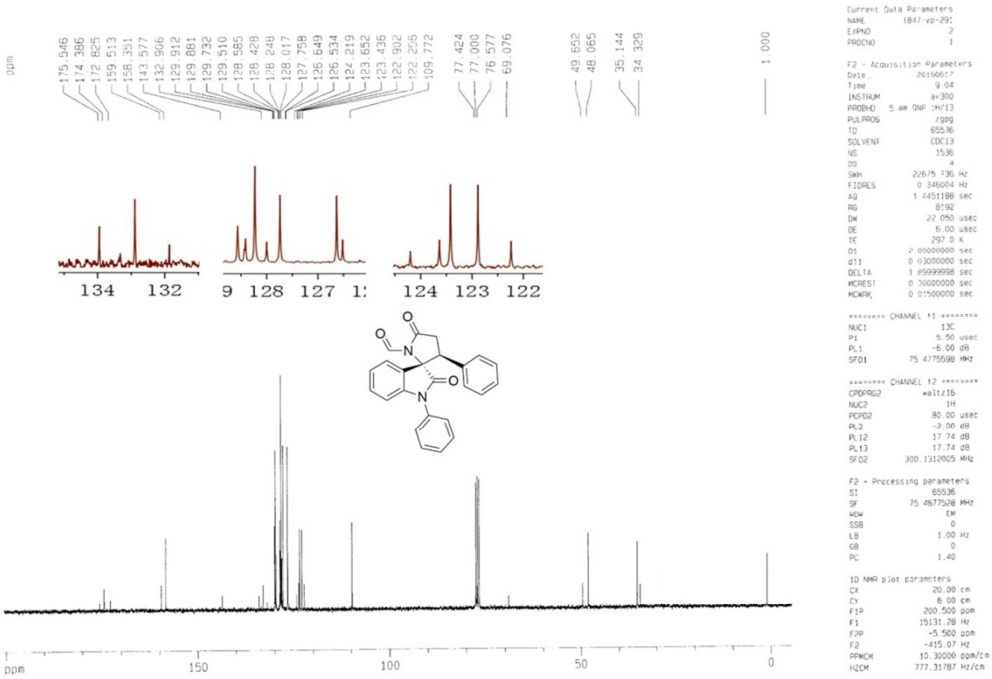


Figure S10': NMR spectra of compound 4i'





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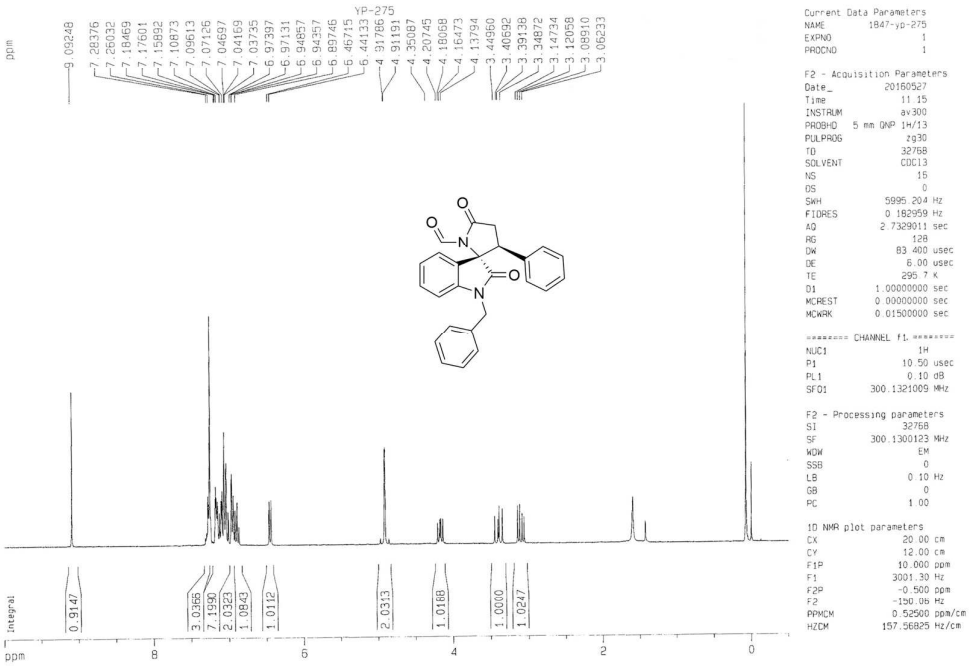


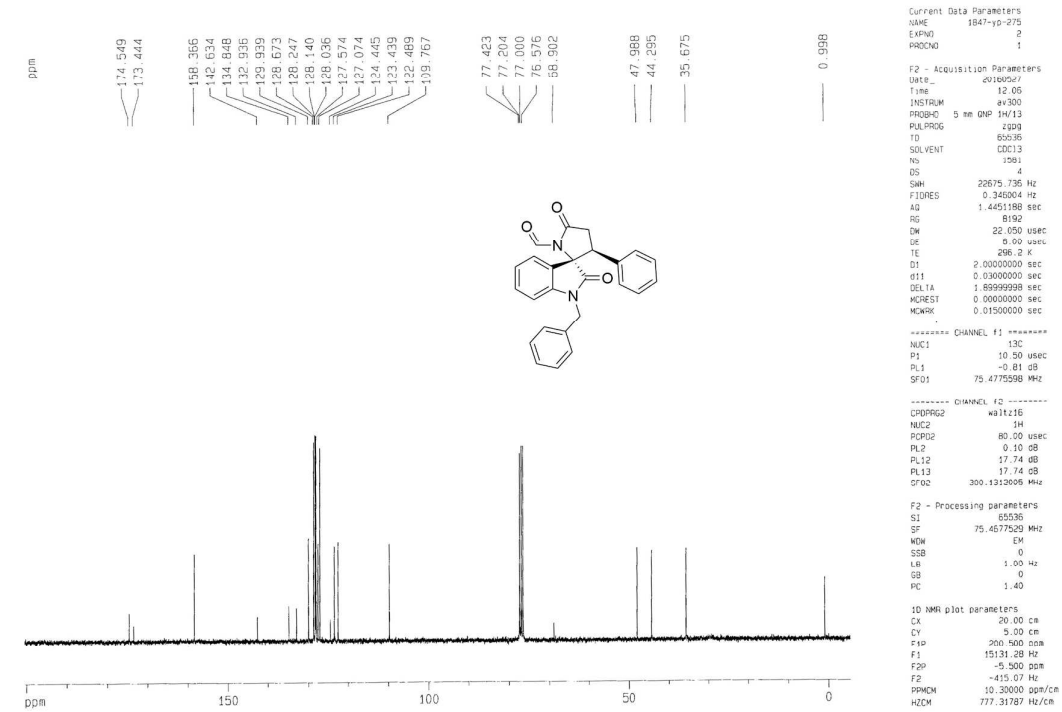
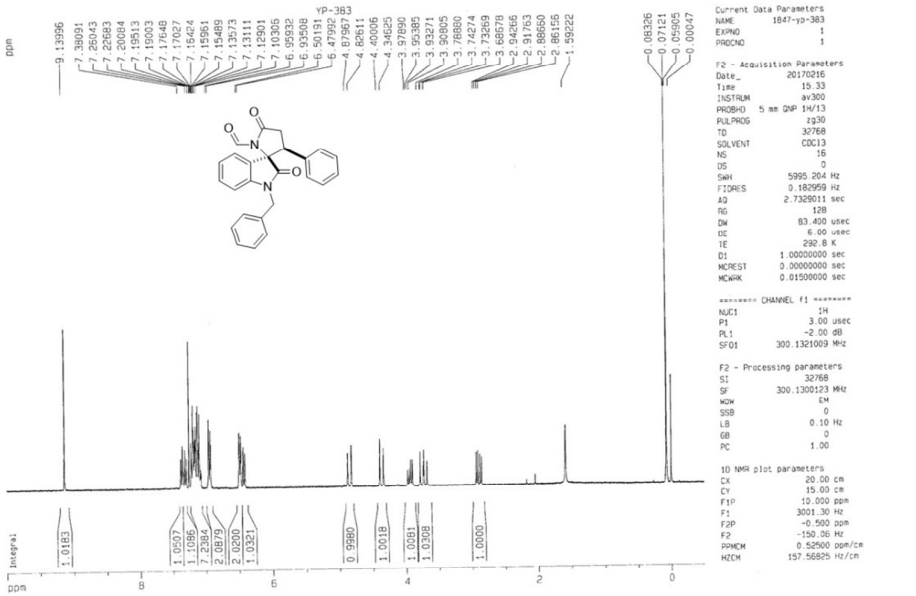


Figure S12: NMR spectra of compound **4k**



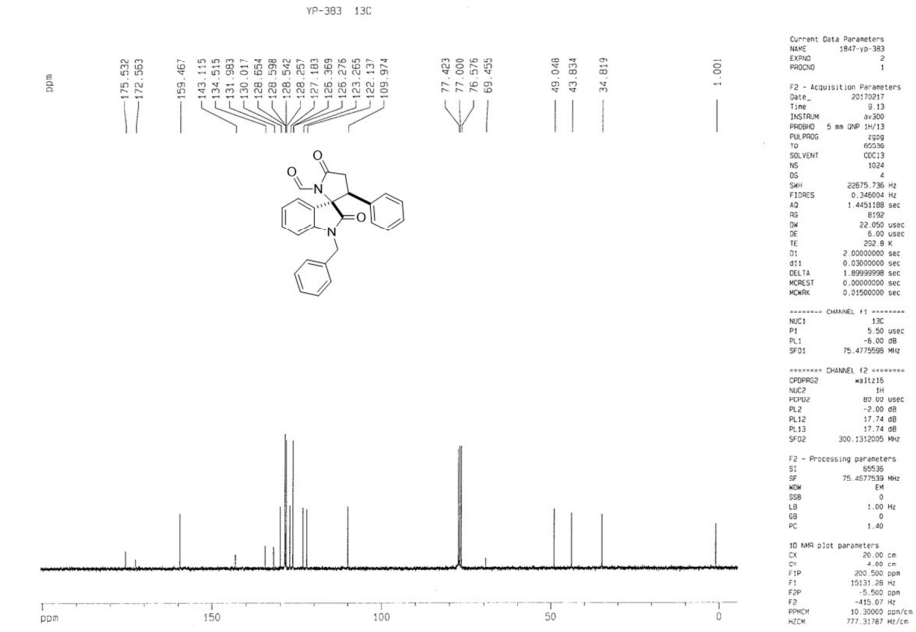
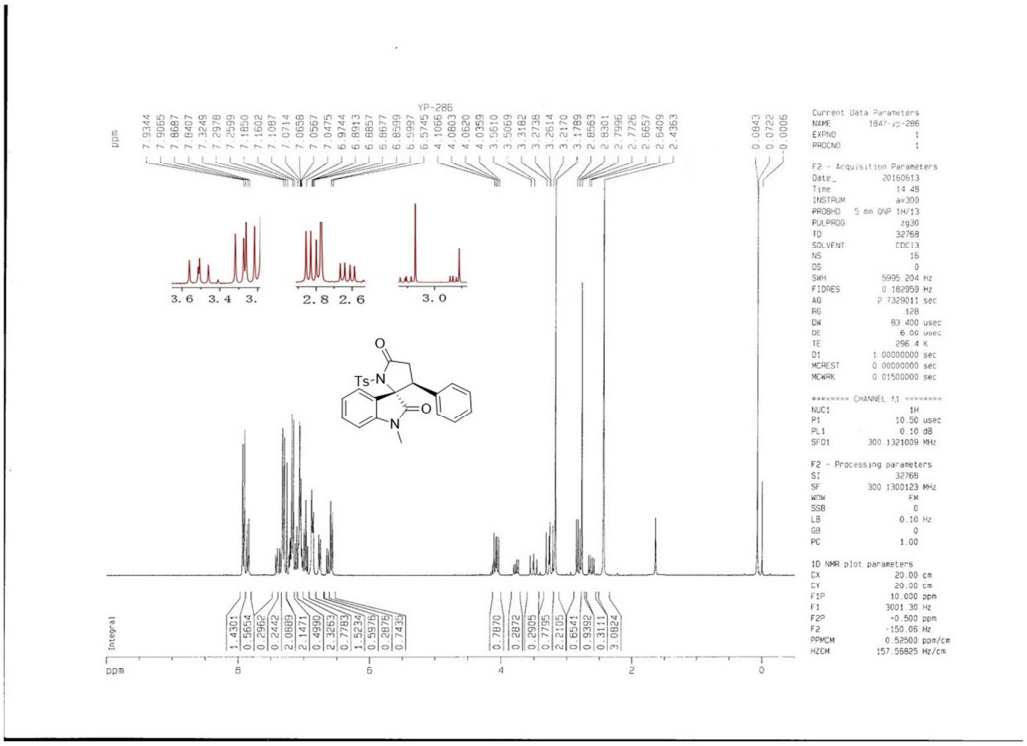


Figure S12': NMR spectra of compound 4k'



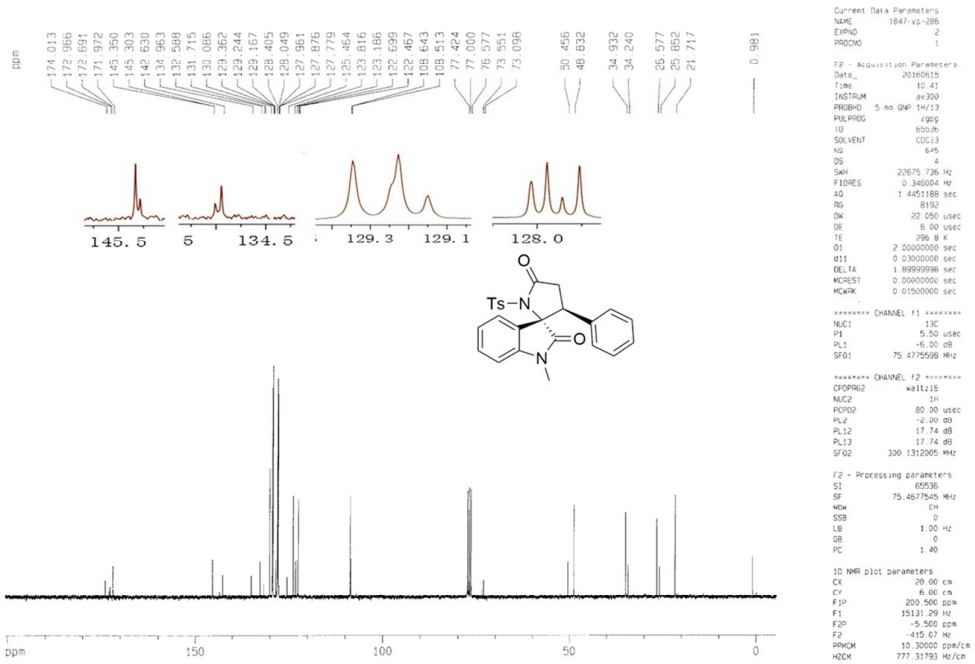


Figure S13: NMR spectra of compound **6a**

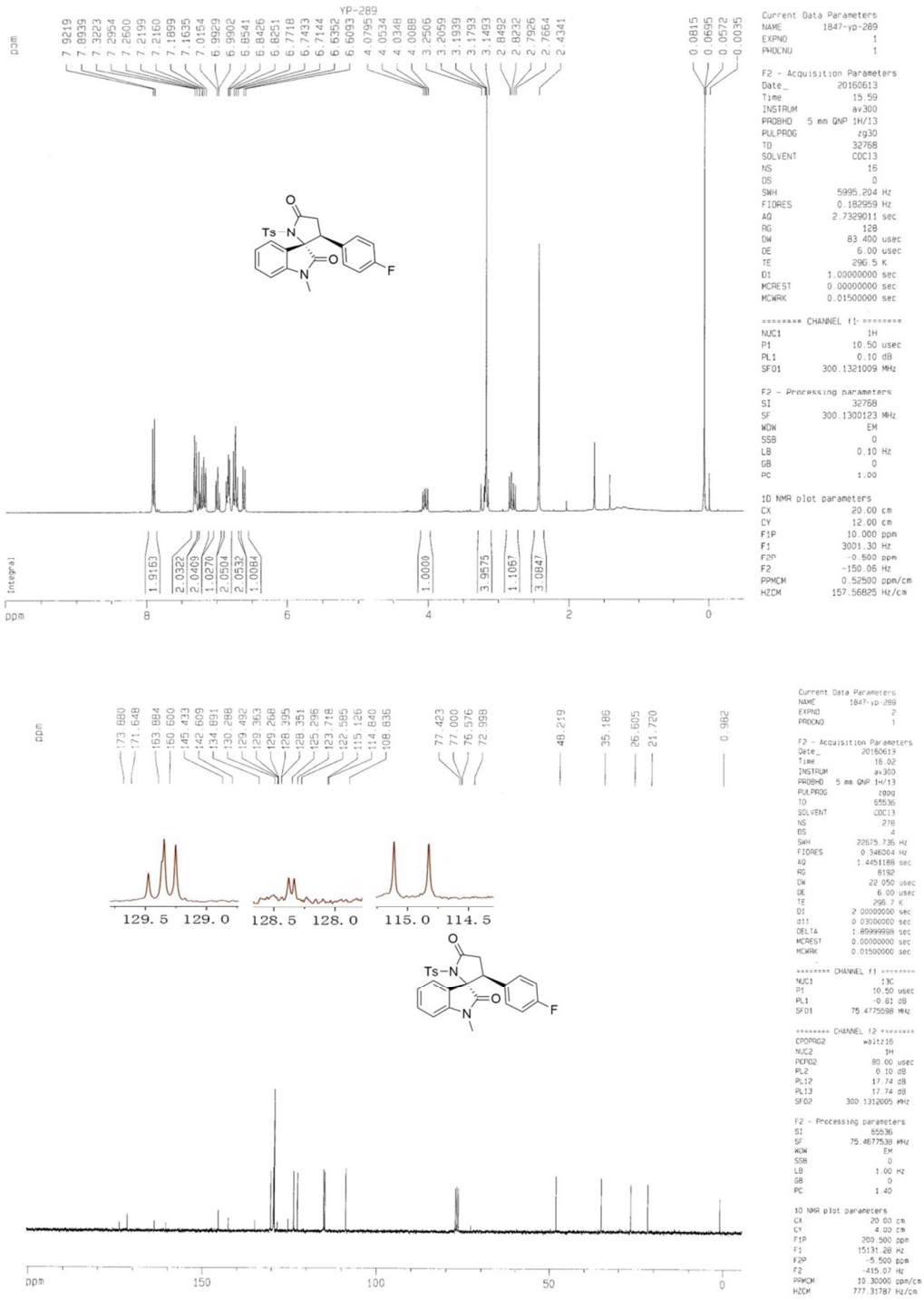
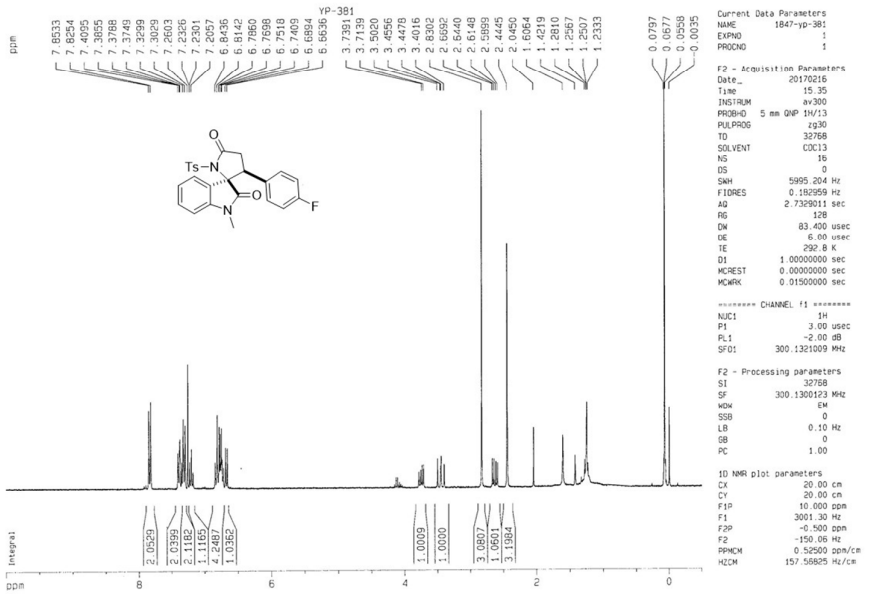


Figure S14: NMR spectra of compound **6b**



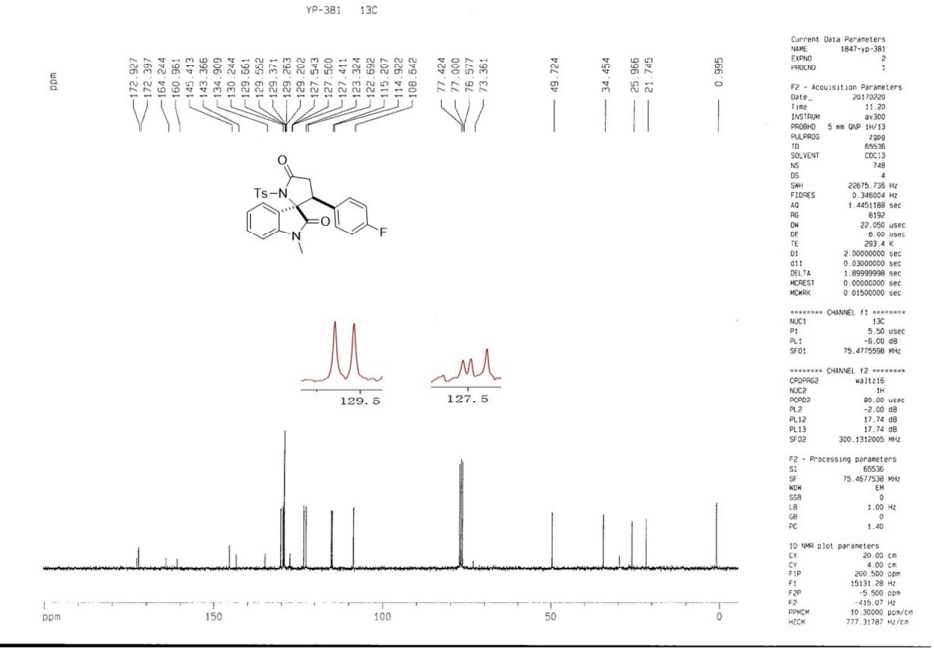
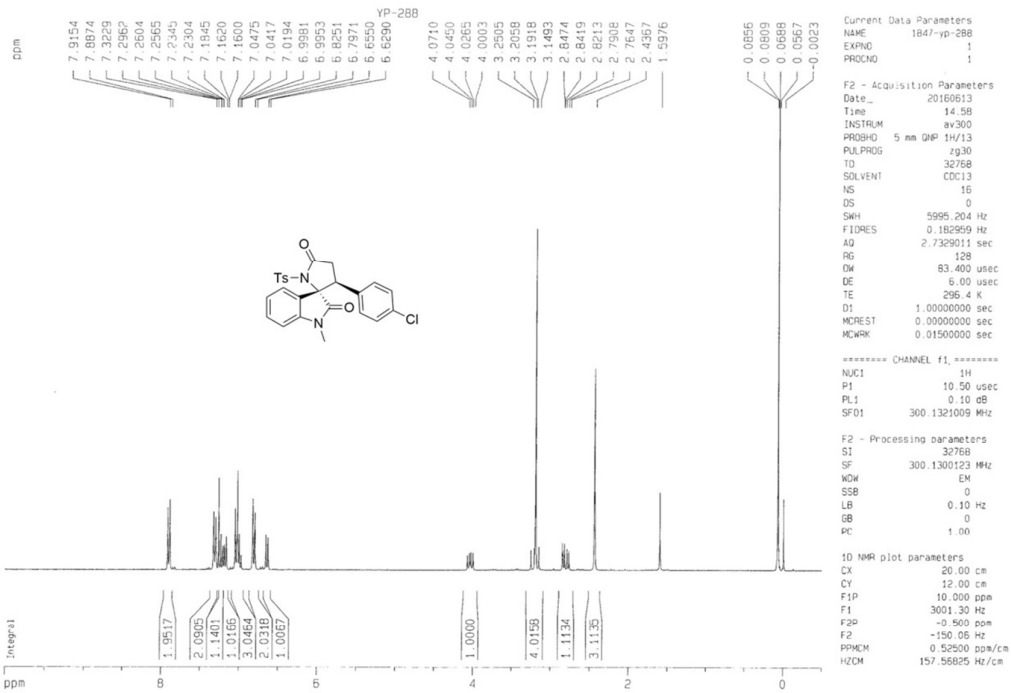


Figure S14': NMR spectra of compound 6b'



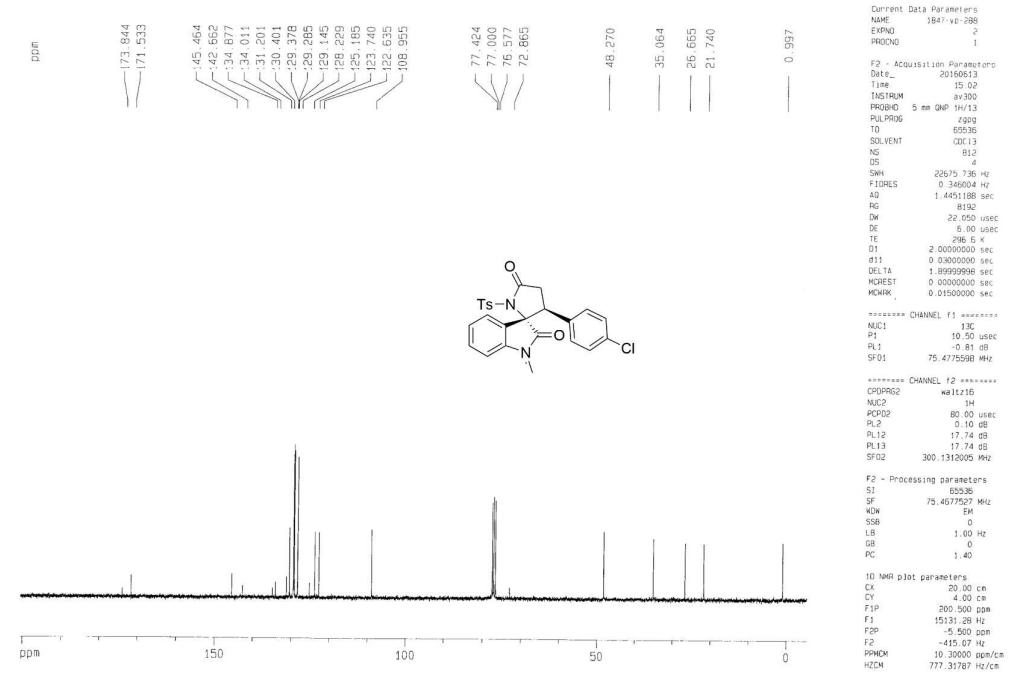
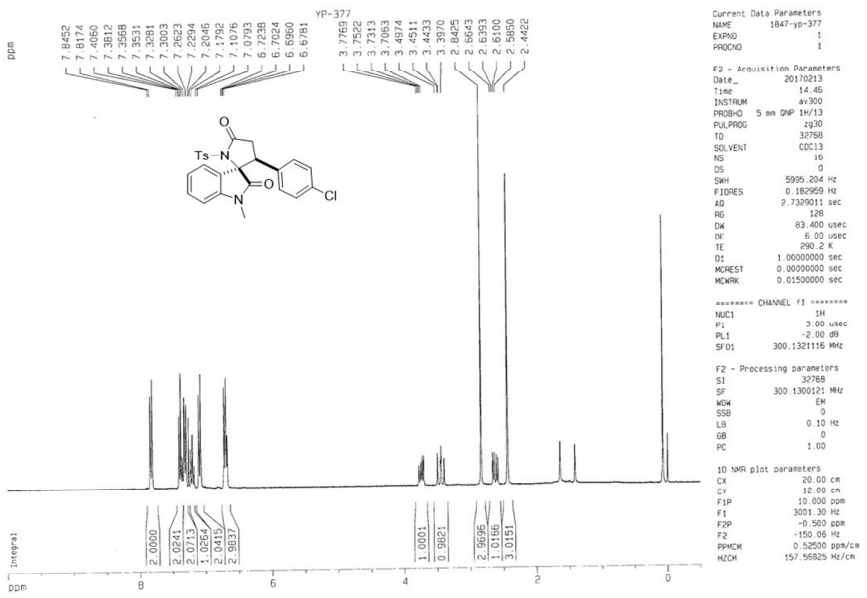


Figure S15: NMR spectra of compound **6c**



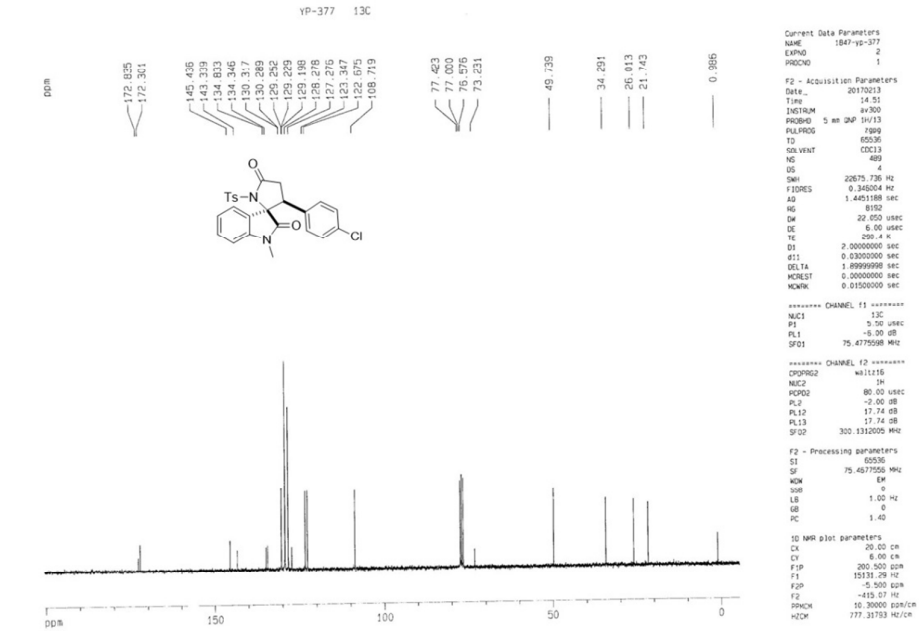


Figure S15': NMR spectra of compound 6c'

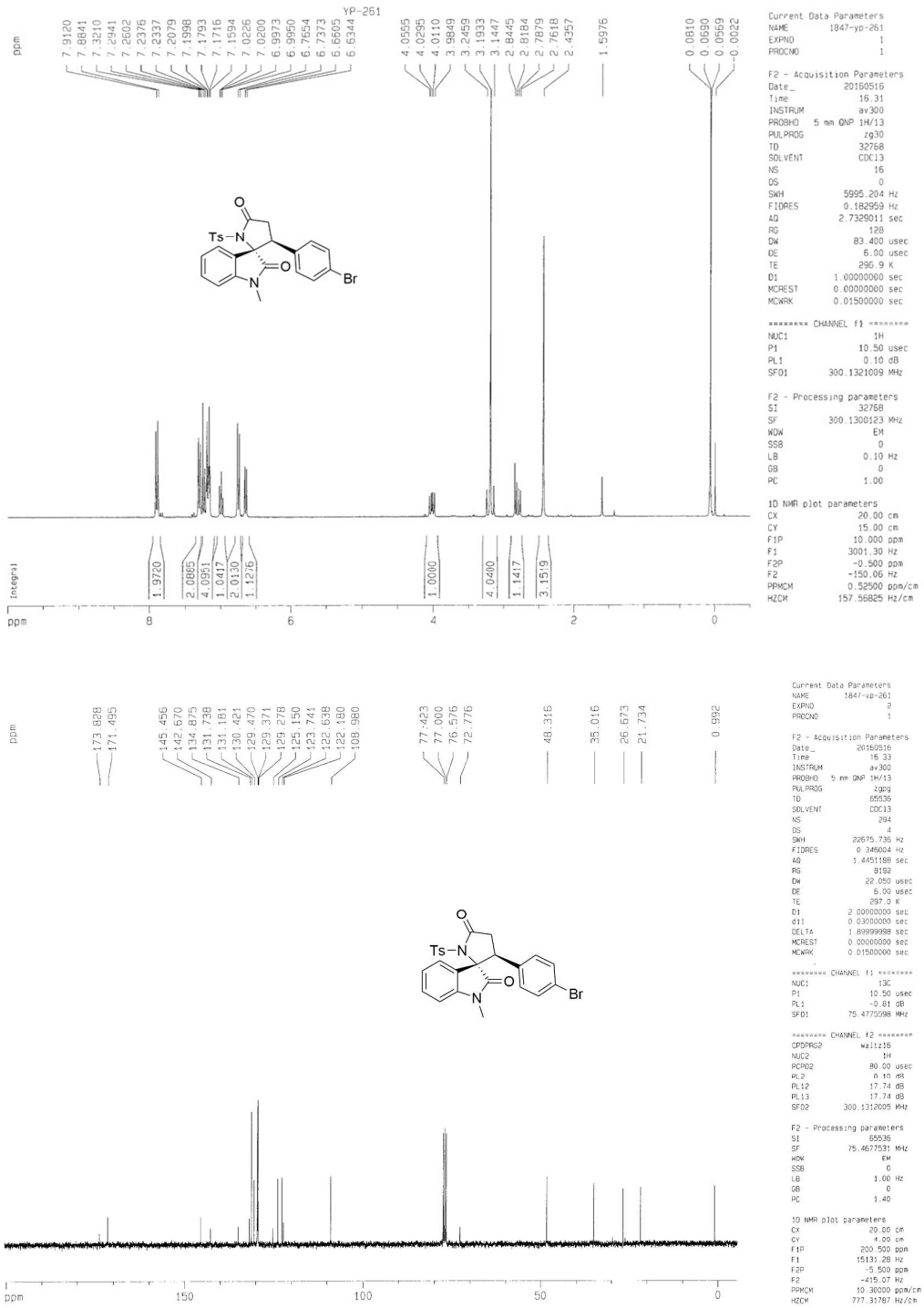
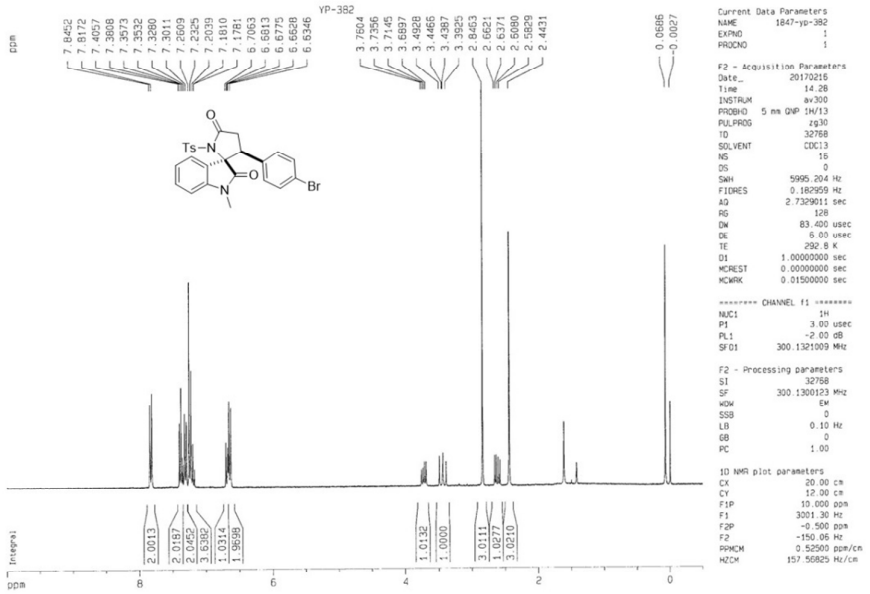


Figure S16: NMR spectra of compound **6d**



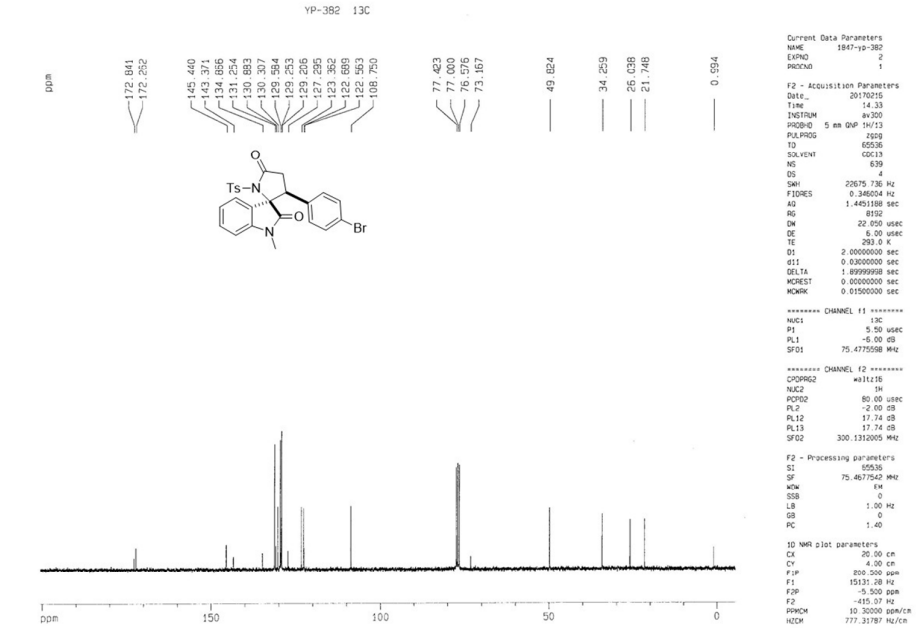


Figure S16': NMR spectra of compound 6d'

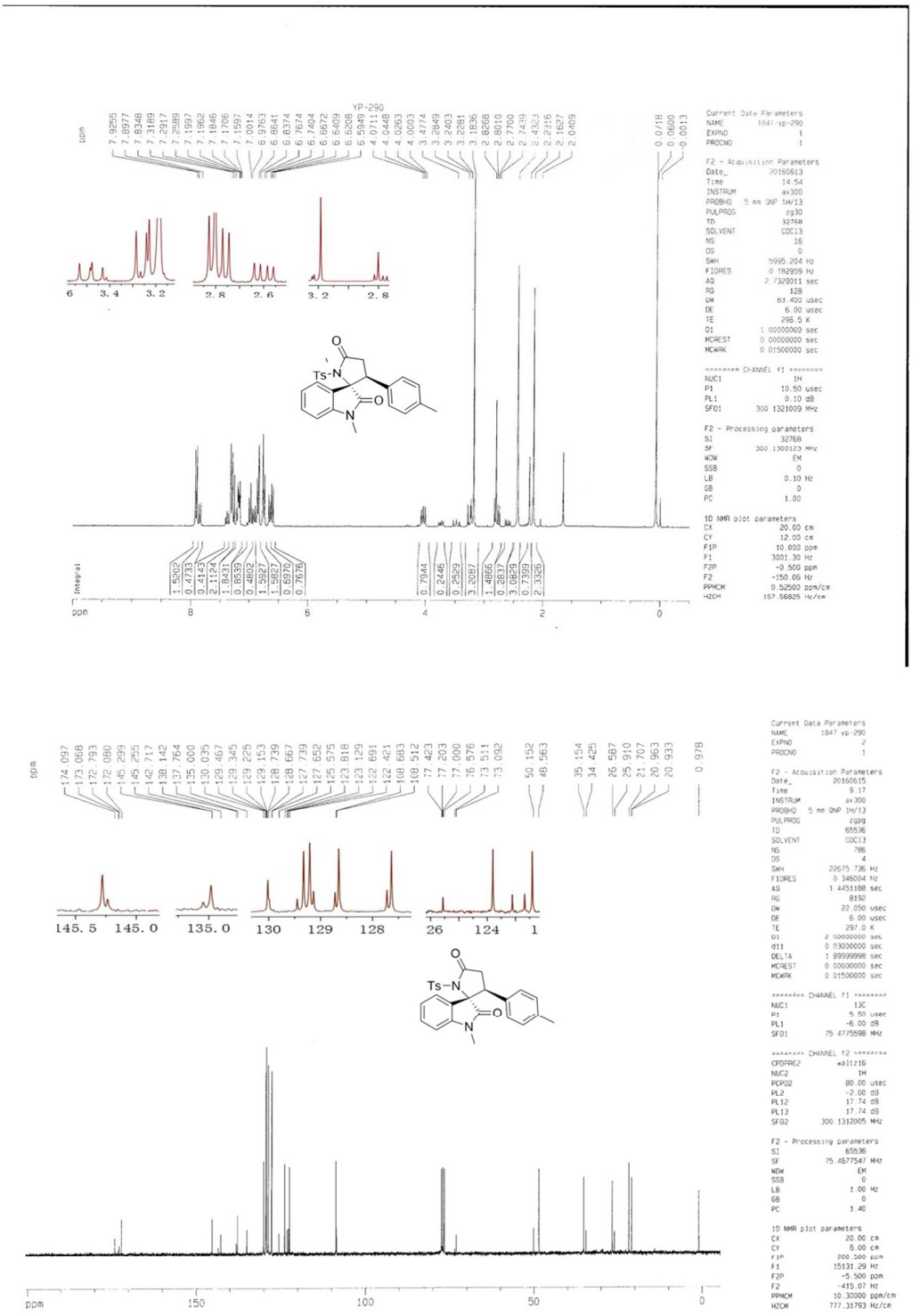


Figure S17: NMR spectra of compound **6e**

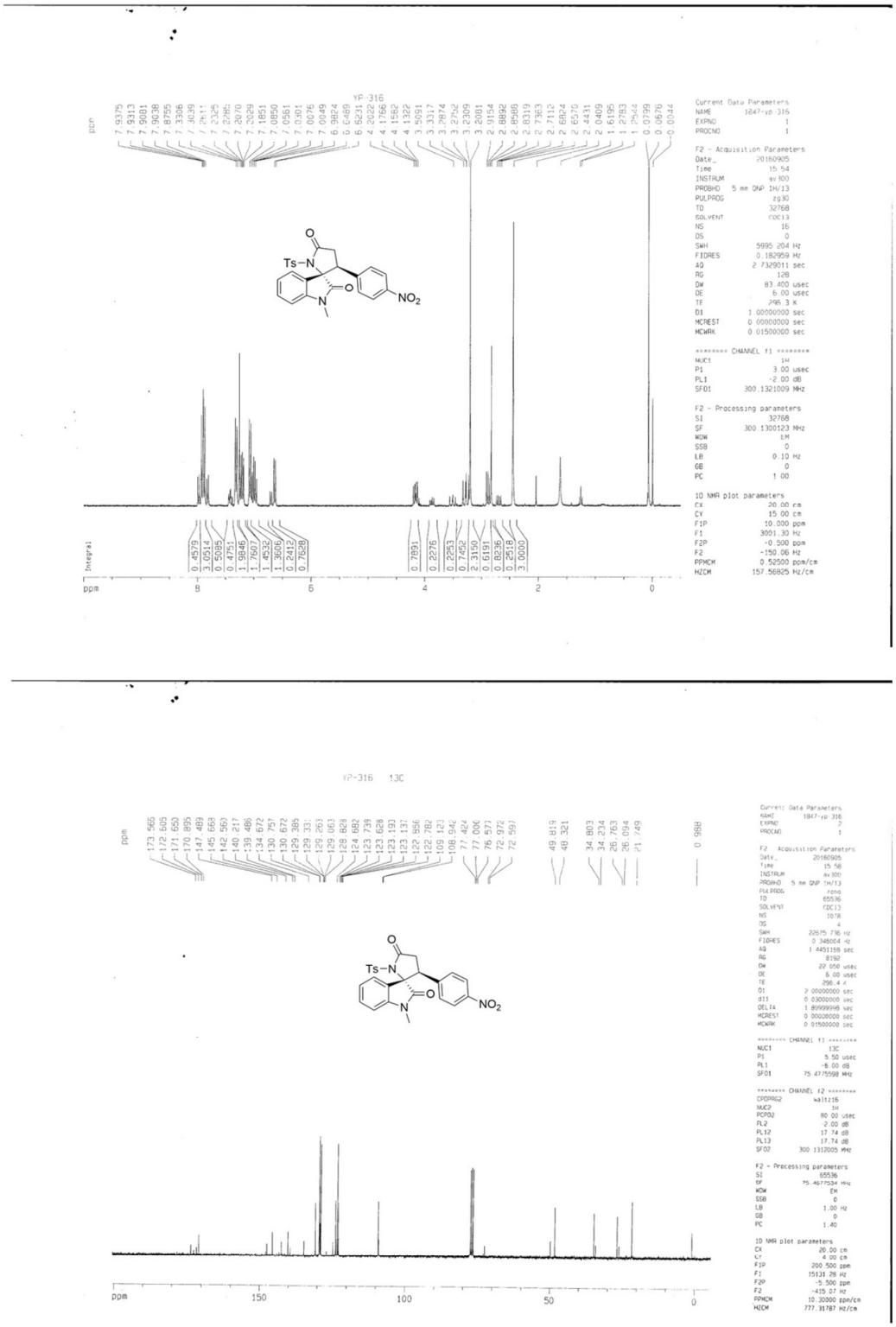


Figure S18: NMR spectra of compound **6f**

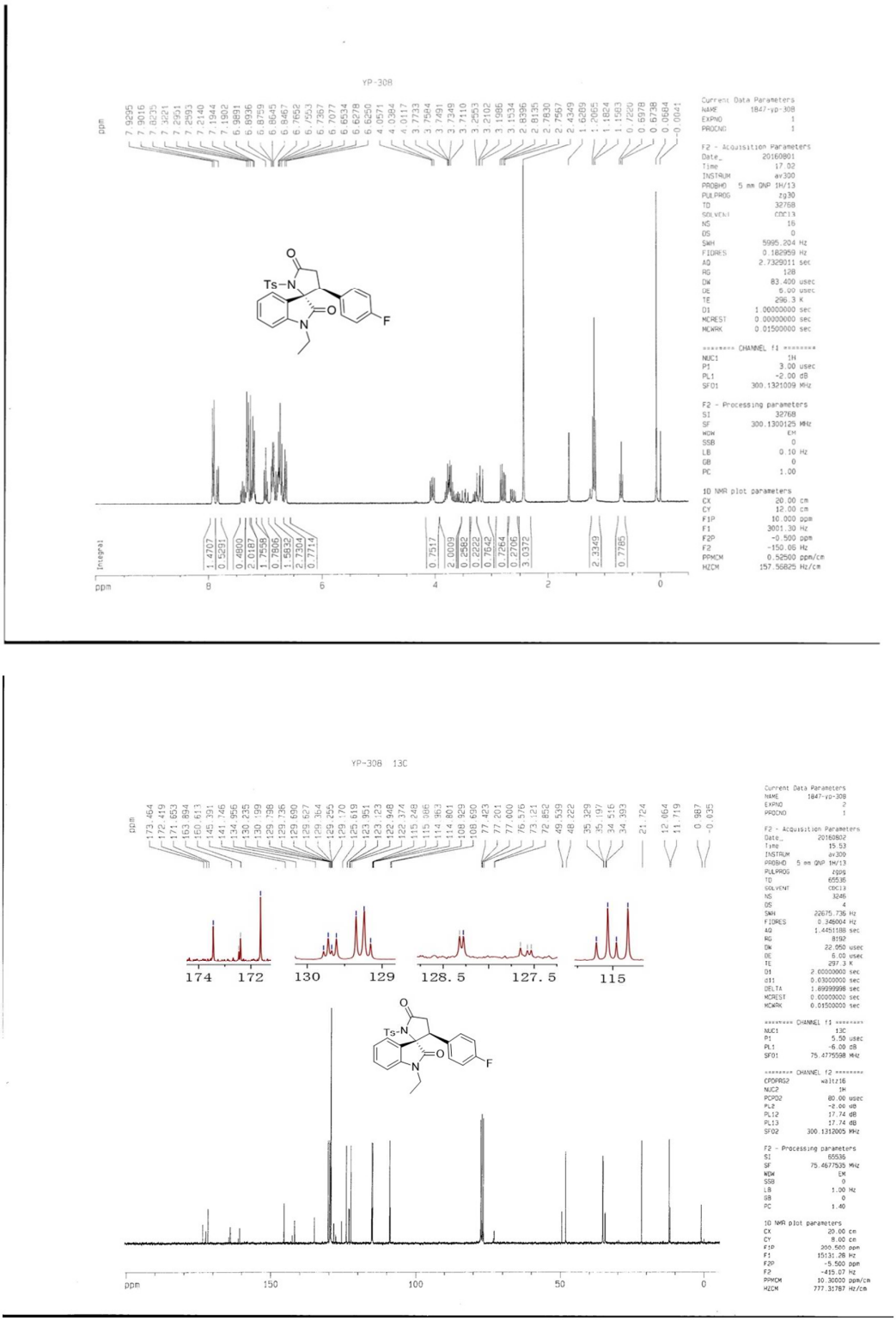
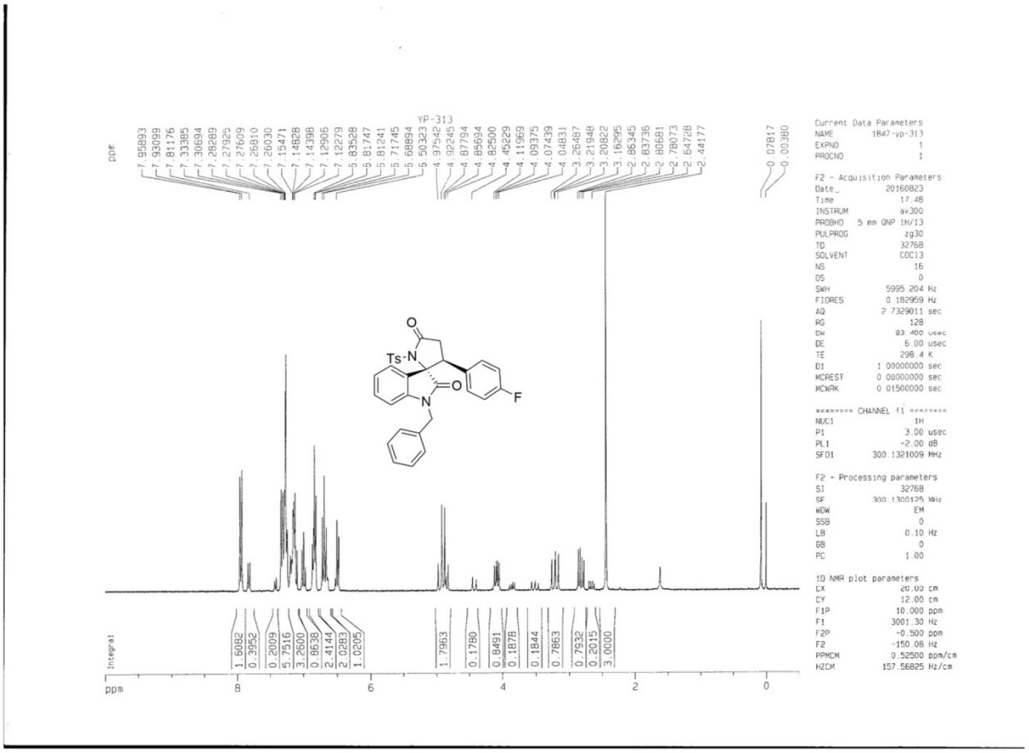


Figure S19: NMR spectra of compound **6g**



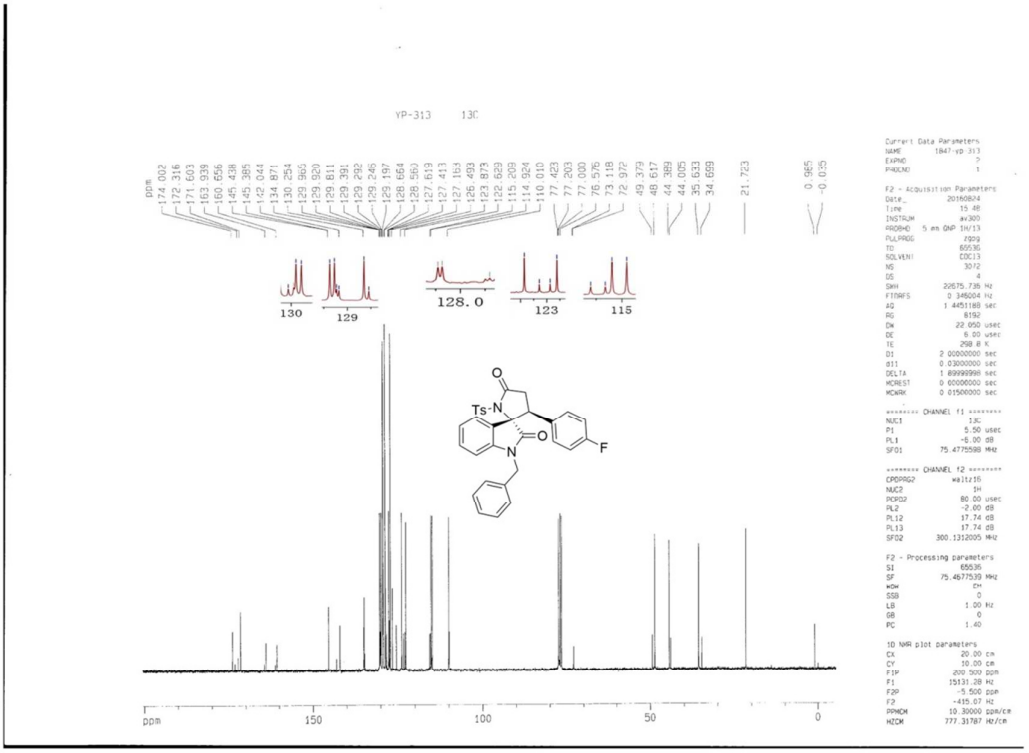
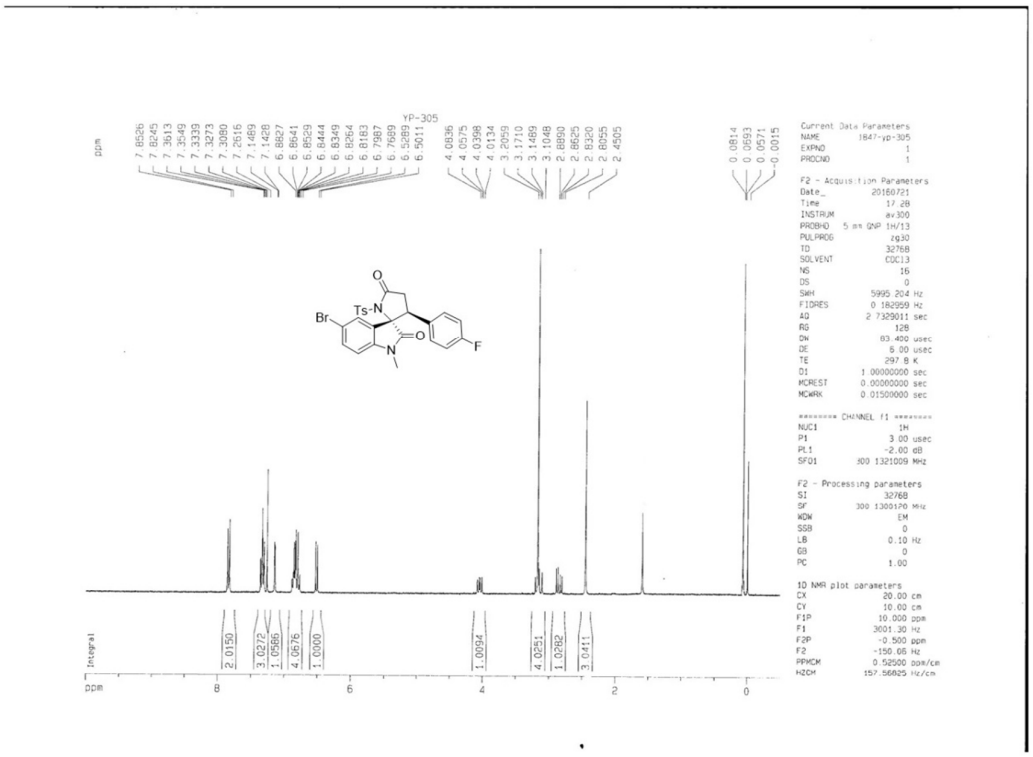


Figure S20: NMR spectra of compound **6h**



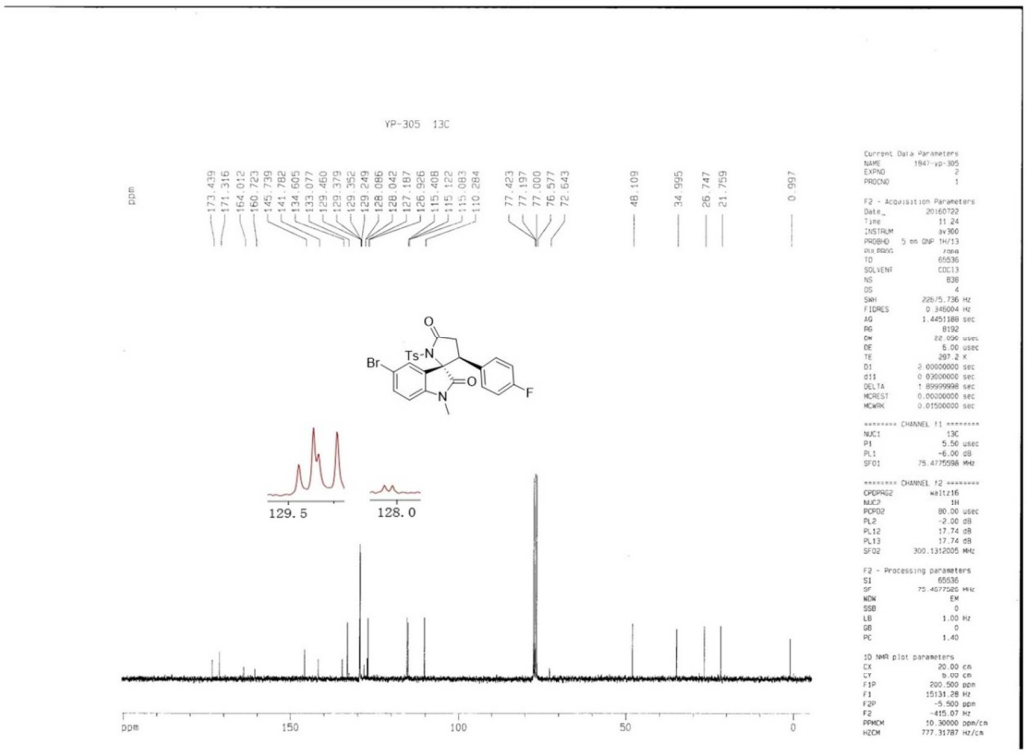
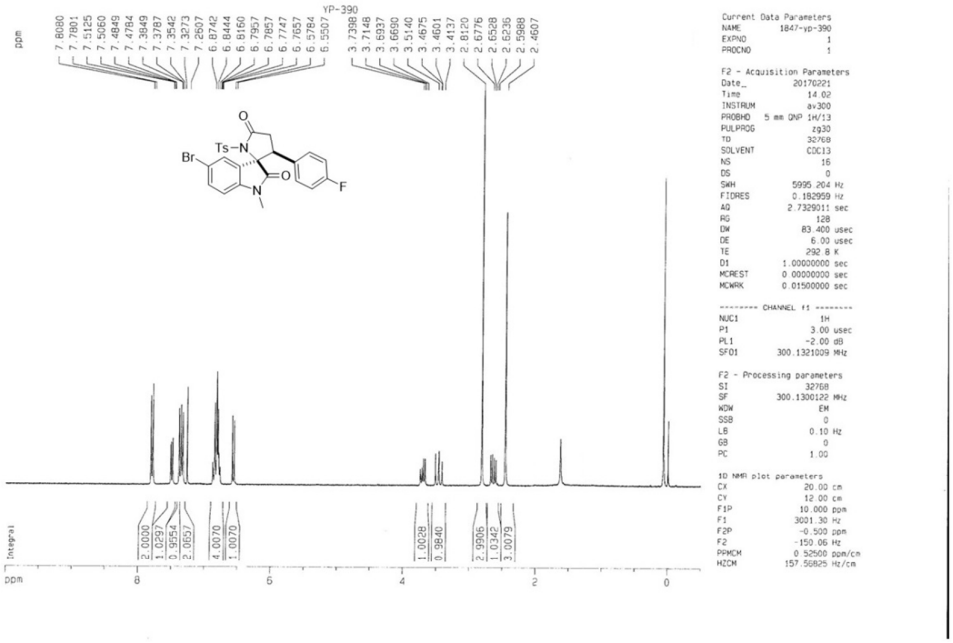


Figure S21: NMR spectra of compound **6i**



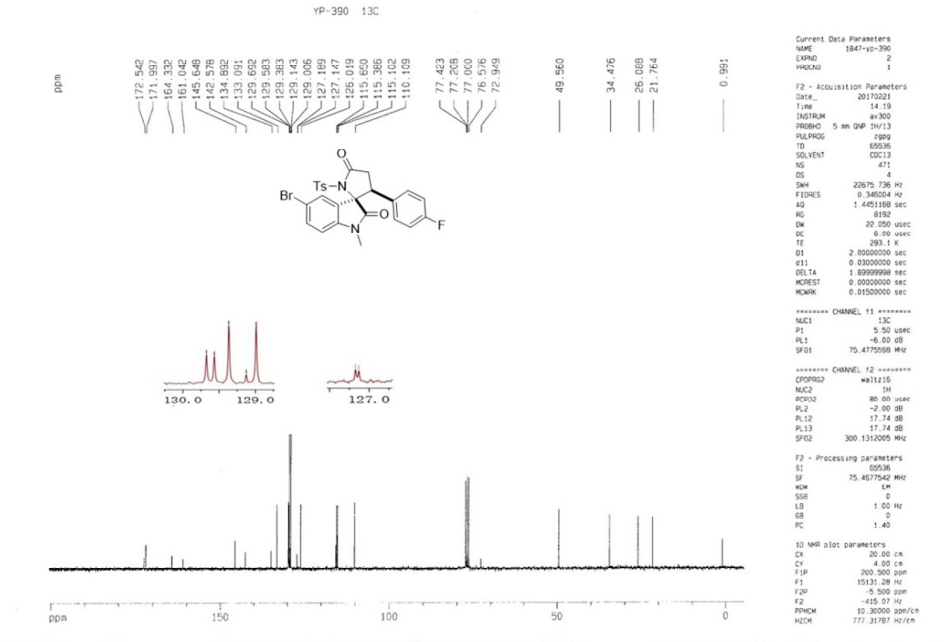
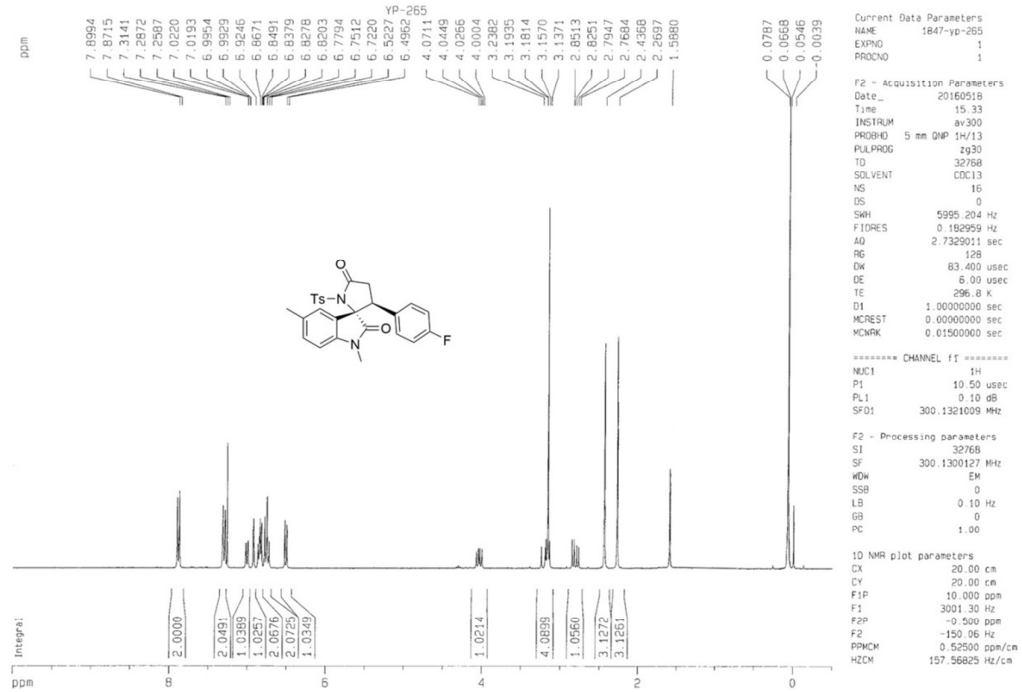


Figure S21': NMR spectra of compound 6i'



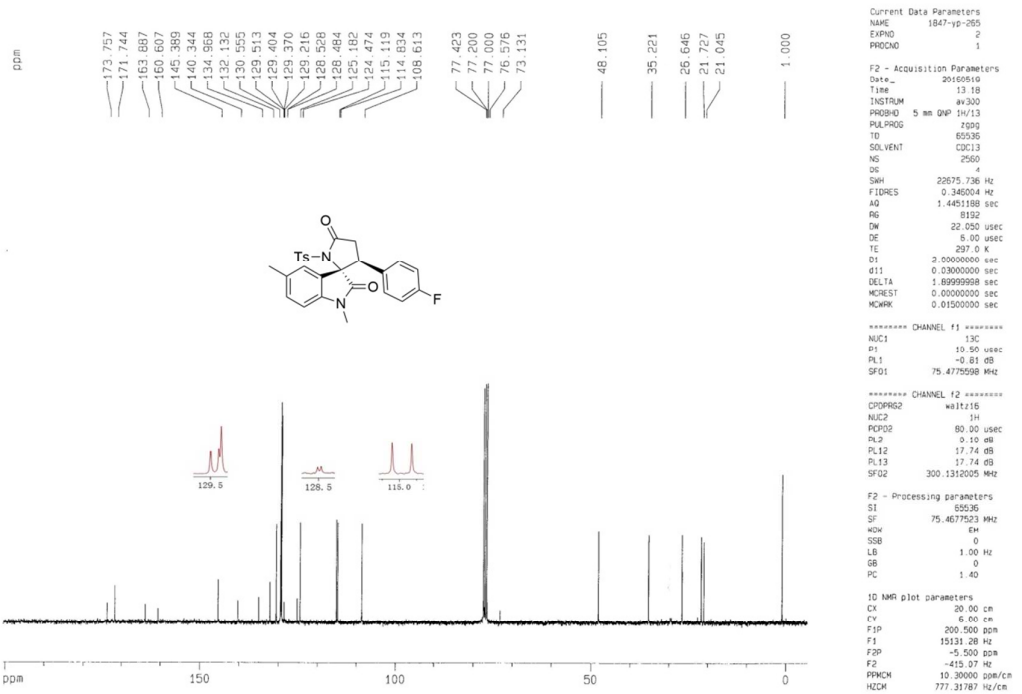
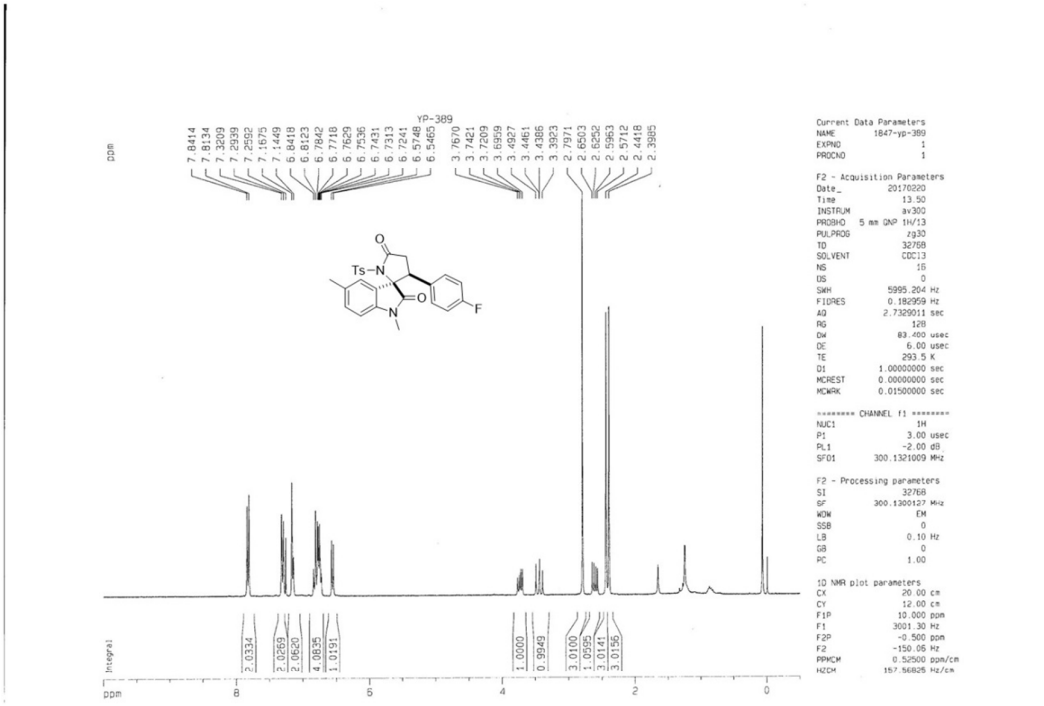


Figure S22: NMR spectra of compound **6j**



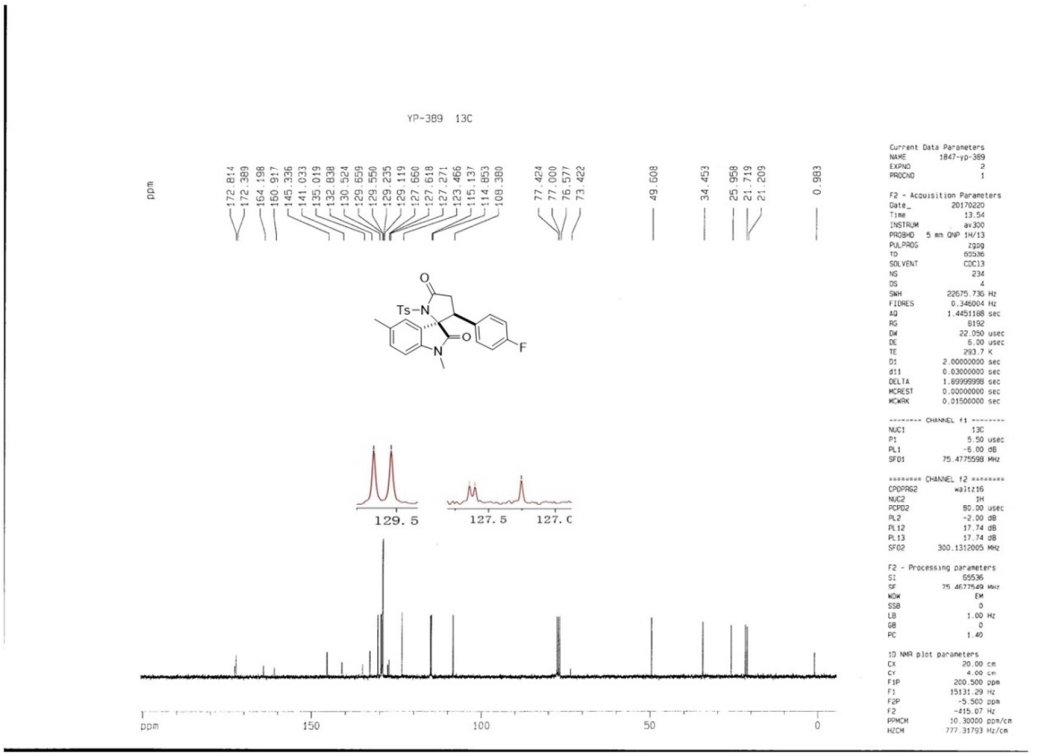
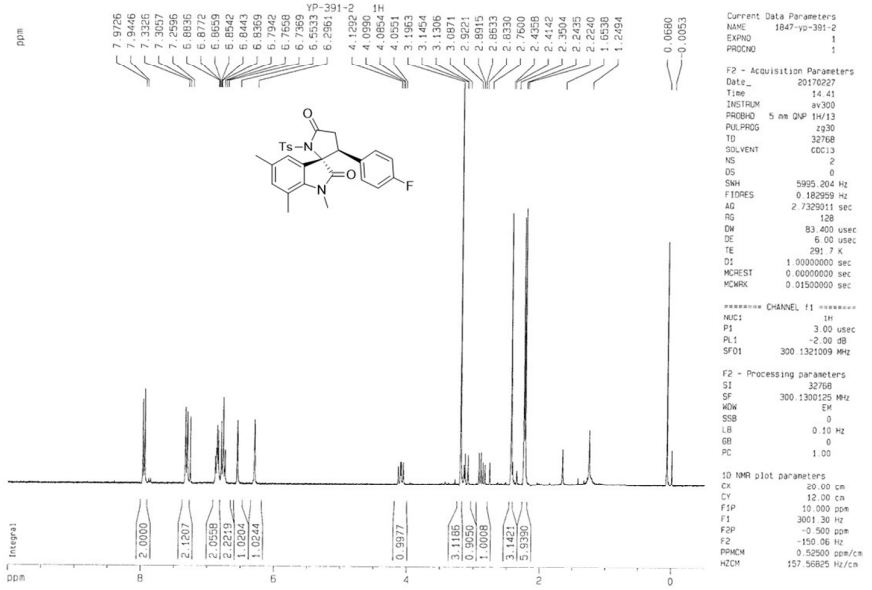


Figure S22': NMR spectra of compound 6j'



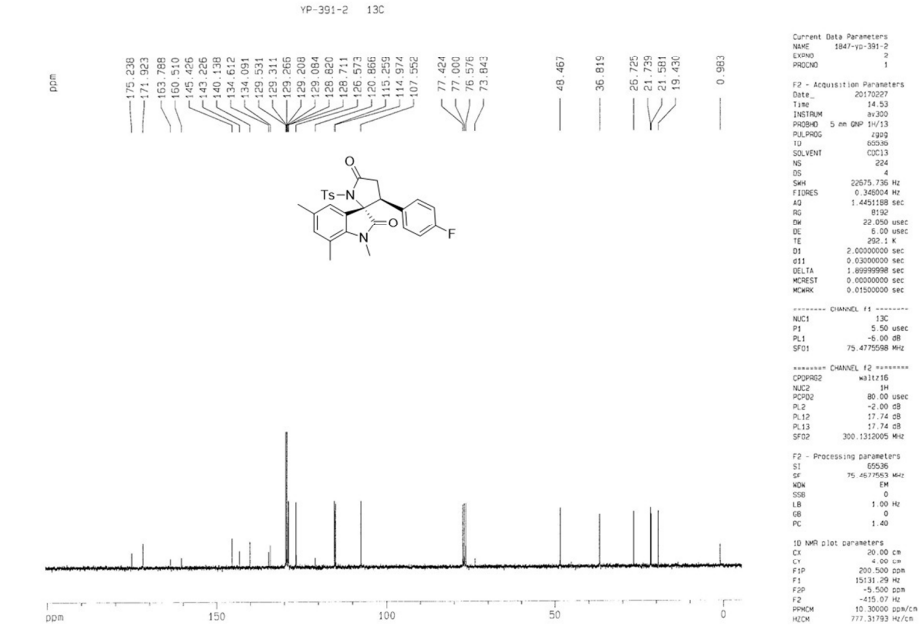
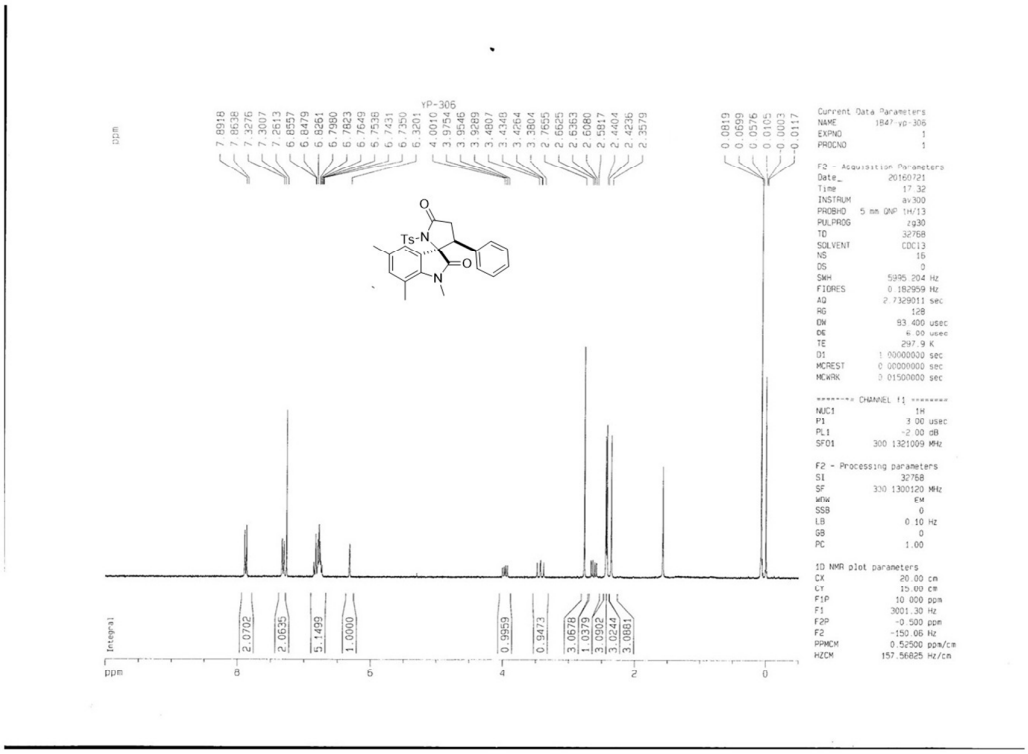


Figure S23: NMR spectra of compound **6k**



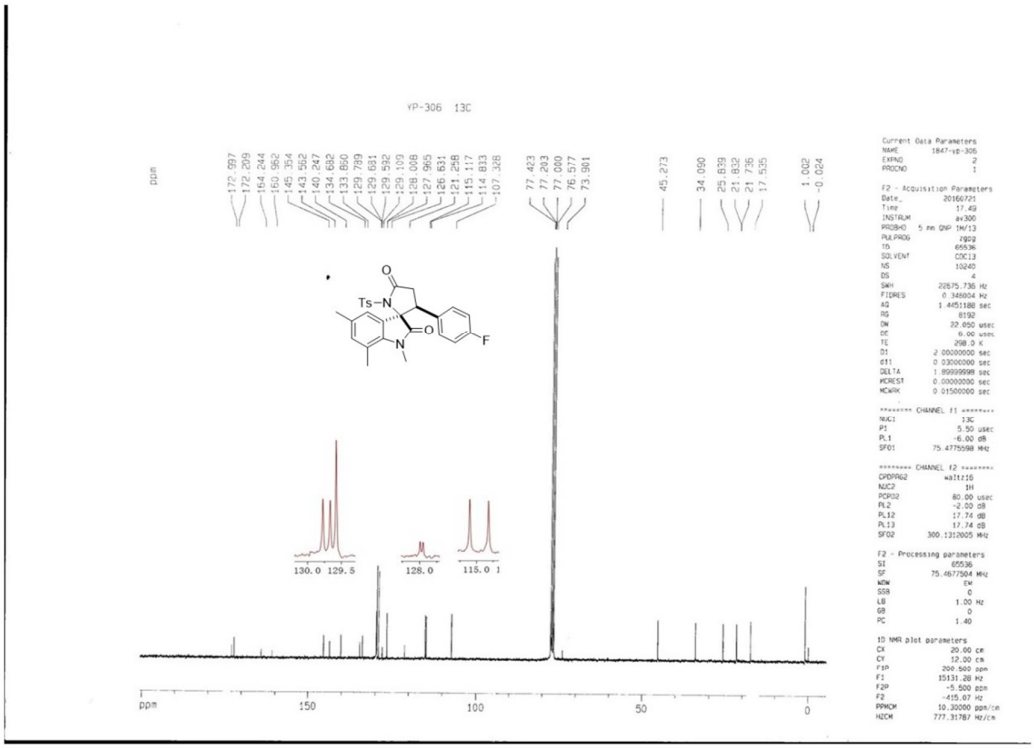
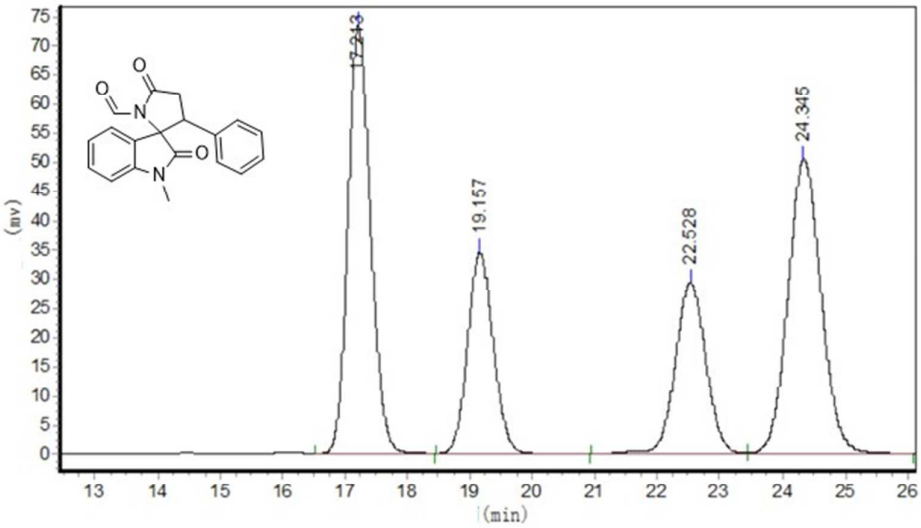
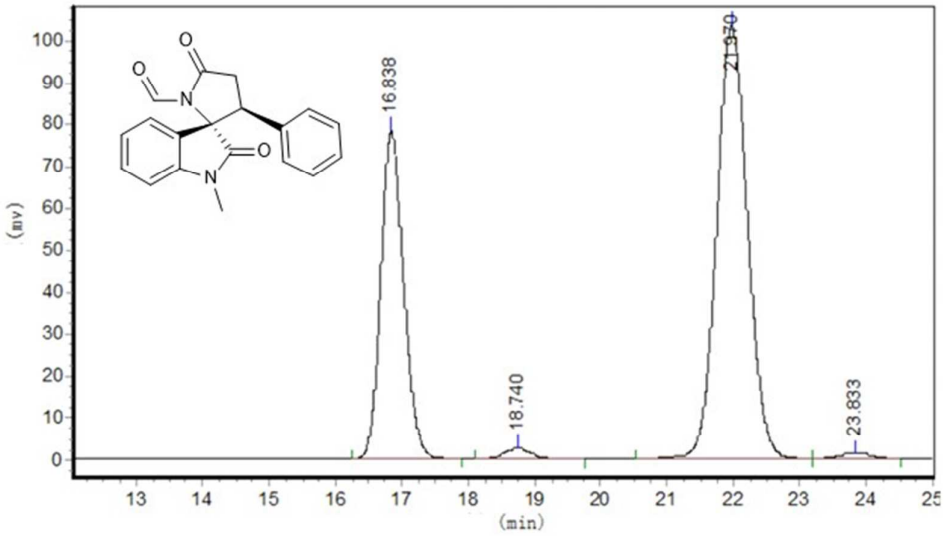


Figure S23': NMR spectra of compound 6k'

3. HPLC analysis of products 4 and 6

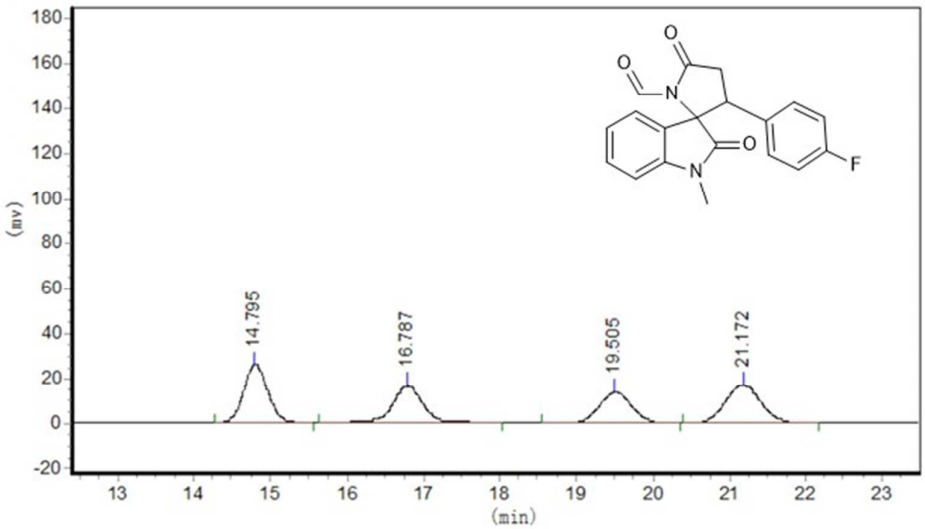


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	17.213	73445.633	1851213.250	32.3193
2	19.157	34596.184	993478.375	17.3446
3	22.528	29380.162	1008400.500	17.6051
4	24.345	50487.691	1874796.375	32.7310
Total		187909.670	5727888.500	100.0000

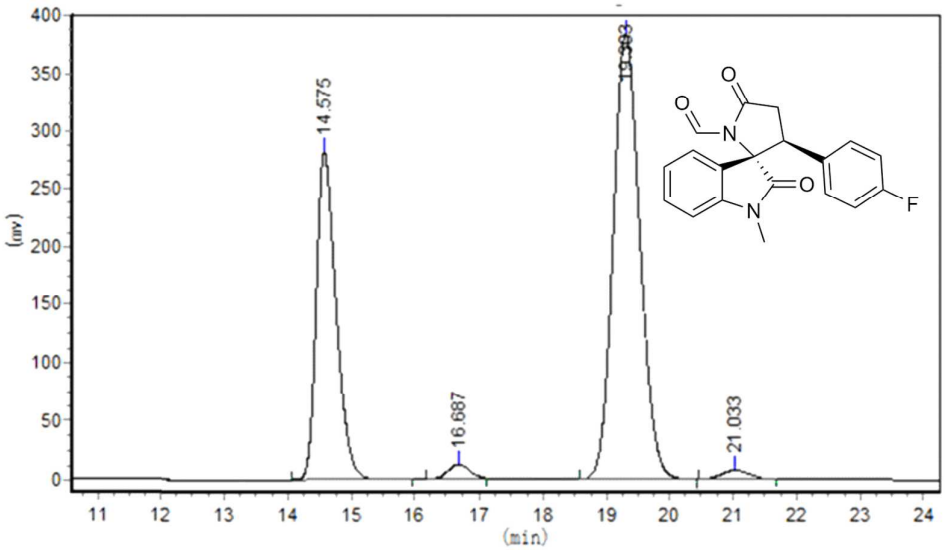


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	16.838	78304.508	1879712.250	35.2345
2	18.740	2433.828	67512.102	1.2655
3	21.970	103668.523	3345516.500	62.7104
4	23.833	1310.300	42124.098	0.7896
Total		185717.159	5334864.949	100.0000

Figure S24: HPLC analysis of the derivative of the compound **4a**

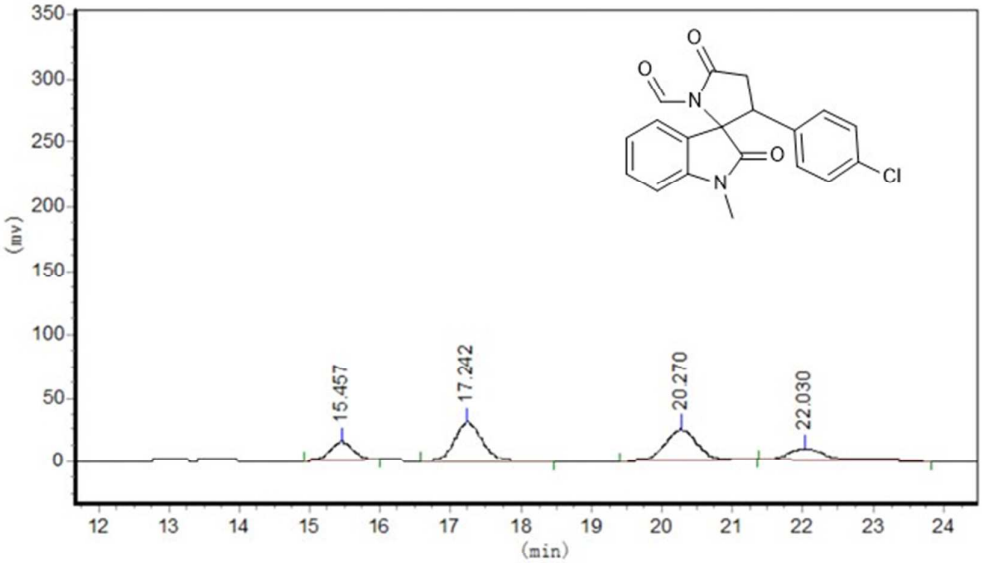


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	14.795	25748.697	555196.750	28.7037
2	16.787	16093.270	426824.344	22.0669
3	19.505	13712.387	401714.906	20.7687
4	21.172	17096.625	550495.500	28.4607
Total		72650.979	13533980.141	100.0000

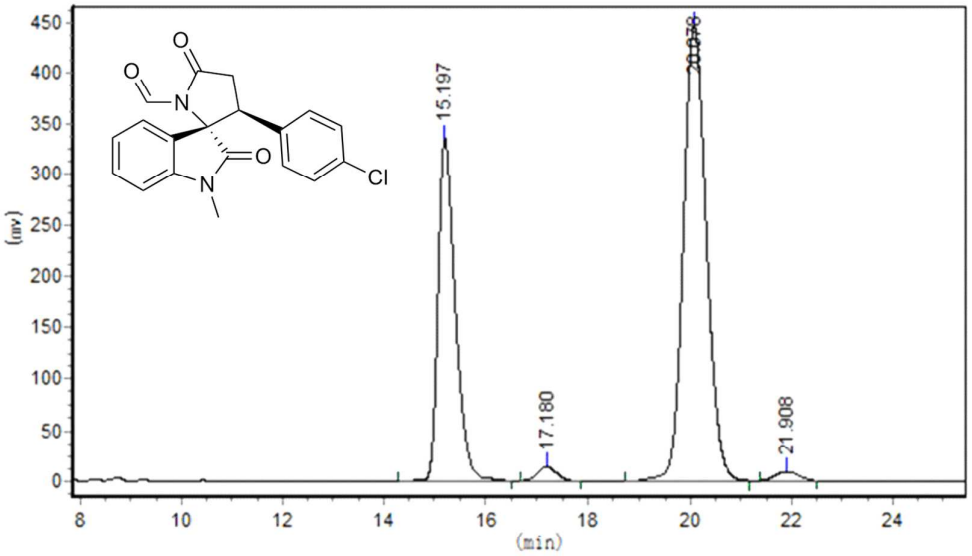


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	14.575	282204.406	6093865.000	34.2349
2	16.687	12134.232	280448.219	1.5755
3	19.303	383512.125	11203562.000	62.9409
4	21.033	7544.945	222259.703	1.2486
Total		685395.708	17800134.922	100.0000

Figure S25: HPLC analysis of the derivative of the compound **4b**

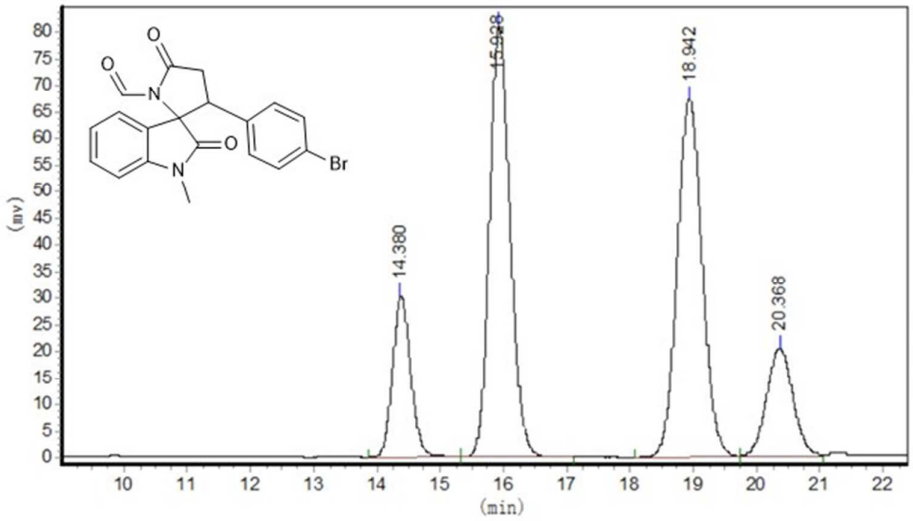


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	15.457	14451.496	326804.438	15.0272
2	17.242	29785.754	774582.438	35.6170
3	20.270	24167.598	748452.813	34.4155
4	22.030	9357.048	324913.594	14.9403
Total		77761.896	2174753.283	100.0000

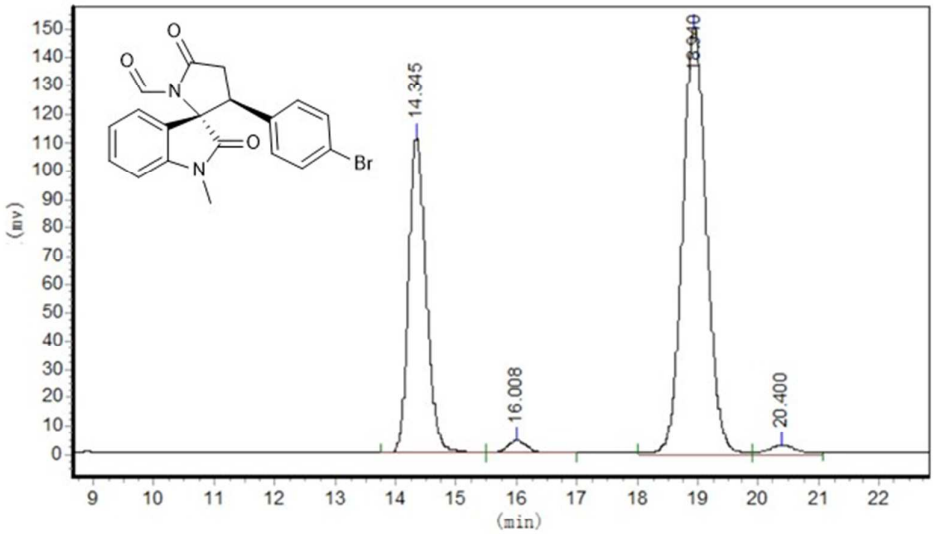


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	15.197	335088.656	7854816.500	34.9275
2	17.180	13507.081	349136.594	1.5525
3	20.078	445487.344	14017182.000	62.3294
4	21.908	8589.218	267749.500	1.1906
Total		802678.299	22488884.594	100.0000

Figure S26: HPLC analysis of the derivative of the compound **4c**

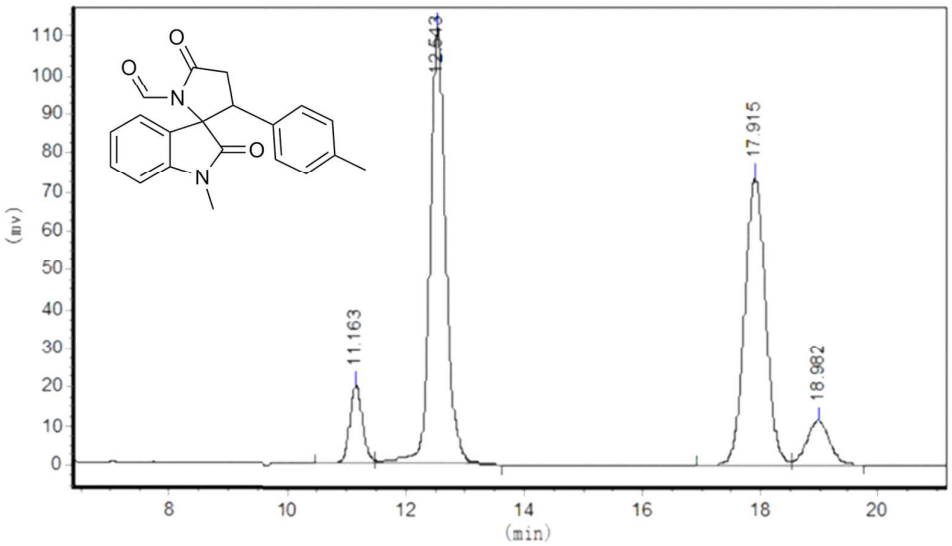


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	14.380	30107.139	605910.250	12.4034
2	15.928	81105.234	1846904.625	37.8073
3	18.942	67511.133	1844419.000	37.7564
4	20.368	20302.709	587818.563	12.0330
Total		199026.215	4885052.438	100.0000

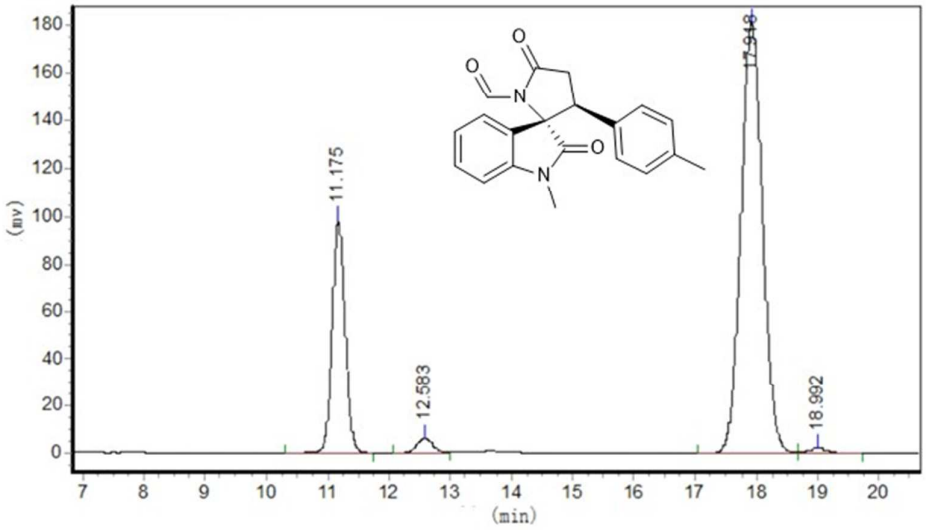


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	14.345	111885.188	2274407.500	34.2727
2	16.008	4605.408	109633.008	1.6520
3	18.940	150914.391	4164859.250	62.7597
4	20.400	2857.247	87302.617	1.3156
Total		270262.233	6636202.375	100.0000

Figure S27: HPLC analysis of the derivative of the compound **4d**

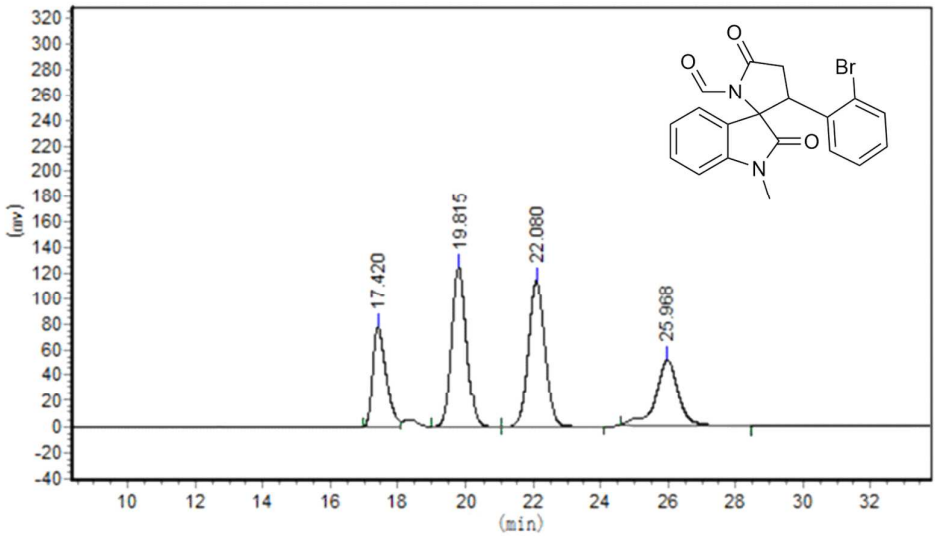


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	11.163	20409.104	308482.375	8.5316
2	12.543	111702.602	1799282.000	41.0805
3	17.915	73479.258	1810462.375	41.9890
4	18.982	11529.353	302524.156	8.3989
Total		202826.276	4220750.906	100.0000

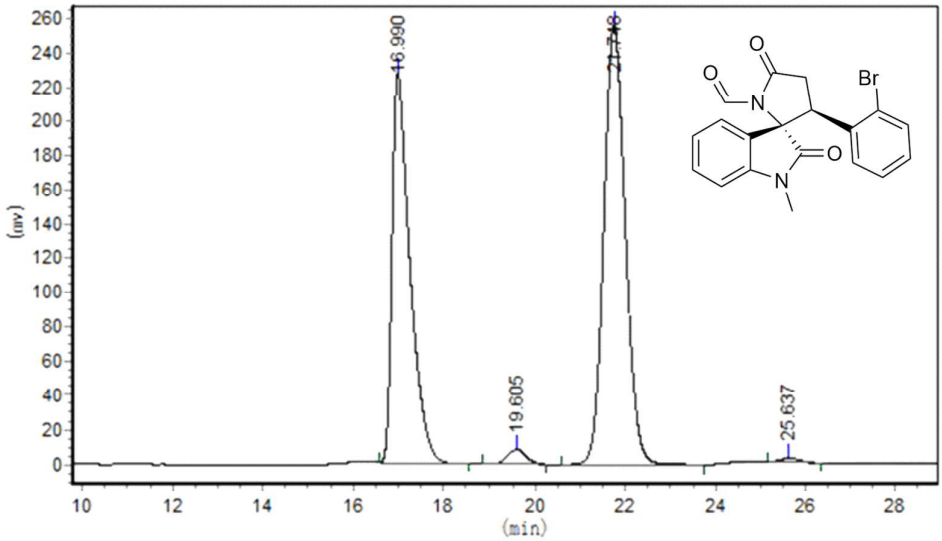


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	11.175	97848.953	1442825.375	23.7075
2	12.583	6141.767	100521.406	1.6517
3	17.918	181476.000	4485496.500	73.7027
4	18.992	2111.000	57091.398	0.9381
Total		287577.720	6085934.680	100.0000

Figure S28: HPLC analysis of the derivative of the compound **4e**

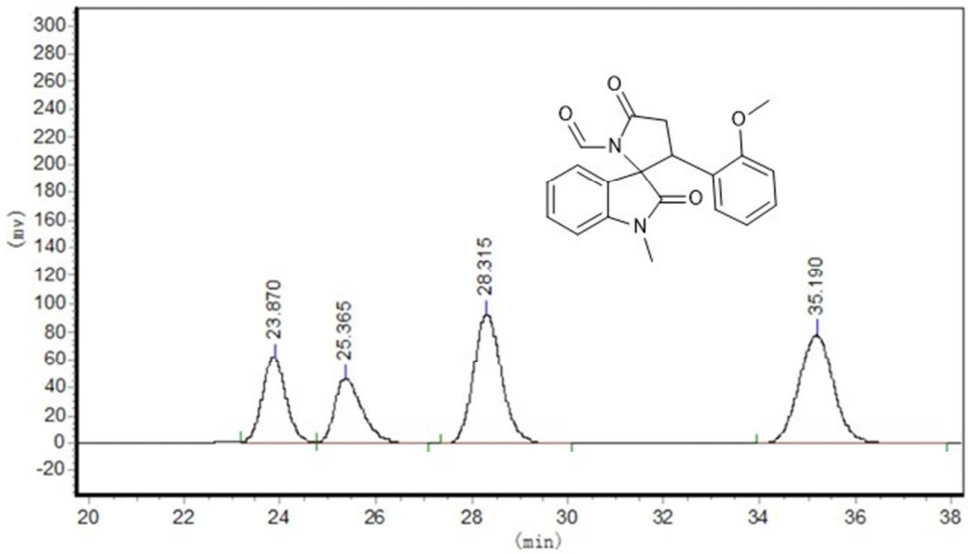


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	17.420	77811.180	2096913.750	17.6518
2	19.815	124560.188	3703499.000	31.1760
3	22.080	113664.227	3860820.250	32.5004
4	25.968	50940.453	2218076.750	18.6718
Total		366976.047	11879309.750	100.0000

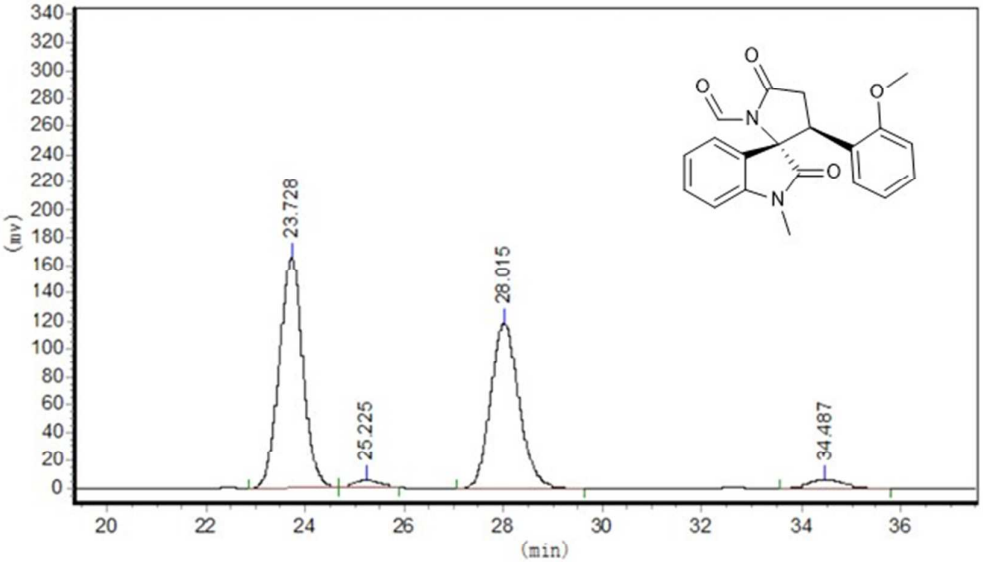


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	16.990	228317.938	6284918.500	41.8443
2	19.605	8210.947	230362.297	1.5337
3	21.748	255655.359	8418613.000	56.0502
4	25.637	2724.500	85885.789	0.5718
Total		494908.745	15019779.586	100.0000

Figure S29: HPLC analysis of the derivative of the compound **4f**

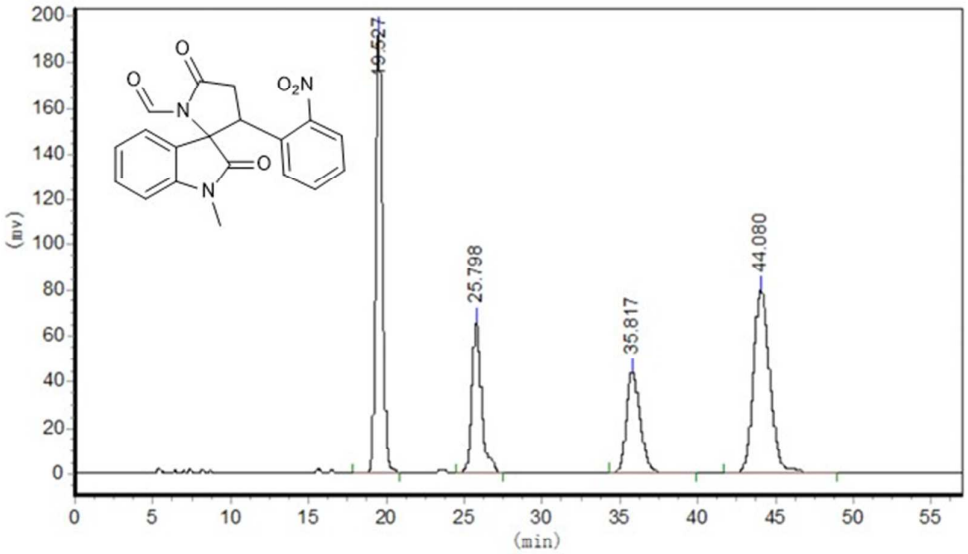


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	23.870	60463.387	1995649.000	17.4929
2	25.365	46078.570	1781505.000	15.6158
3	28.315	92504.430	3749179.750	32.8636
4	35.190	77579.000	3881988.000	34.0277
Total		276625.387	11408321.750	100.0000

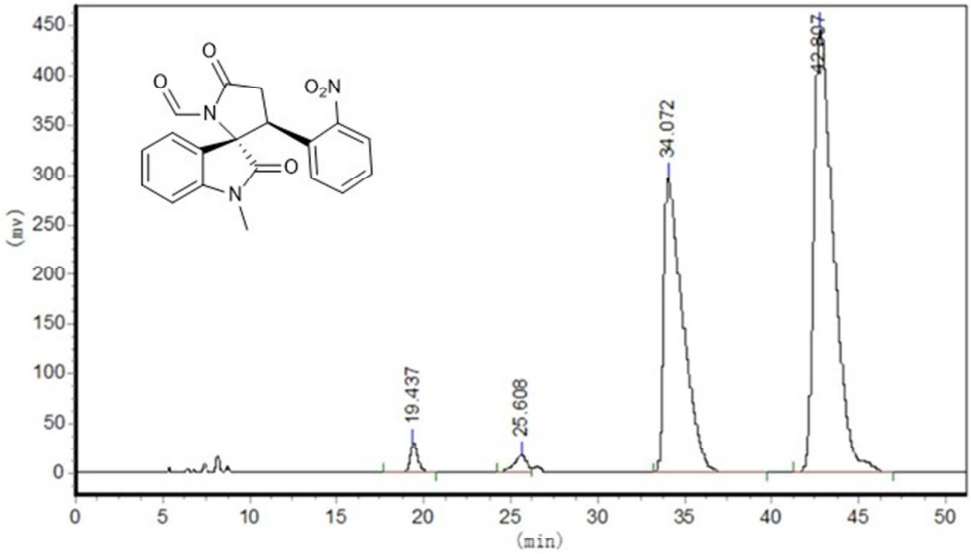


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	23.728	165747.203	5485291.500	51.3356
2	25.225	5656.745	196165.203	1.8359
3	28.015	118481.734	4690816.500	43.9003
4	34.487	6612.722	312895.094	2.9283
Total		296498.404	10685168.297	100.0000

Figure S30: HPLC analysis of the derivative of the compound **4g**

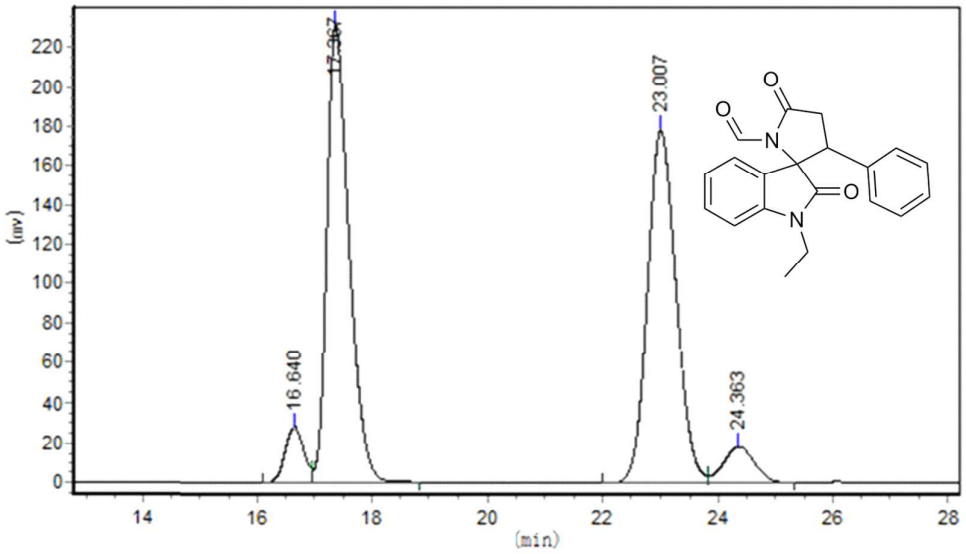


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	19.527	193489.078	5820158.000	33.6290
2	25.798	66243.203	2913718.750	16.8355
3	35.817	43878.266	2721077.750	15.7224
4	44.080	79752.078	5852008.500	33.8130
Total		383362.625	17306963.000	100.0000

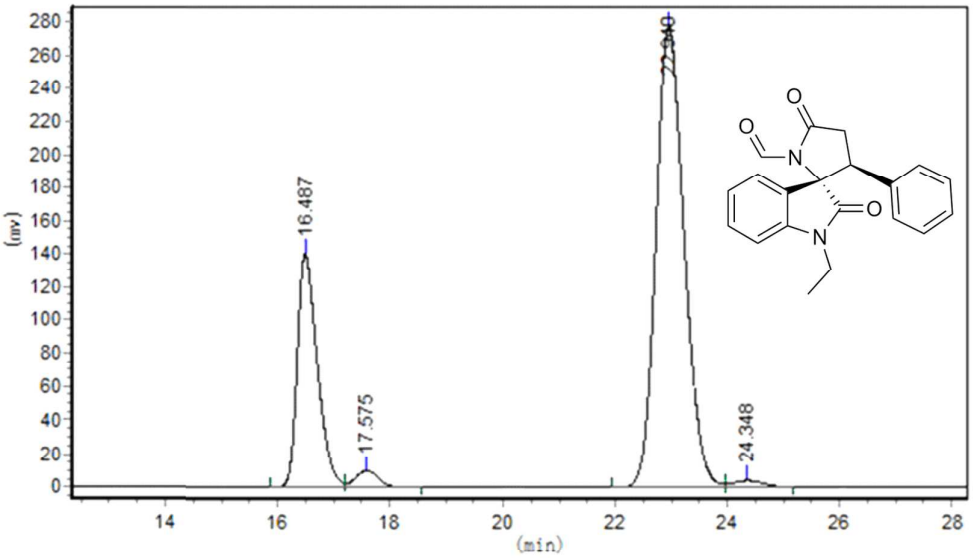


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	19.437	28512.357	874215.250	1.4675
2	25.608	17253.621	872670.188	1.4649
3	34.072	297066.281	22471314.000	37.7210
4	42.807	446864.063	35354240.000	59.3466
Total		789696.322	59572439.438	100.0000

Figure S31: HPLC analysis of the derivative of the compound **4h**

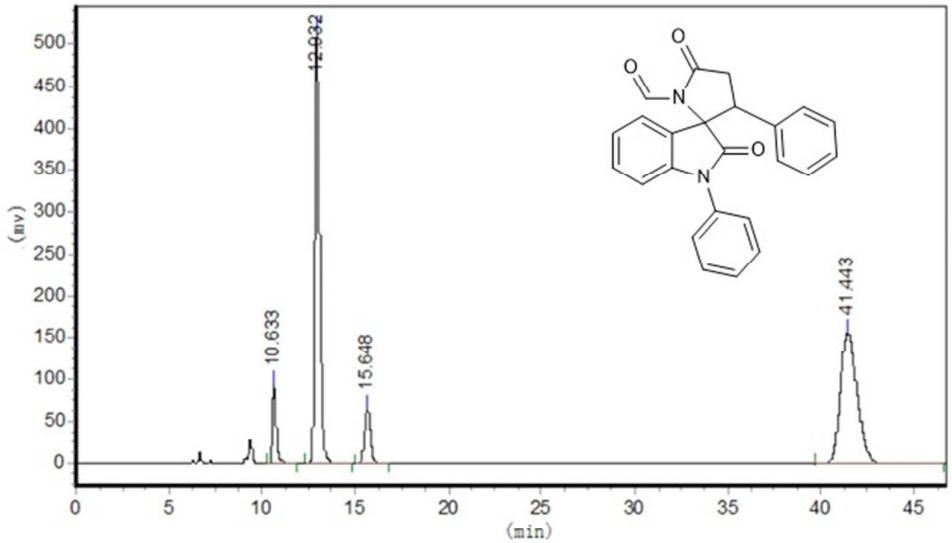


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	16.640	27969.863	633172.625	4.6510
2	17.367	230960.047	6155042.000	45.2119
3	23.007	177671.672	6148425.000	45.1633
4	24.363	18184.555	677132.875	4.9739
Total		454786.137	13613772.500	100.0000

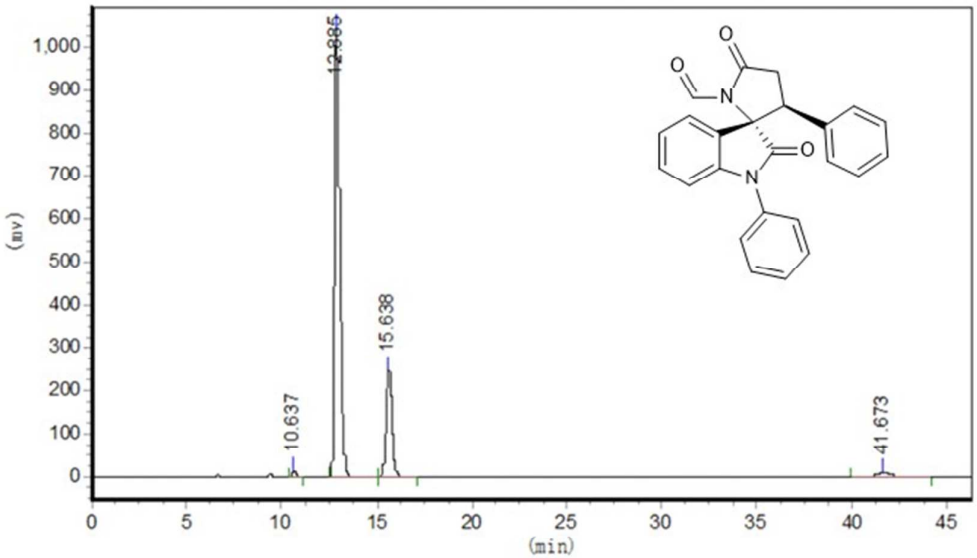


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	16.487	140557.781	3458876.500	25.5570
2	17.575	10113.960	266967.656	1.9726
3	22.940	277121.938	9654126.000	71.3325
4	24.348	4032.926	154009.984	1.1380
Total		431826.604	13533980.141	100.0000

Figure S32: HPLC analysis of the derivative of the compound **4i**

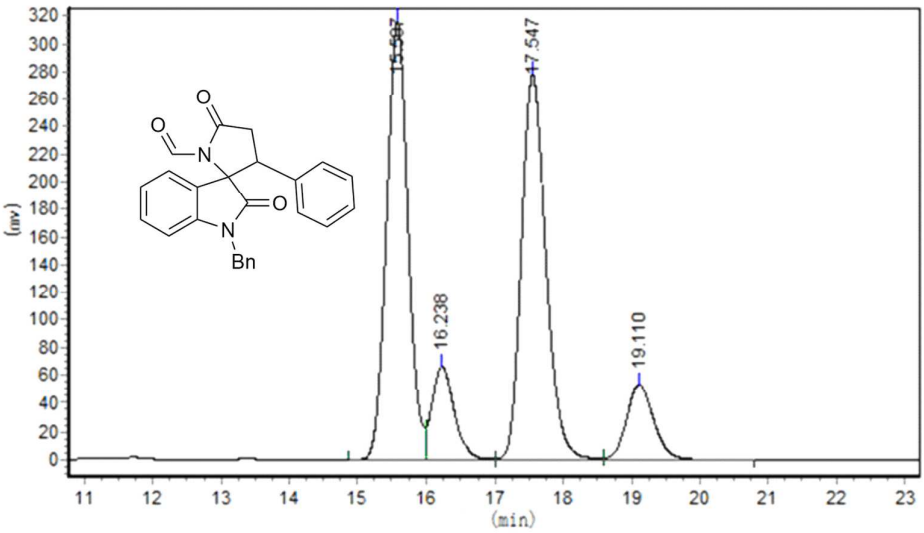


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	10.633	95076.547	1445859.875	6.4638
2	12.932	518476.875	9758604.000	43.6263
3	15.648	65151.438	1403712.375	6.2754
4	41.443	156458.219	9760472.000	43.6346
Total		835163.078	22368648.250	100.0000

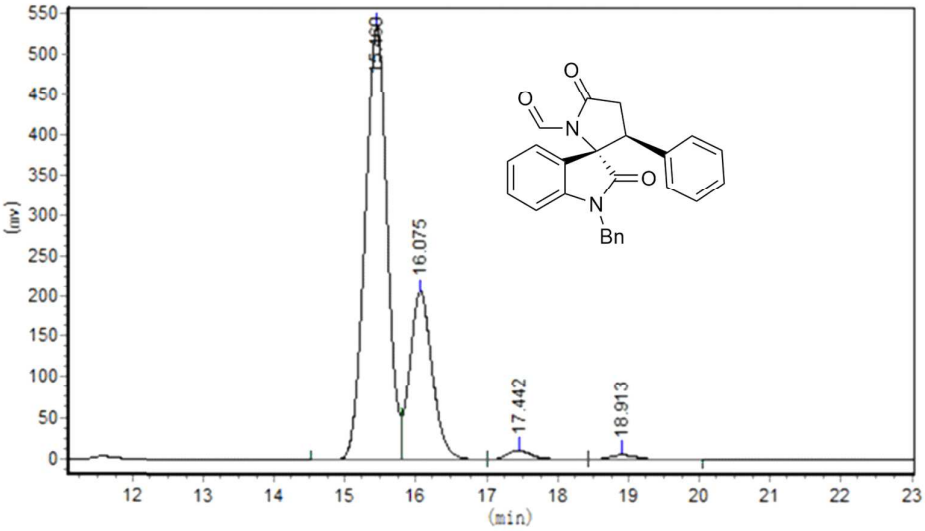


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	10.637	14458.800	206724.563	0.7618
2	12.885	1037717.750	20977684.000	77.3027
3	15.638	248866.891	5332804.500	19.6514
4	41.673	10159.583	619855.063	2.2842
Total		1311203.023	27137068.125	100.0000

Figure S33: HPLC analysis of the derivative of the compound **4j**

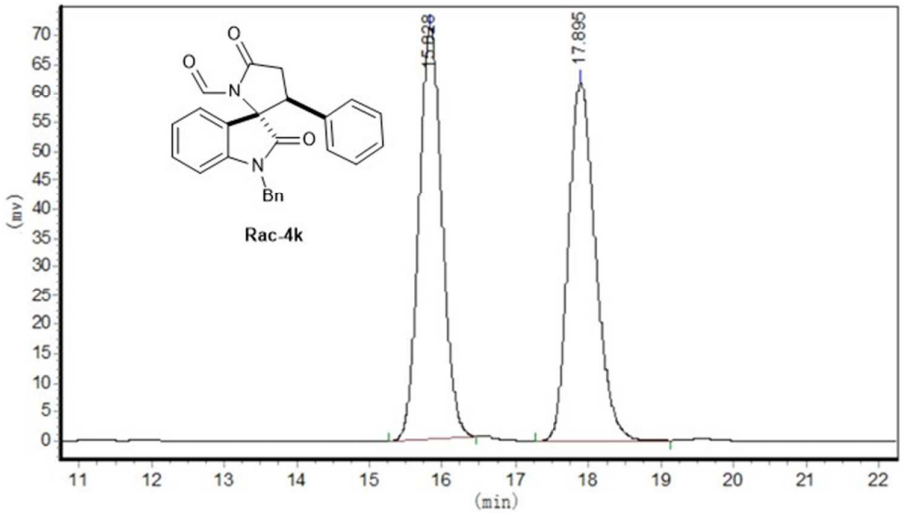


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	15.587	315136.938	6865188.500	41.1325
2	16.238	66181.930	1482413.875	8.8818
3	17.547	277758.938	6891294.500	41.2889
4	19.110	53016.566	1451531.375	8.6968
Total		712094.371	16690428.250	100.0000

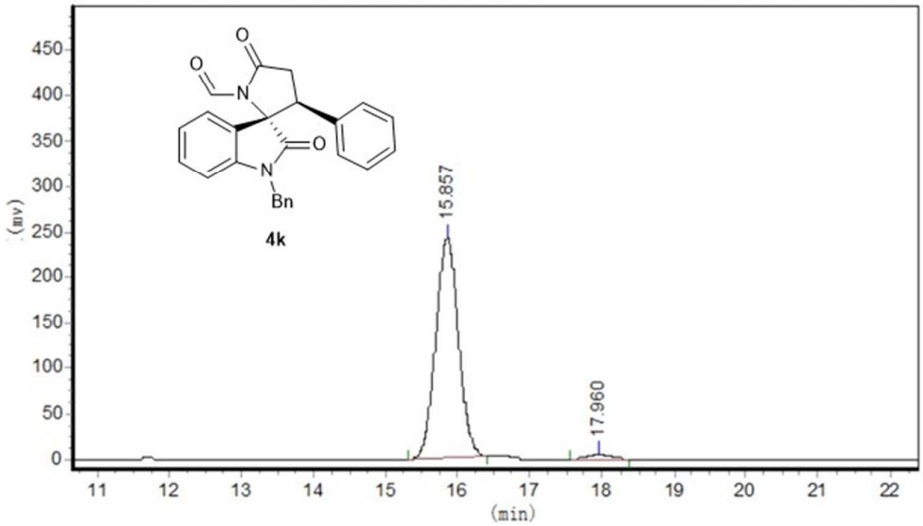


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	15.460	534459.500	11562757.000	69.9719
2	16.075	206195.844	4504408.000	27.2584
3	17.442	10902.589	295884.813	1.7905
4	18.913	5952.388	161806.625	0.9792
Total		757510.321	16524856.438	100.0000

Figure S34: HPLC analysis of the derivative of the compound **4k**

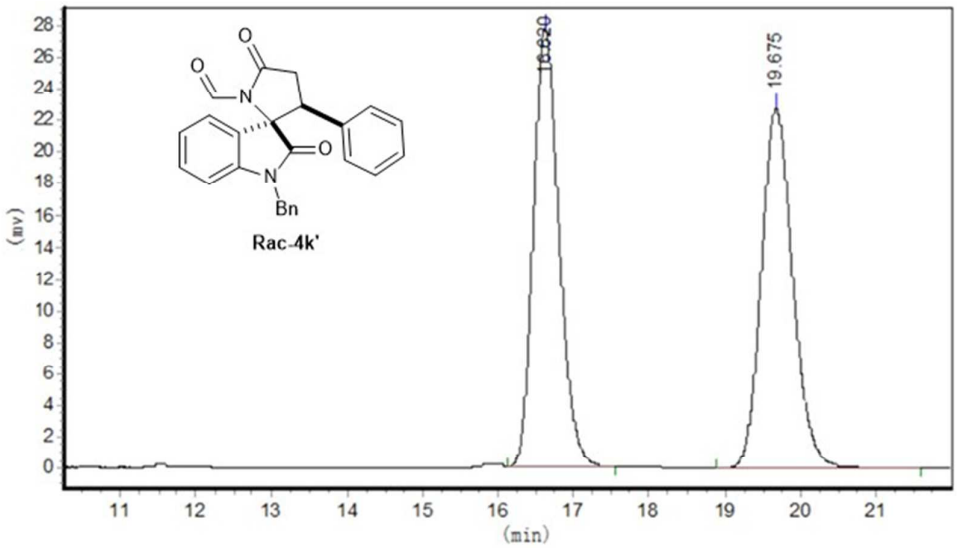


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	15.828	71097.805	1567694.750	49.5476
2	17.895	61761.566	1596319.750	50.4524
Total		132859.371	3164014.500	100.0000

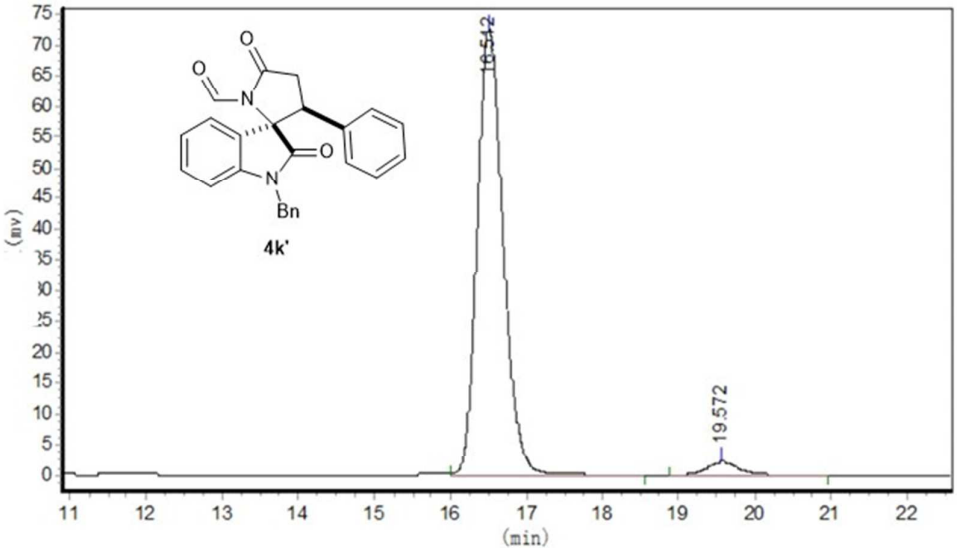


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	15.857	242460.125	5306438.000	97.9944
2	17.960	4725.675	108604.594	2.0056
Total		247185.800	5415042.594	100.0000

Figure S34': HPLC analysis of the major product **4k**

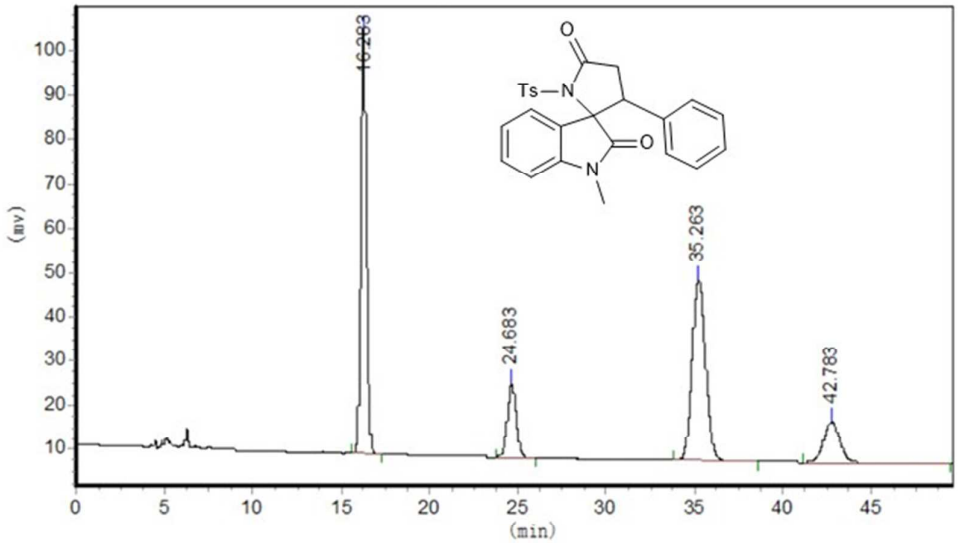


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	16.620	27709.166	651178.063	49.7473
2	19.675	22754.287	657794.000	50.2527
Total		50463.453	1308972.063	100.0000

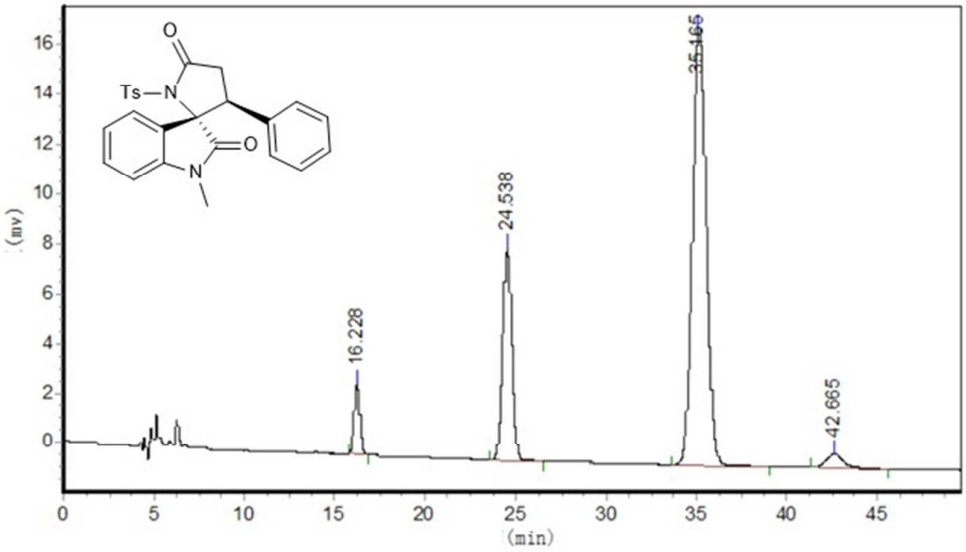


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	16.512	72589.297	1708768.125	96.5913
2	19.572	2096.028	60302.750	3.4087
Total		74685.325	1769070.875	100.0000

Figure S34'': HPLC analysis of the minor product **4k'**

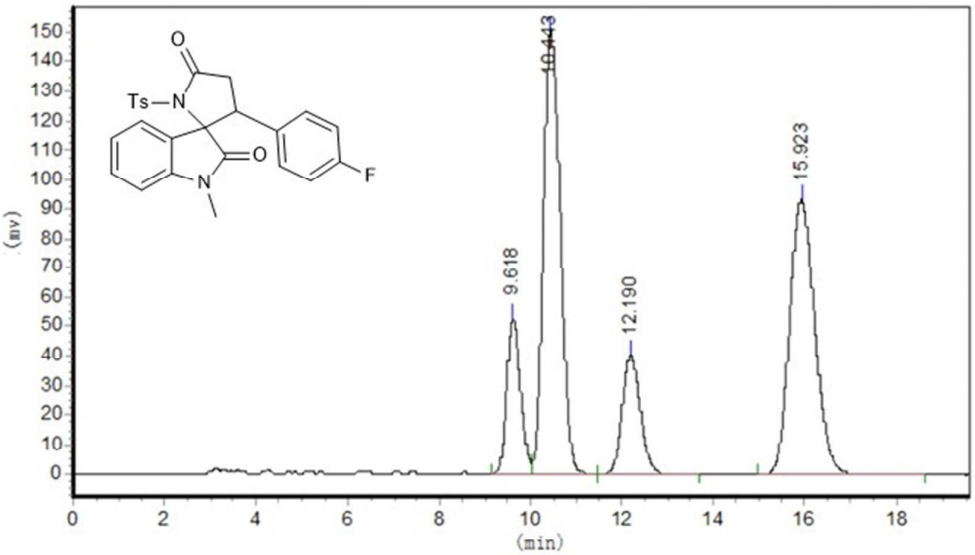


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	16.283	95960.258	2156805.750	39.2085
2	24.683	16692.230	586916.000	10.6695
3	35.263	40711.801	2167222.750	39.3978
4	42.783	9191.500	589923.188	10.7242
Total		162555.789	5500867.688	100.0000

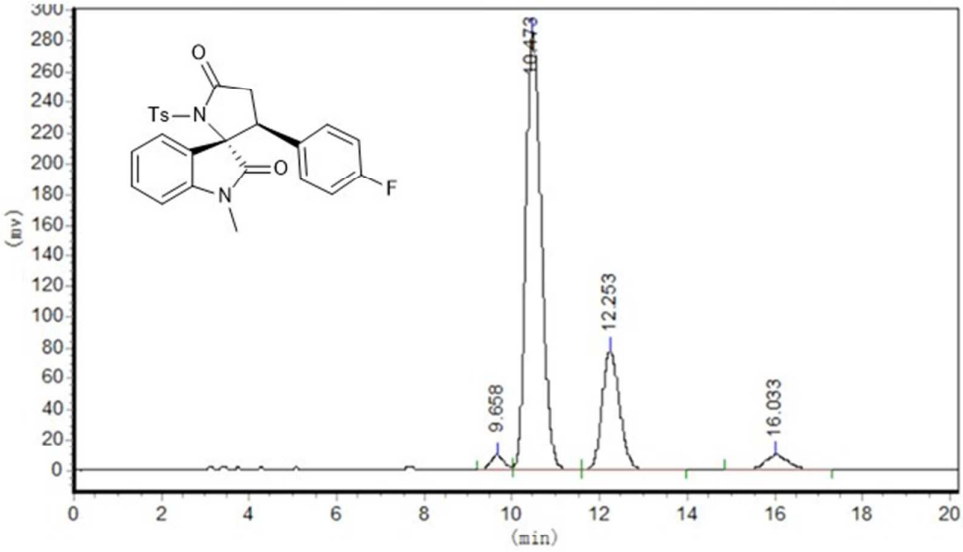


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	16.228	2805.919	61471.301	4.6696
2	24.538	8490.557	297341.750	22.5871
3	35.165	17493.730	923905.938	70.1831
4	42.665	526.897	33703.098	2.5602
Total		29317.103	1316422.086	100.0000

Figure S35: HPLC analysis of the derivative of the compound **6a**

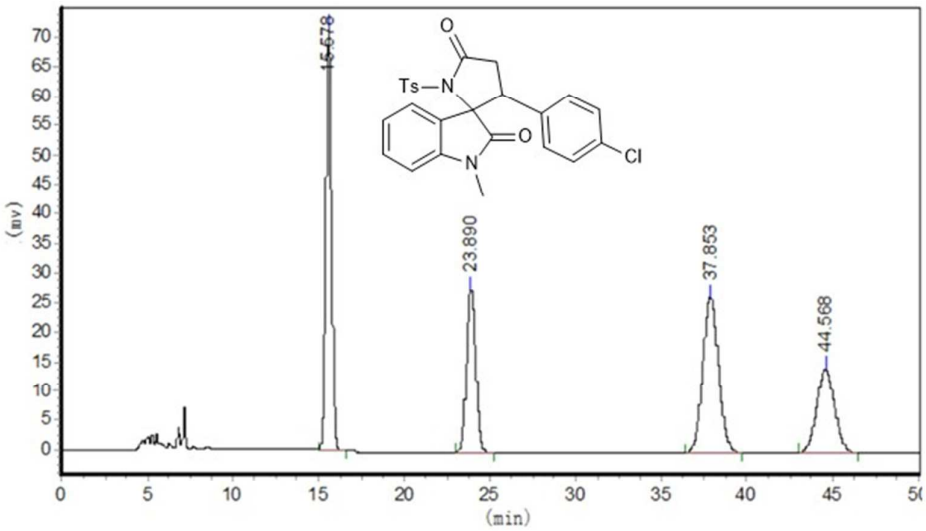


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	9.618	52371.730	1092901.500	11.3959
2	10.443	150555.828	3843354.250	40.0754
3	12.190	40320.730	1098408.625	11.4533
4	15.923	93197.750	3555640.500	37.0754
Total		336336.039	9590304.875	100.0000

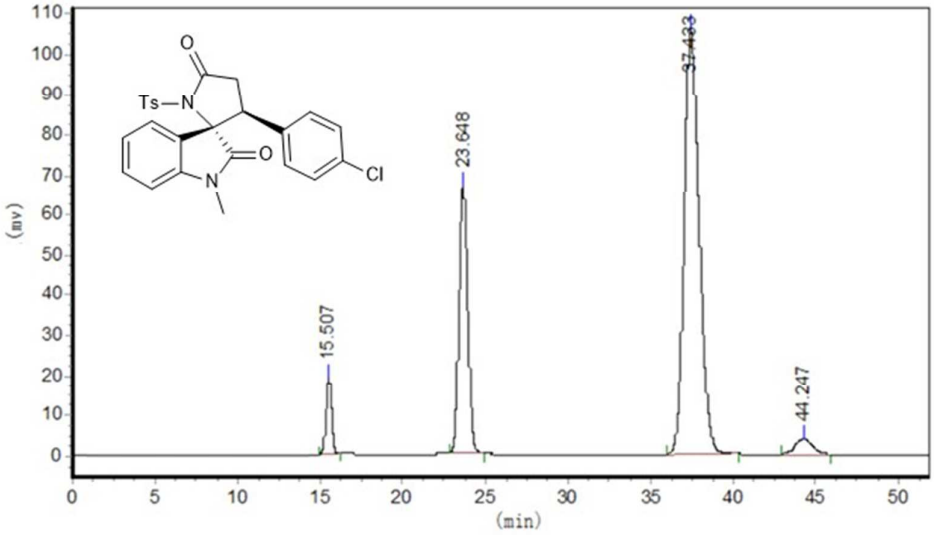


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	9.658	9429.484	193230.656	1.9816
2	10.473	285871.375	7055648.000	72.3575
3	12.253	77903.820	2149178.250	22.0404
4	16.033	9258.839	353042.906	3.6205
Total		382563.519	9751099.813	100.0000

Figure S36: HPLC analysis of the derivative of the compound **6b**

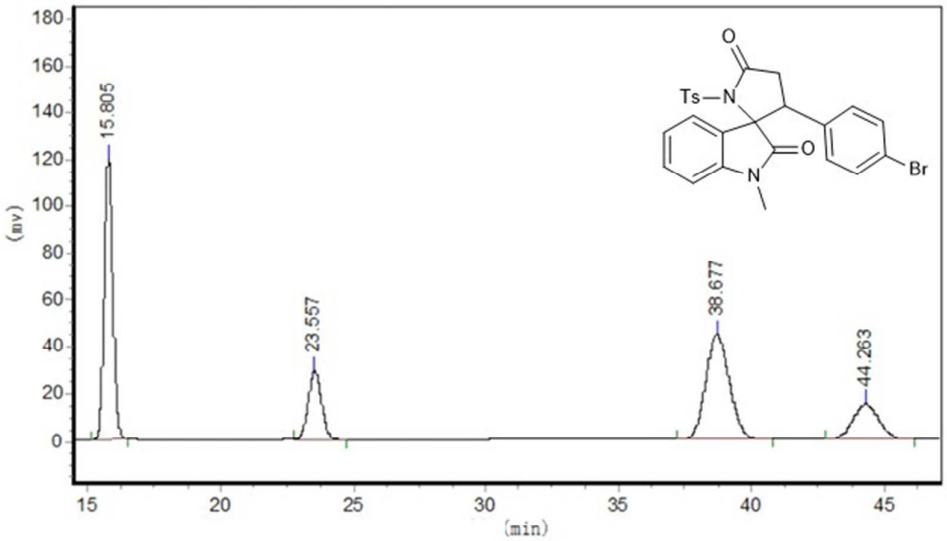


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	15.578	71204.047	1671757.625	31.0322
2	23.890	27518.275	1026594.625	19.0563
3	37.853	26234.779	1670909.000	31.0164
4	44.568	13984.000	1017911.875	18.8951
Total		138941.102	5387173.125	100.0000

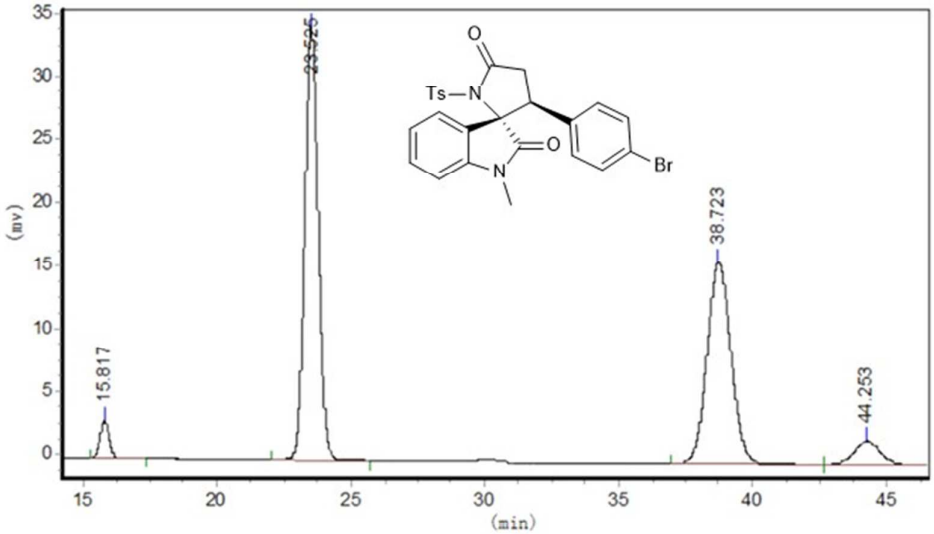


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	15.507	18480.129	416474.688	4.2227
2	23.648	66886.883	2482009.250	25.1655
3	37.433	105732.836	6757789.000	68.5182
4	44.247	3238.398	206484.797	2.0936
Total		194338.246	9862757.734	100.0000

Figure S37: HPLC analysis of the derivative of the compound **6c**

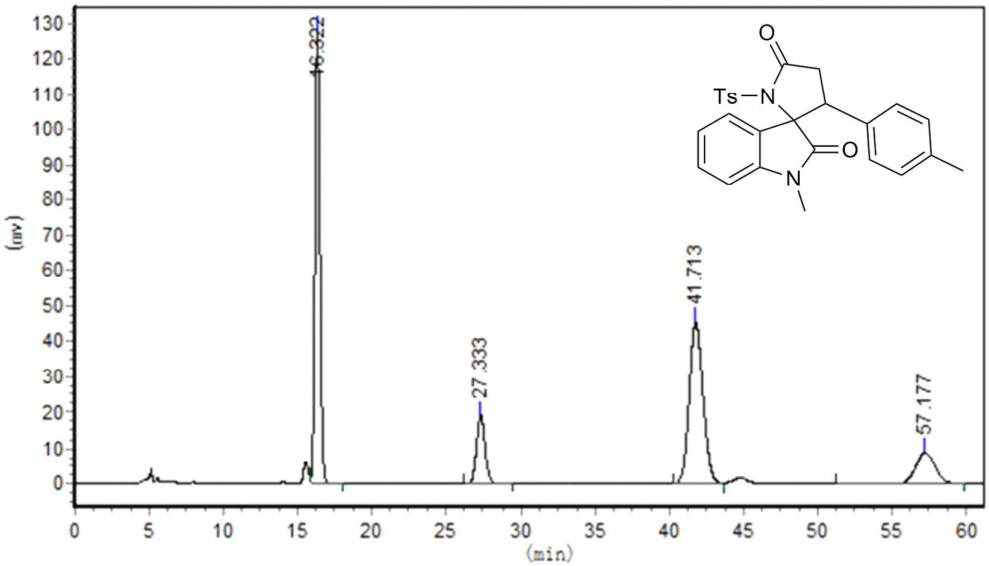


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	15.805	119893.211	2695767.750	36.1604
2	23.557	29099.709	1011407.188	13.5668
3	38.677	44520.699	2738495.000	36.7335
4	44.263	14799.054	1009355.563	13.5393
Total		208312.673	7455025.500	100.0000

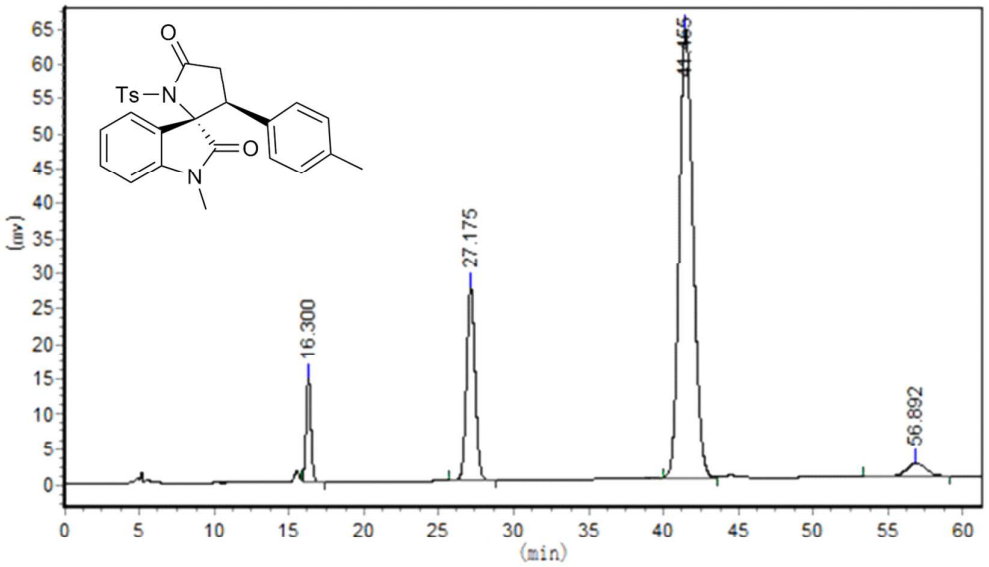


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	15.817	2987.620	67486.148	2.8427
2	23.525	34344.875	1193273.000	50.2645
3	38.723	16002.278	984862.000	41.4856
4	44.253	1816.945	128366.531	5.4072
Total		55151.718	2373987.680	100.0000

Figure S38: HPLC analysis of the derivative of the compound **6d**

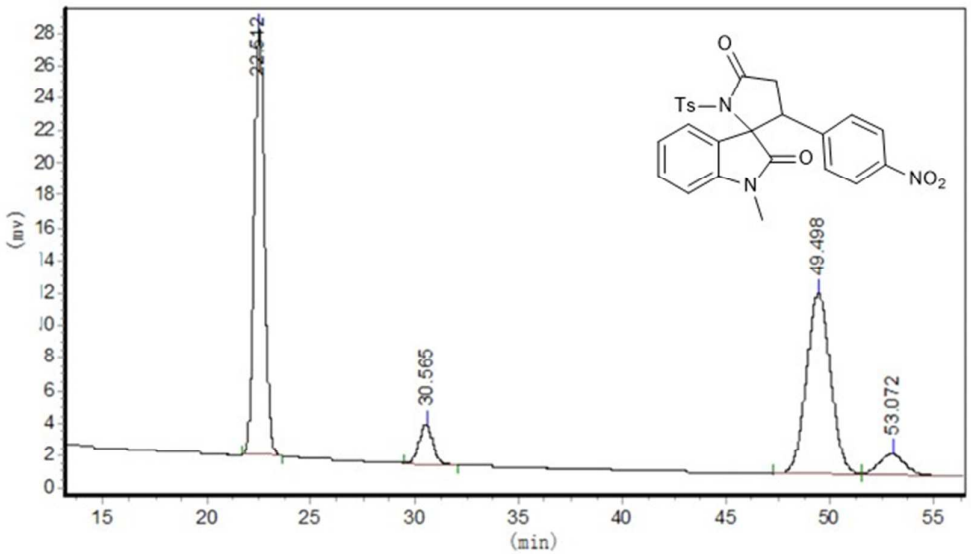


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	16.322	128081.797	2949755.500	39.7040
2	27.333	19027.605	764903.125	10.2957
3	41.713	45285.813	2953126.250	39.7493
4	57.177	8698.386	761589.125	10.2511
Total		201093.601	7429374.000	100.0000

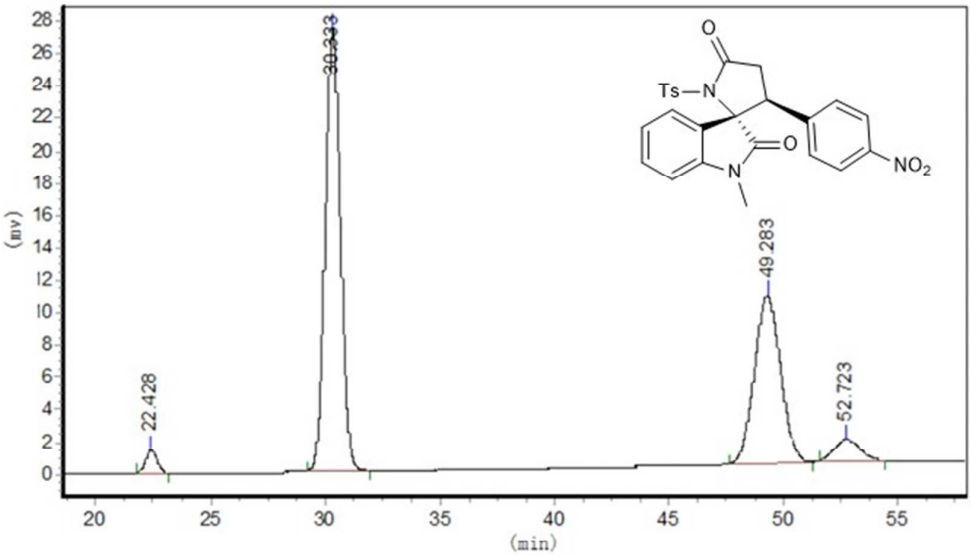


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	16.300	14867.250	339807.406	5.9326
2	27.175	27525.668	1094602.125	19.1102
3	41.455	63839.402	4137428.500	72.2338
4	56.892	1801.667	155990.500	2.7234
Total		108033.987	5727828.531	100.0000

Figure S39: HPLC analysis of the derivative of the compound **6e**

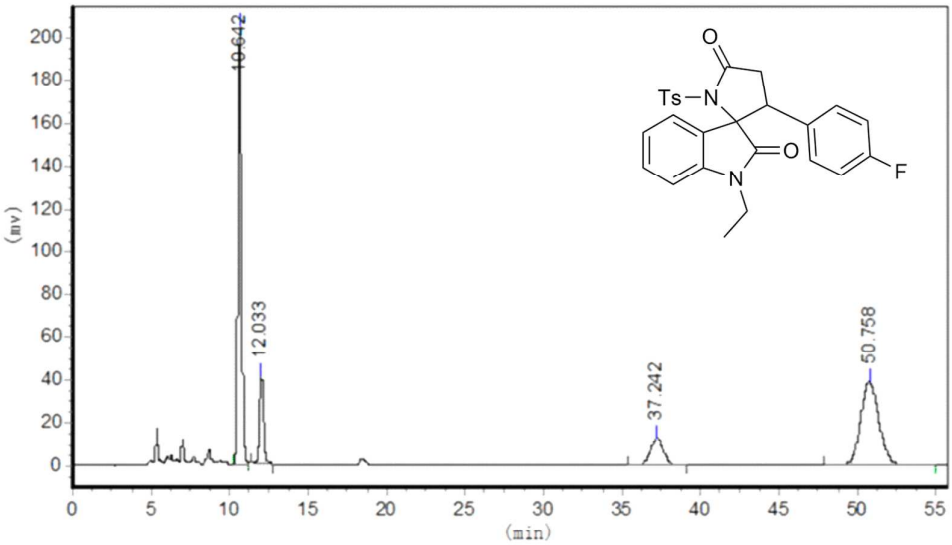


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	22.512	26248.293	897069.375	44.2807
2	30.565	2409.732	111987.703	5.5279
3	49.498	11117.335	904054.563	44.6255
4	53.072	1321.298	112757.609	5.5659
Total		41096.658	2025869.250	100.0000

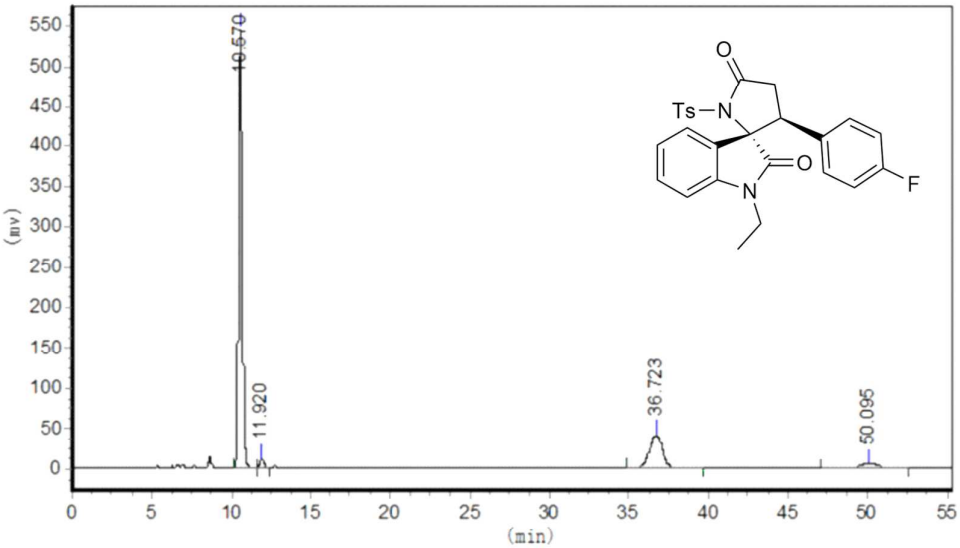


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	22.428	1459.460	48682.398	2.1811
2	30.333	27375.078	1260905.750	56.4921
3	49.283	10315.465	819468.438	36.7145
4	52.723	1310.766	102946.000	4.6123
Total		40460.769	2232002.586	100.0000

Figure S40: HPLC analysis of the derivative of the compound **6f**

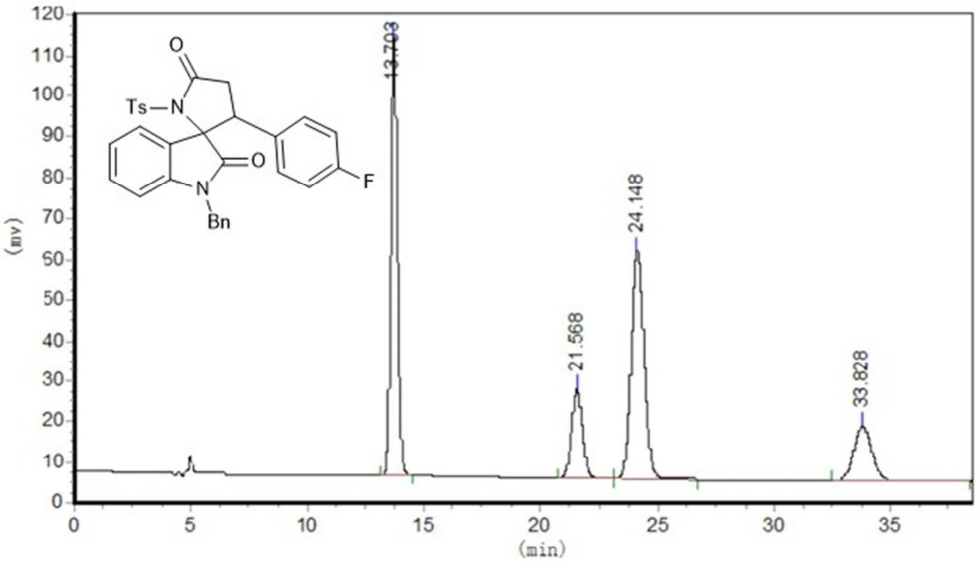


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	10.642	203863.719	3124426.000	40.8028
2	12.033	40331.215	706721.000	9.2293
3	37.242	12530.040	707049.625	9.2336
4	50.758	38741.438	3119178.000	40.7343
Total		295466.411	7657374.625	100.0000

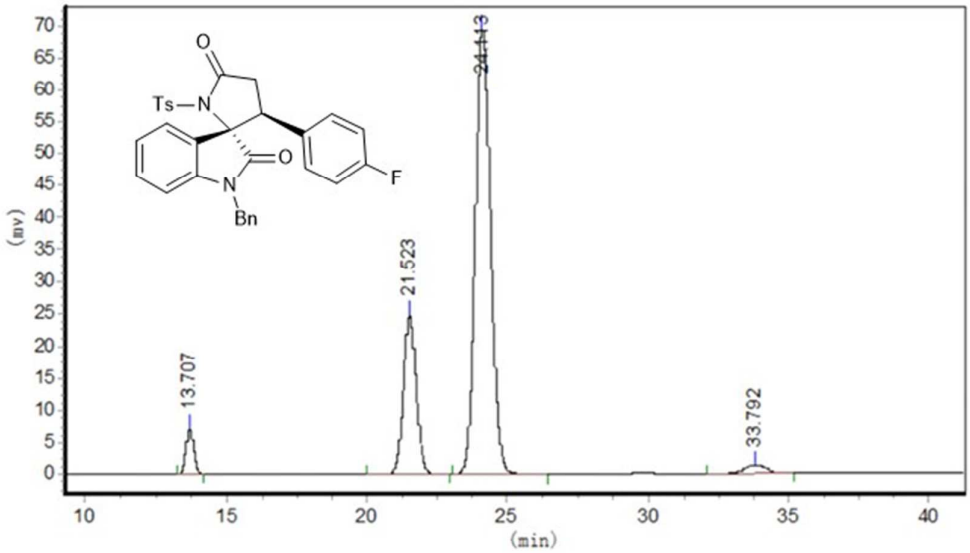


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	10.570	545997.750	8379773.000	73.6527
2	11.920	11160.707	195920.531	1.7220
3	36.723	41800.160	2346442.000	20.6237
4	50.095	5816.649	455281.125	4.0016
Total		604775.266	11377416.656	100.0000

Figure S41: HPLC analysis of the derivative of the compound **6g**

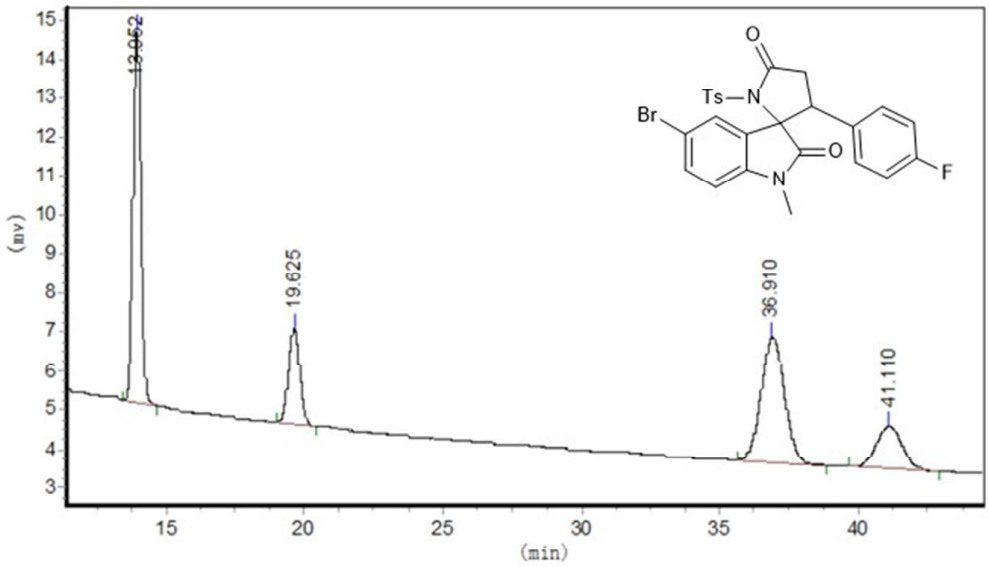


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	13.703	107880.523	2116852.500	37.3566
2	21.568	21976.832	708034.375	12.4948
3	24.148	55938.199	2135814.500	37.6912
4	33.828	13248.703	705914.813	12.4574
Total		199044.258	5666616.188	100.0000

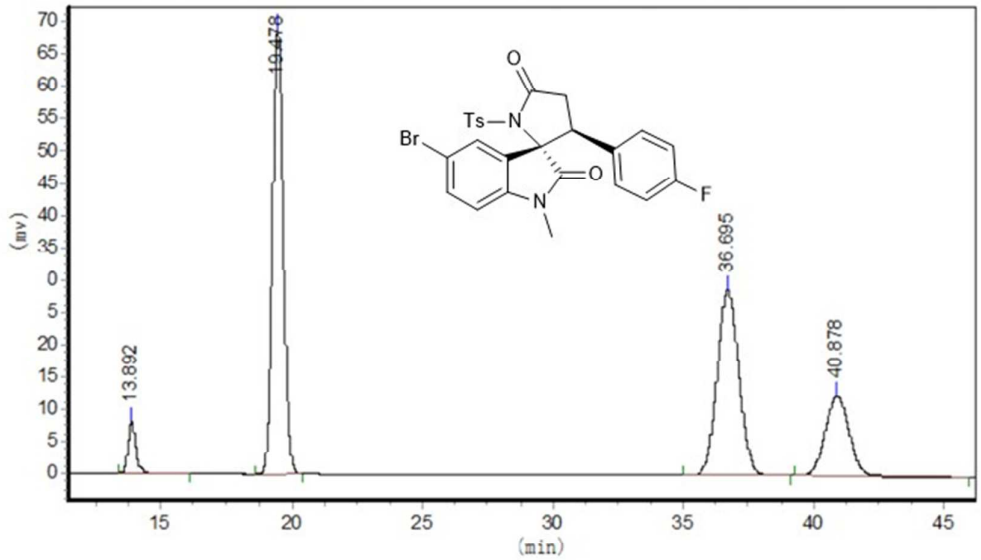


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	13.707	7139.207	138498.688	3.7957
2	21.523	24841.398	798716.188	21.8899
3	24.113	69597.781	2639642.000	72.3430
4	33.792	1366.960	71928.250	1.9713
Total		102945.347	3648785.125	100.0000

Figure S42: HPLC analysis of the derivative of the compound **6h**

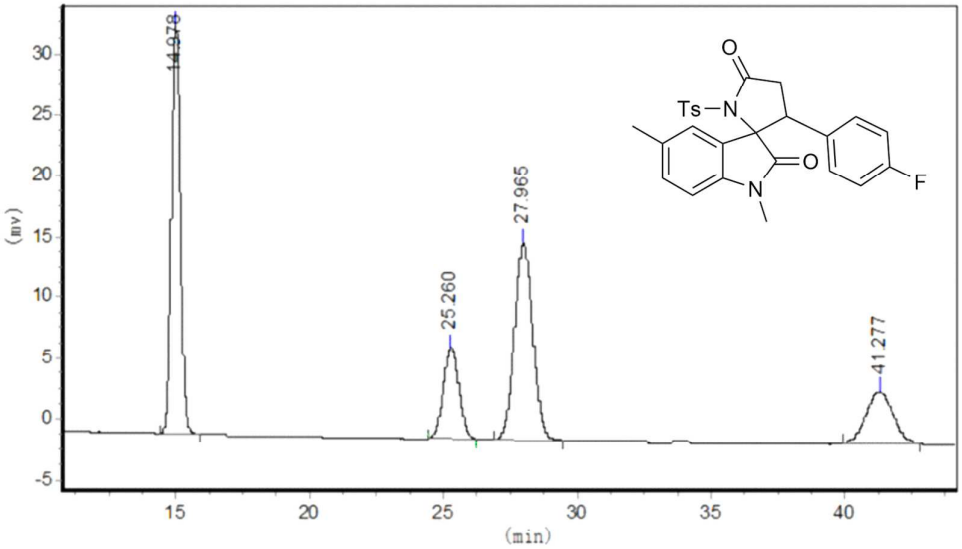


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	13.952	9571.640	187772.594	36.3344
2	19.625	2478.906	70644.602	13.6699
3	36.910	3204.860	187667.844	36.3141
4	41.110	1101.015	70705.109	13.6816
Total		16356.421	516790.148	100.0000

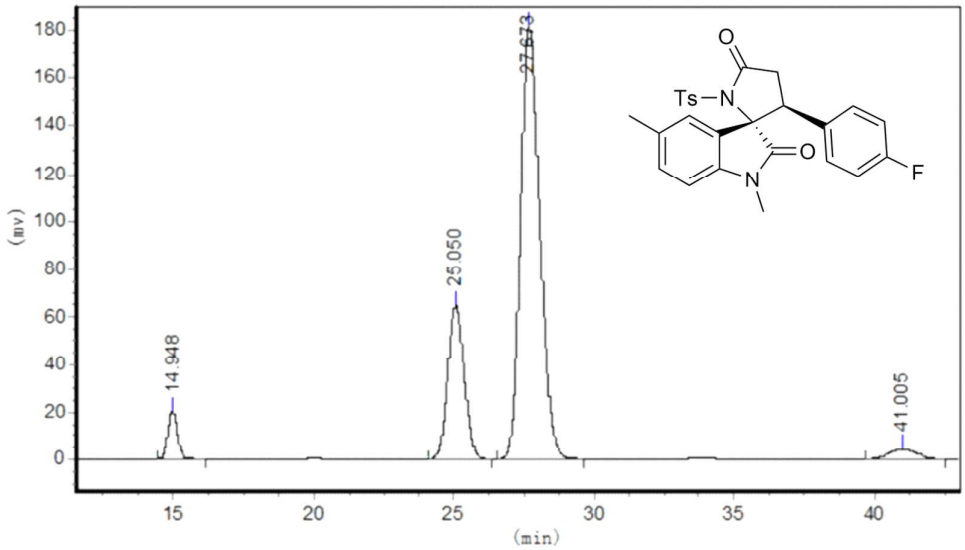


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	13.892	8002.604	168389.656	4.2489
2	19.478	69174.969	1968393.250	49.6674
3	36.695	28809.291	1679094.625	42.3677
4	40.878	4164.682	147271.109	3.7160
Total		110151.545	3963148.641	100.0000

Figure S43: HPLC analysis of the derivative of the compound **6i**

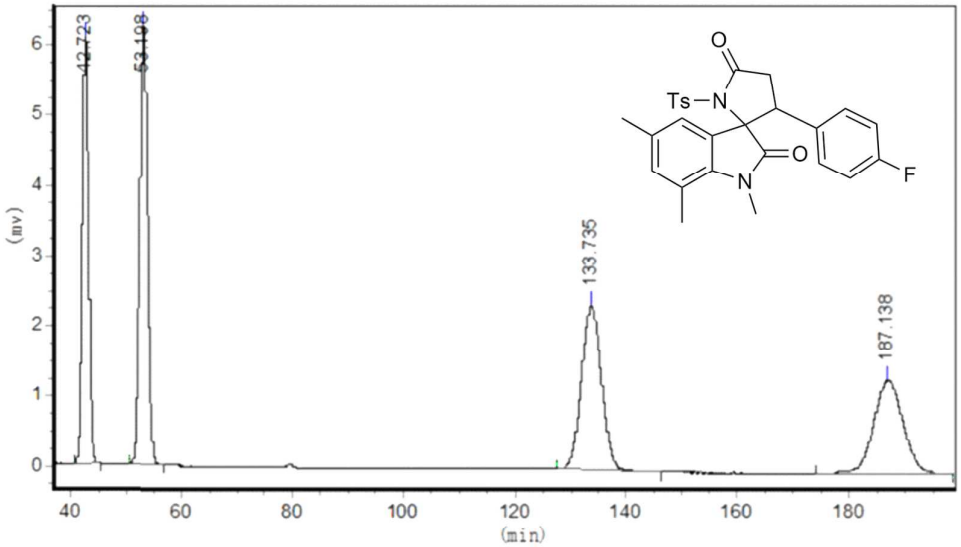


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	14.978	33555.133	781900.500	36.1238
2	25.260	7483.700	305846.000	14.1310
3	27.965	16281.564	777441.813	35.9179
4	41.277	4307.528	299311.125	13.8282
Total		61627.925	2164499.438	100.0000

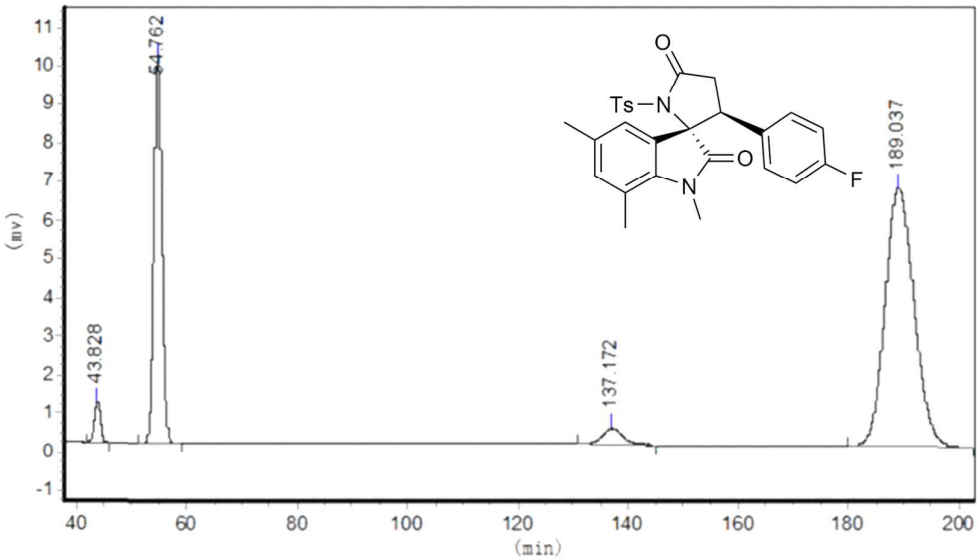


Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	14.948	20804.432	489496.094	4.0217
2	25.050	64700.176	26329999.000	21.6327
3	27.673	181545.188	8731026.000	71.7341
4	41.005	4642.184	317856.406	2.6115
Total		271691.979	12171377.500	100.0000

Figure S44: HPLC analysis of the derivative of the compound **6j**



Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	42.723	6084.131	471536.500	21.9751
2	53.198	6241.219	601442.250	28.0291
3	133.735	2336.161	589283.063	27.4625
4	187.138	1318.529	483513.344	22.5333
Total		15980.041	2145775.157	100.0000



Peak	RT (min)	Height (mV*sec)	Area (mV)	Area (%)
1	43.828	1050.881	86172.305	2.3509
2	54.762	10020.166	1015470.875	27.7030
3	137.172	426.065	109703.688	2.9928
4	189.037	6711.934	2454215.250	66.9533
Total		18209.046	3665562.117	100.0000

Figure S45: HPLC analysis of the derivative of the compound **6k**