

Enantio- and diastereoselective synthesis of spiropyrazolones via organocatalytic [1+2+3] multicomponent reaction

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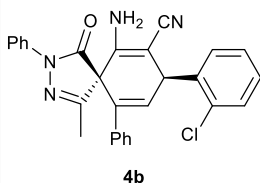
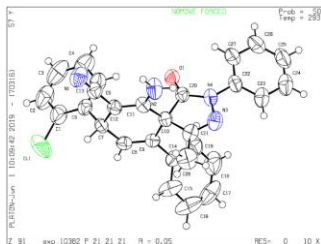
^bHospital of Chengdu University of Traditional Chinese Medicine, Chengdu, 610072, China. E-mail: zhenzhen41@cdutcm.edu.cn

[‡] These authors contributed equally to this work

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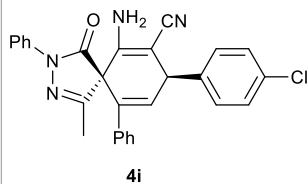
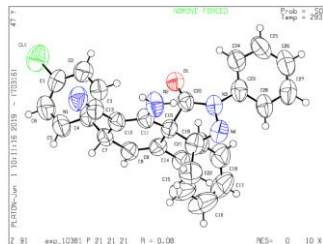
1. X-Ray Diffraction Data

1.1 X-Ray Diffraction Data for Compound 4b



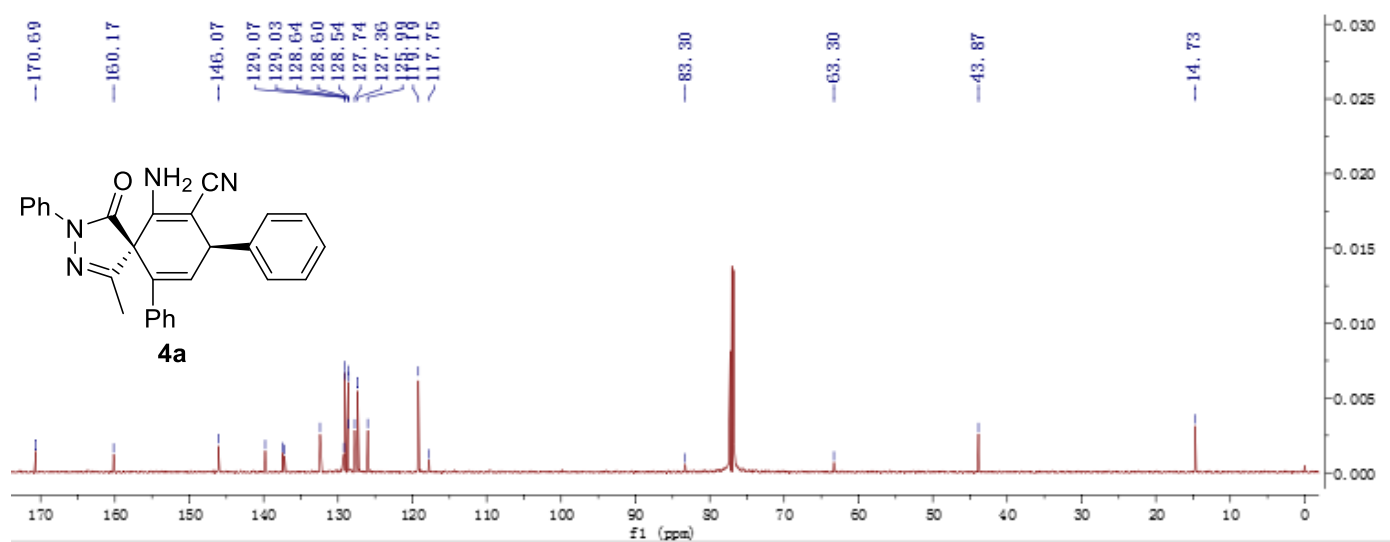
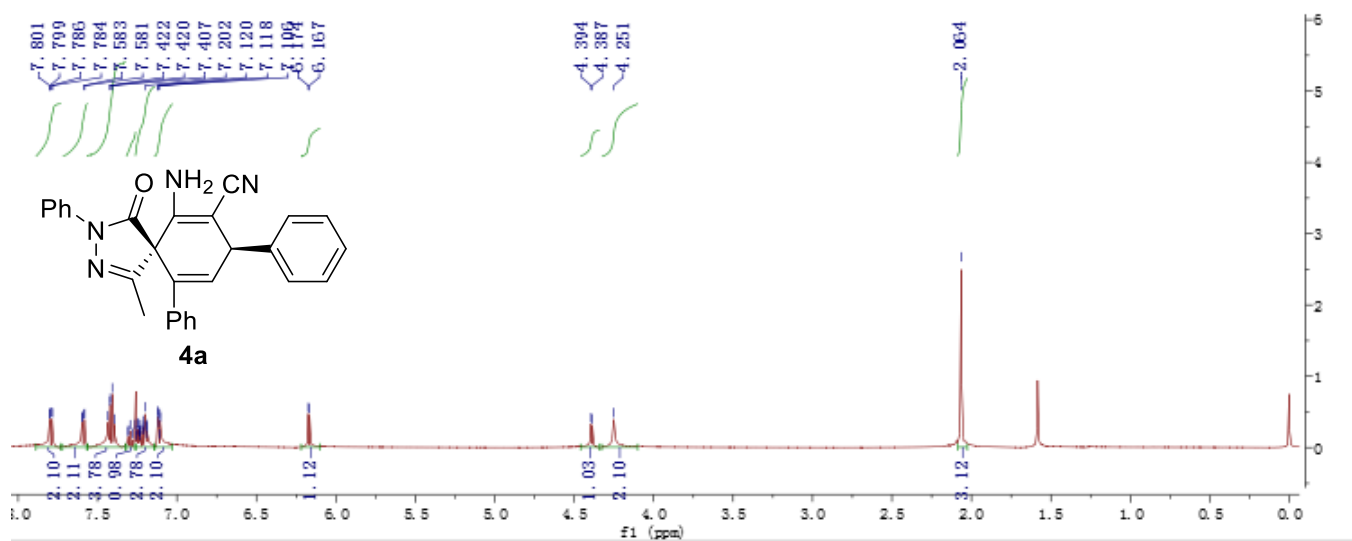
Identification code	exp_10382
Empirical formula	C ₂₈ H ₂₁ ClN ₄ O
Formula weight	464.94
Temperature	293(2) K
Wavelength	1.54184 Å
Unit cell dimensions	a = 7.5705(5) Å alpha = 90 deg. b = 11.9923(7) Å beta = 90 deg. c = 27.772(2) Å gamma = 90 deg.
Volume	2521.3(3) Å ³
Z, Calculated density	4, 1.225 Mg/m ³
Absorption coefficient	1.549 mm ⁻¹
F(000)	968
Crystal size	0.13 x 0.12 x 0.11 mm
Theta range for data collection	3.18 to 67.24 deg.
Limiting indices	-9 ≤ h ≤ 8, -11 ≤ k ≤ 14, -23 ≤ l ≤ 33
Reflections collected / unique	5868 / 3882 [R(int) = 0.0302]
Completeness to theta = 67.24	99.8 %
Max. and min. transmission	0.8480 and 0.8240
Refinement method	Full-matrix least-squares on F ²
Data / restraints / parameters	3882 / 0 / 309
Goodness-of-fit on F ²	1.039
Final R indices [I > 2sigma(I)]	R1 = 0.0481, wR2 = 0.1120
R indices (all data)	R1 = 0.0605, wR2 = 0.1235
Absolute structure parameter	-0.01(3)
Extinction coefficient	0.0063(3)
Largest diff. peak and hole	0.261 and -0.227 e. Å ⁻³

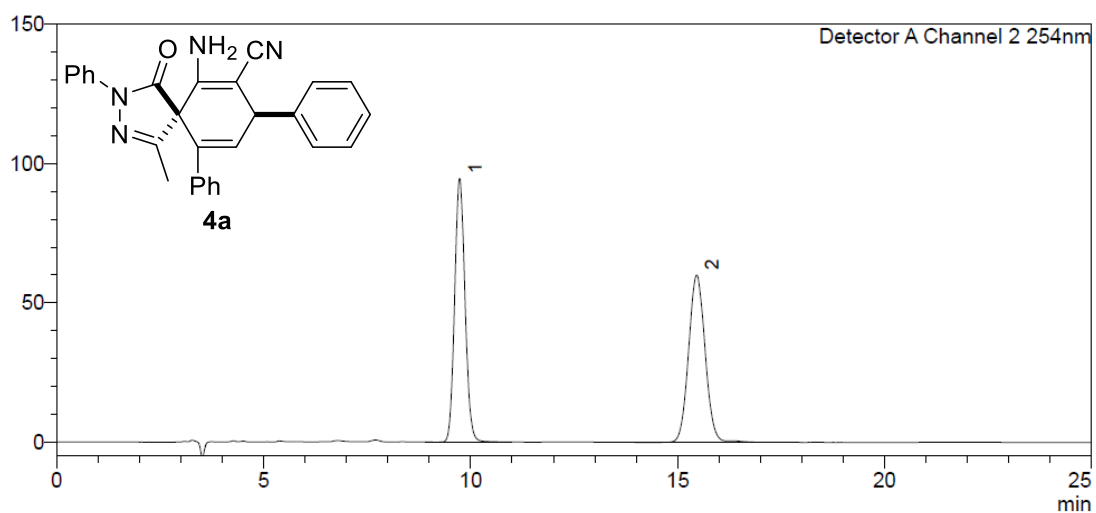
1.2 X-Ray Diffraction Data for Compound 4i



Identification code	exp_10381
Empirical formula	C ₂₈ H ₂₁ ClN ₄ O
Formula weight	464.94
Temperature	293(2) K
Wavelength	1.54184 Å
Unit cell dimensions	a = 7.6734(5) Å alpha = 90 deg. b = 11.9044(8) Å beta = 90 deg. c = 27.0141(17) Å gamma = 90 deg.
Volume	2467.7(3) Å ³
Z, Calculated density	4, 1.251 Mg/m ³
Absorption coefficient	1.583 mm ⁻¹
F(000)	968
Crystal size	0.09 x 0.08 x 0.08 mm
Theta range for data collection	3.27 to 67.23 deg.
Limiting indices	-9<=h<=8, -14<=k<=14, -20<=l<=32
Reflections collected / unique	16107 / 4413 [R(int) = 0.0699]
Completeness to theta = 67.23	100.0 %
Max. and min. transmission	0.8838 and 0.8706
Refinement method	Full-matrix least-squares on F ²
Data / restraints / parameters	4413 / 0 / 309
Goodness-of-fit on F ²	1.058
Final R indices [I>2sigma(I)]	R1 = 0.0785, wR2 = 0.1863
R indices (all data)	R1 = 0.1326, wR2 = 0.2507
Absolute structure parameter	-0.05(4)
Extinction coefficient	0.0133(11)
Largest diff. peak and hole	0.210 and -0.221 e. Å ⁻³

2. NMR and HPLC spectra of compound 4

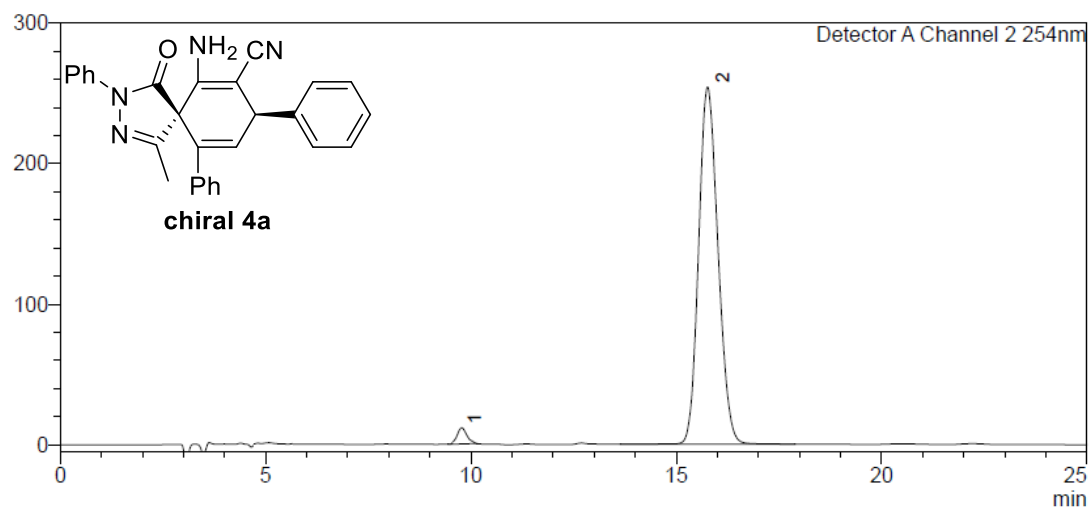




<Peak Table>

Detector A Channel 2 254nm

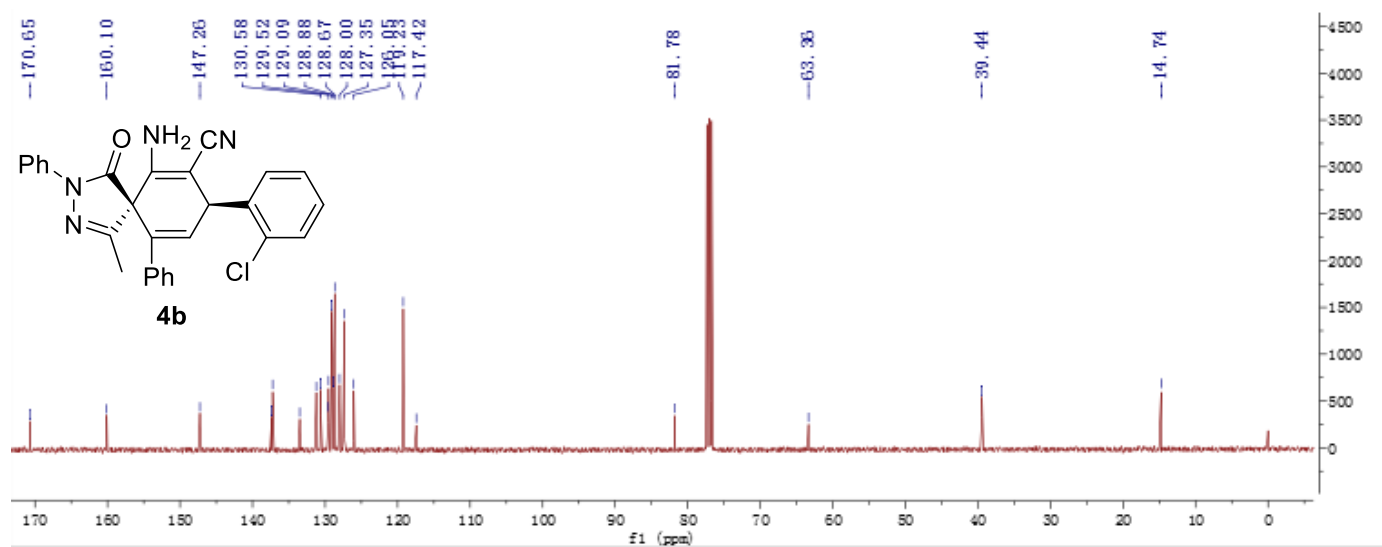
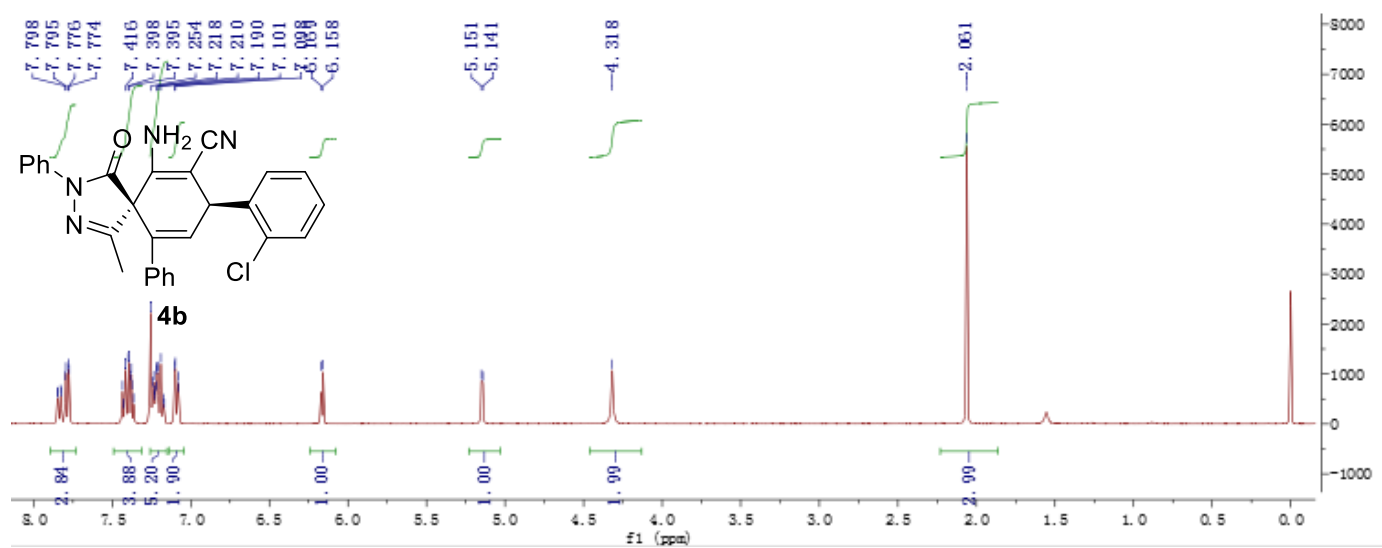
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	9.734	1653246	94758	49.669	49.669
2	15.462	1675249	60116	50.331	50.331
Total		3328495	154875		100.000

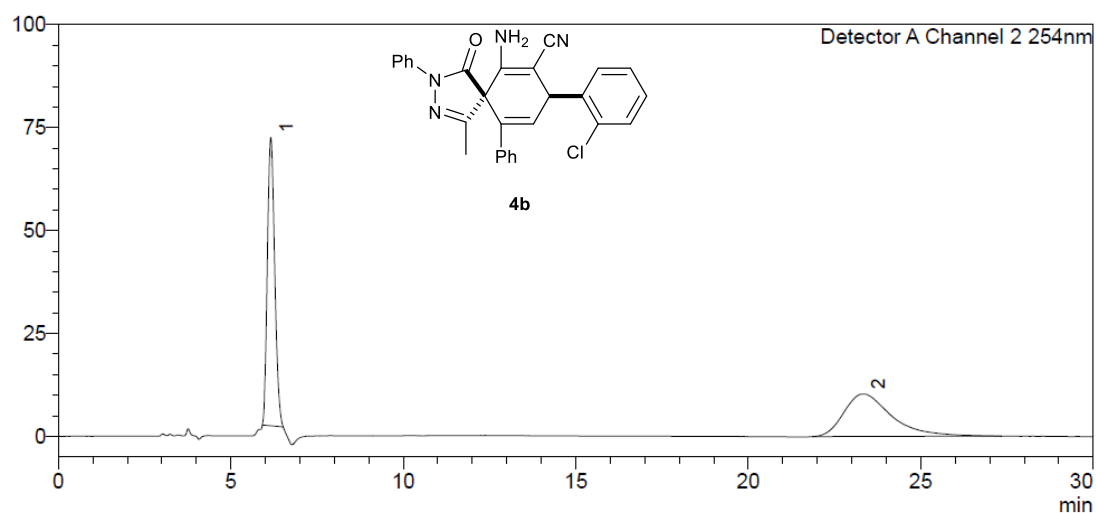


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	9.715	150748	7188	1.772	1.772
2	15.762	8355702	254226	98.228	98.228
Total		8506451	261414		100.000

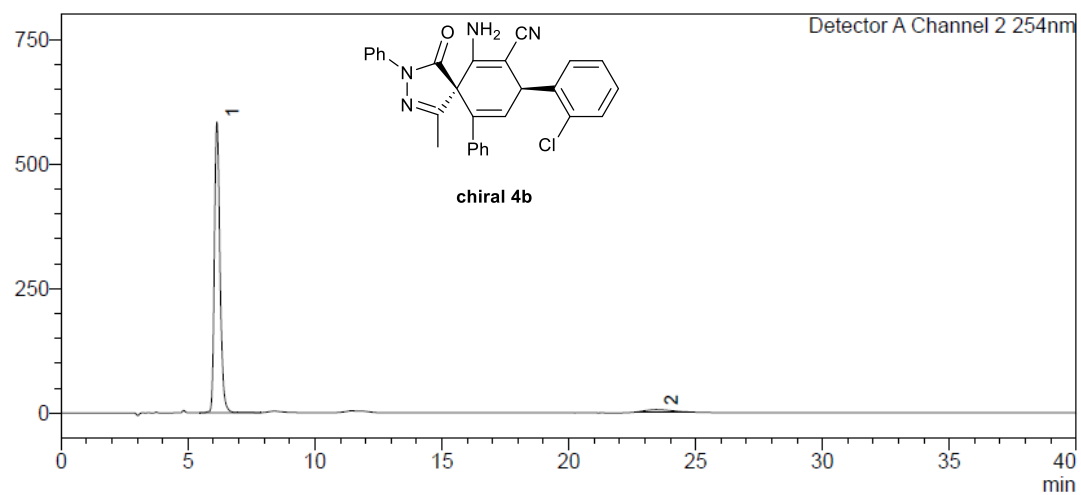




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Detector A Channel 2 254nm

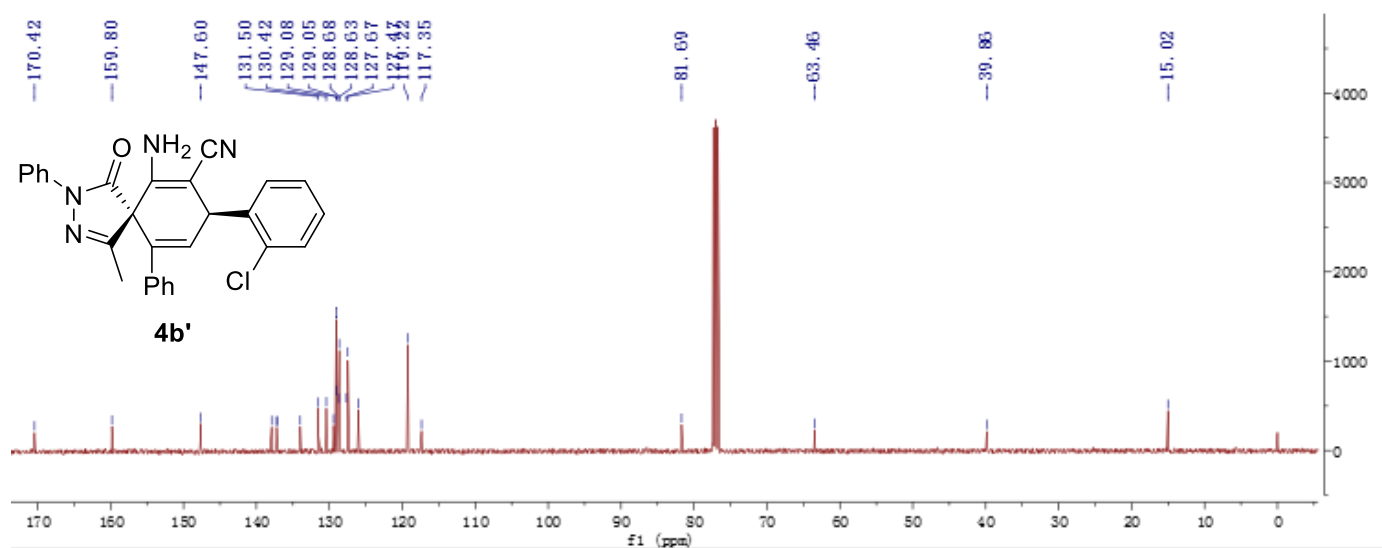
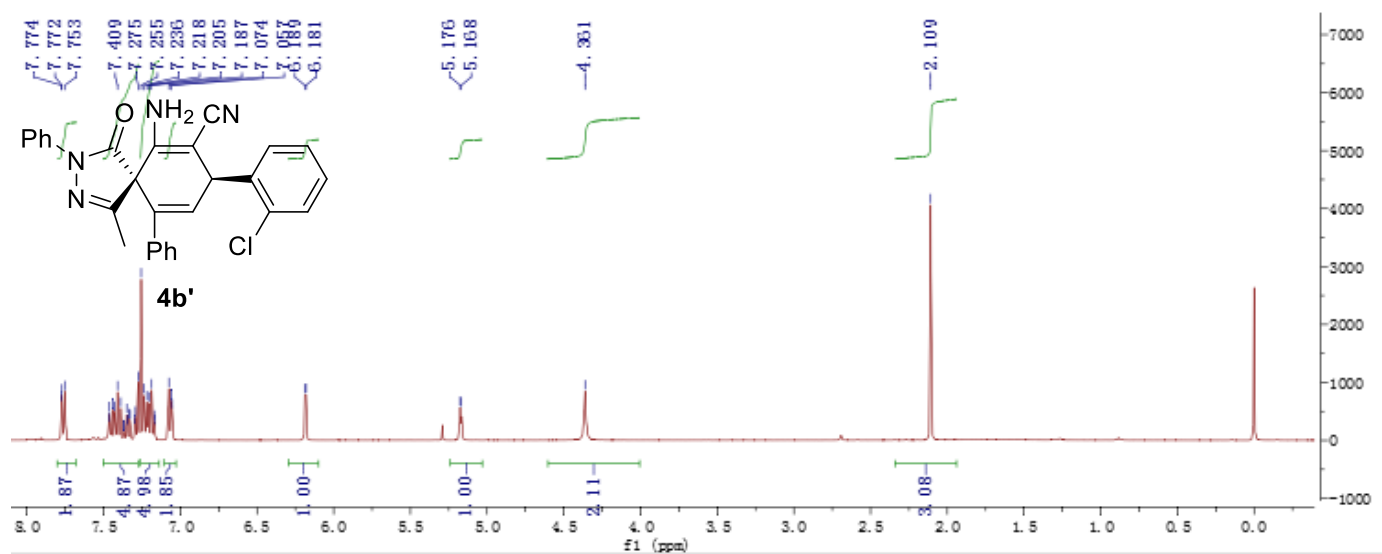
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	6.154	1031373	70053	50.959	50.959
2	23.323	992570	10329	49.041	49.041
Total		2023943	80382		100.000

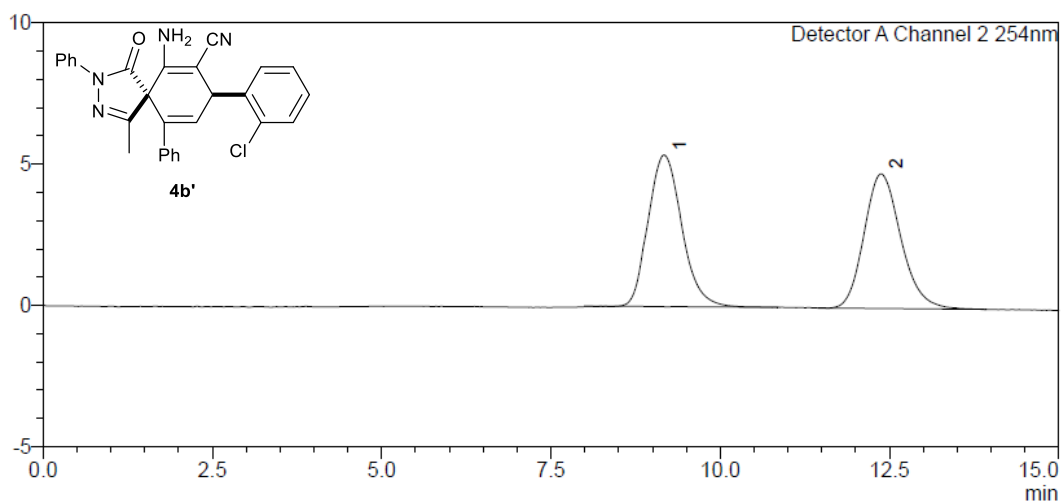


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	6.133	8938424	583772	96.438	96.438
2	23.444	330163	4747	3.562	3.562
Total		9268587	588519		100.000

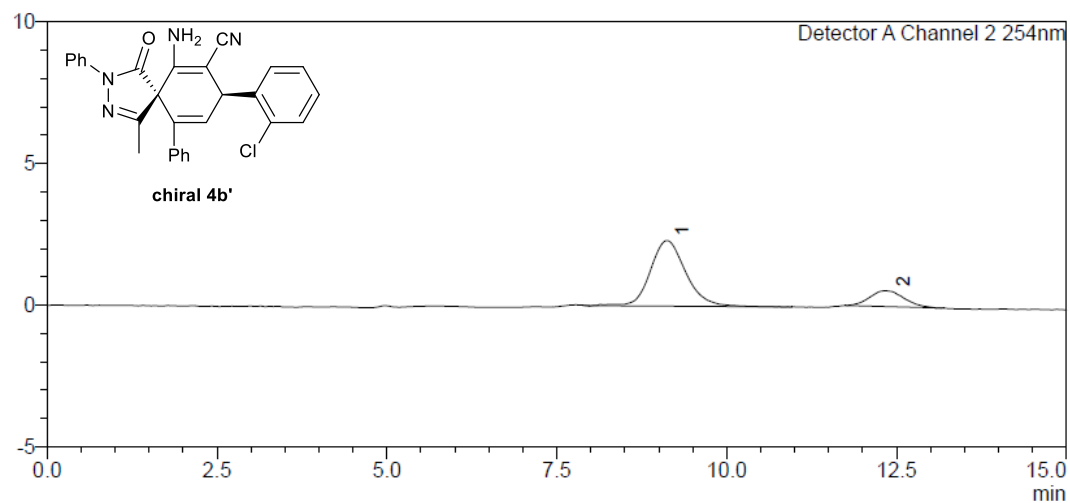




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Detector A Channel 2 254nm

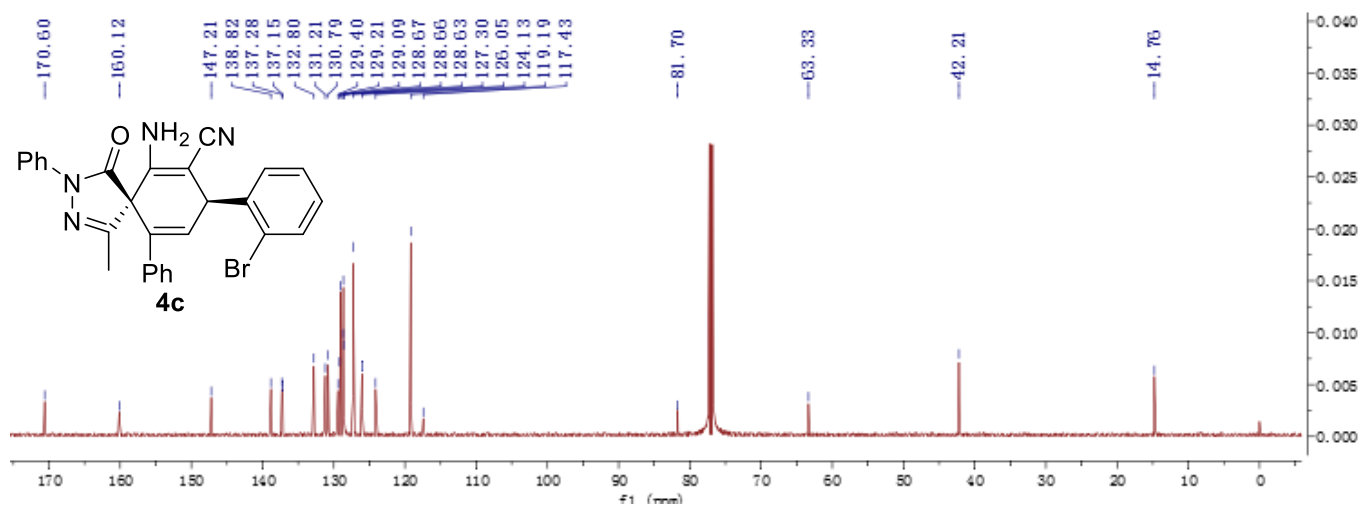
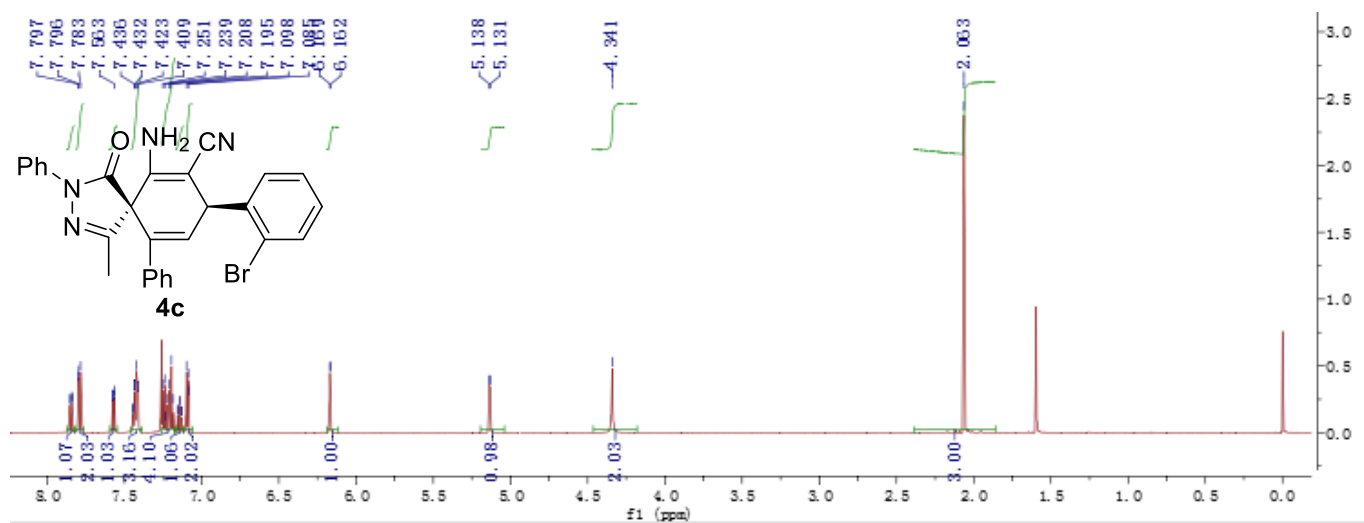
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	9.172	175011	5153	49.272	49.272
2	12.377	180185	4768	50.728	50.728
Total		355196	9920		100.000

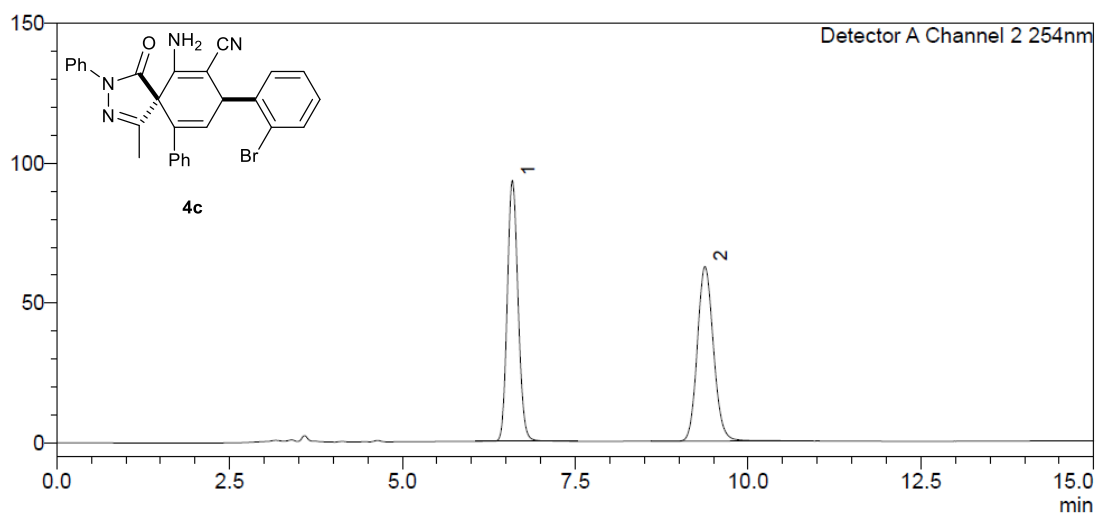


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	9.119	84579	2319	81.338	81.338
2	12.375	19406	552	18.662	18.662
Total		103985	2871		100.000

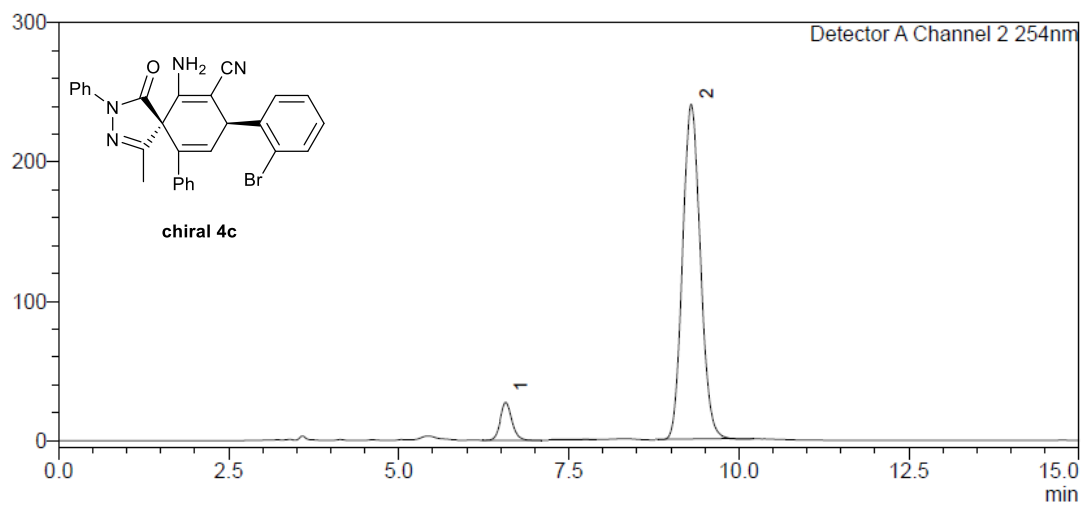




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Detector A Channel 2 254nm

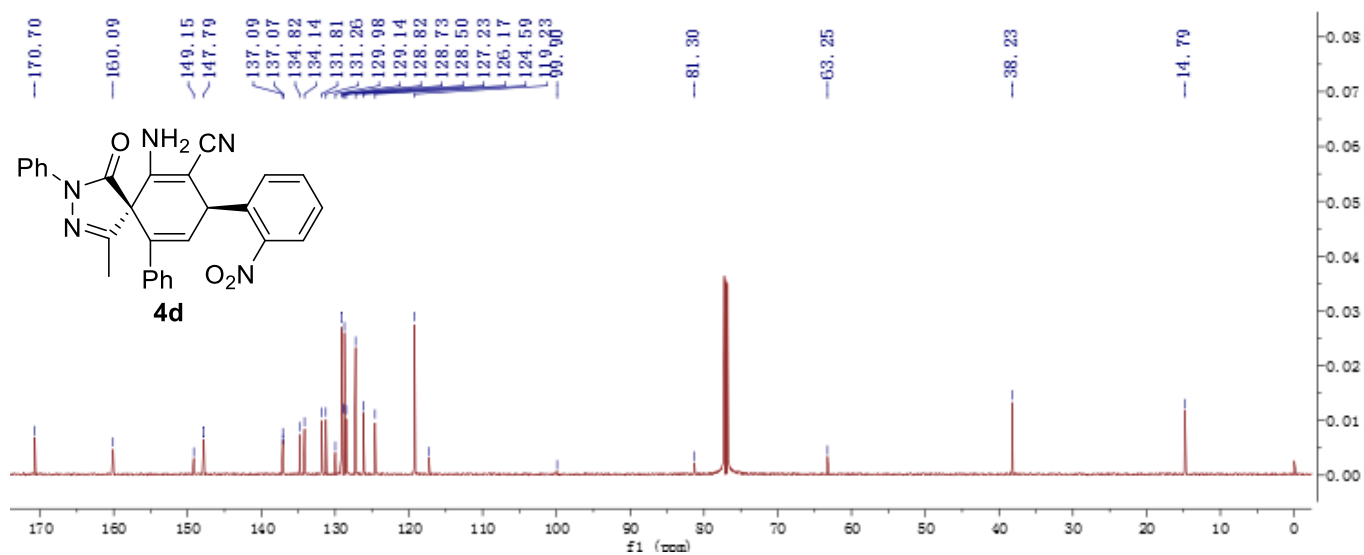
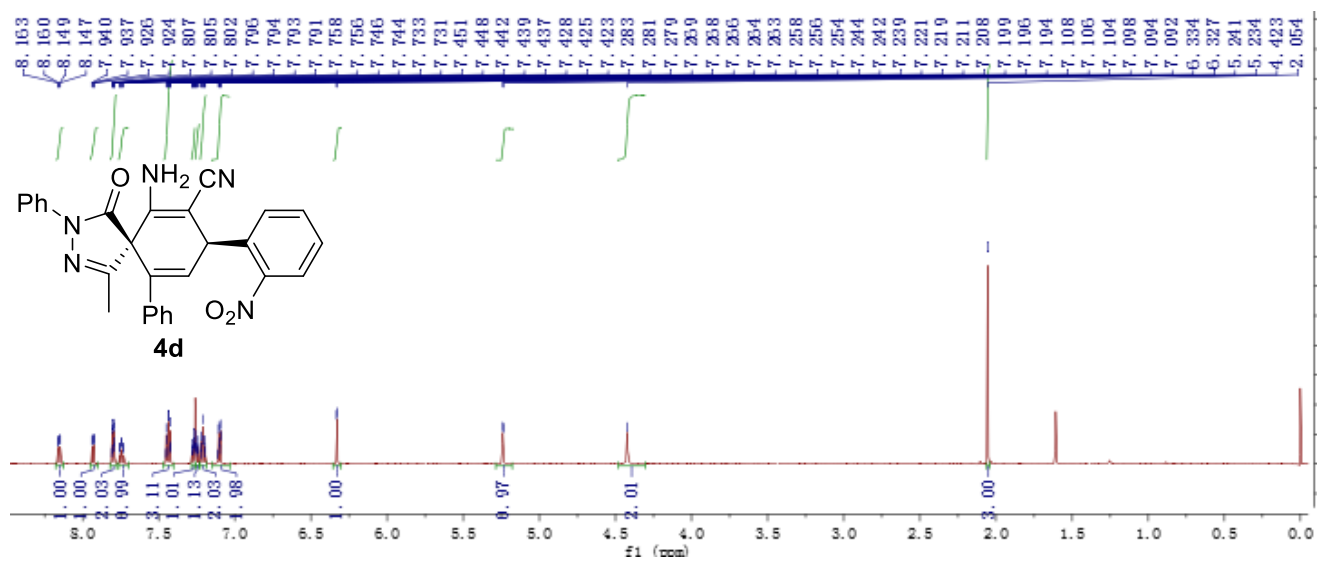
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	6.591	1009584	93348	49.817	49.817
2	9.377	1017016	62460	50.183	50.183
Total		2026600	155808		100.000

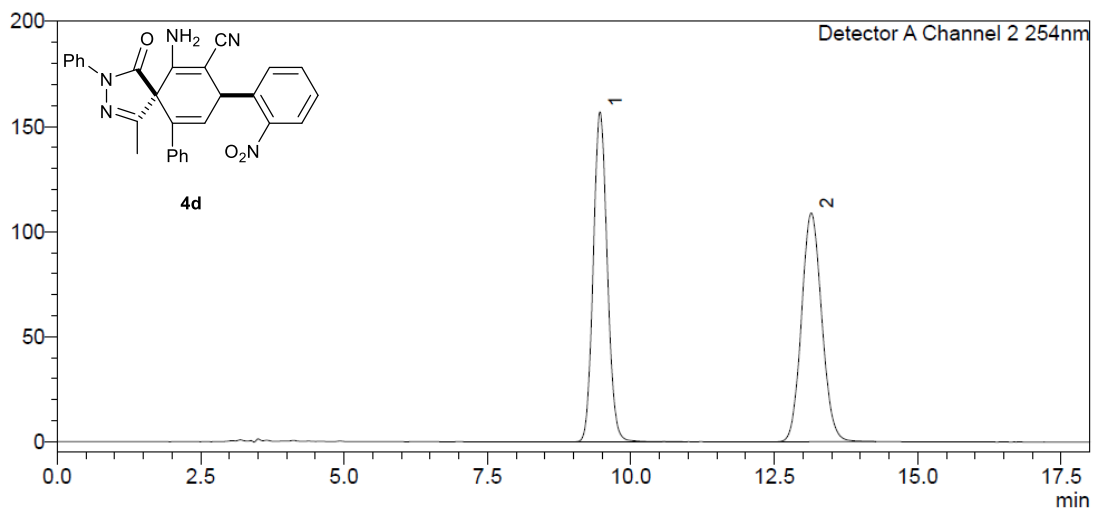


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	6.571	209208	22202	4.567	4.567
2	9.299	4371653	240375	95.433	95.433
Total		4580861	262577		100.000

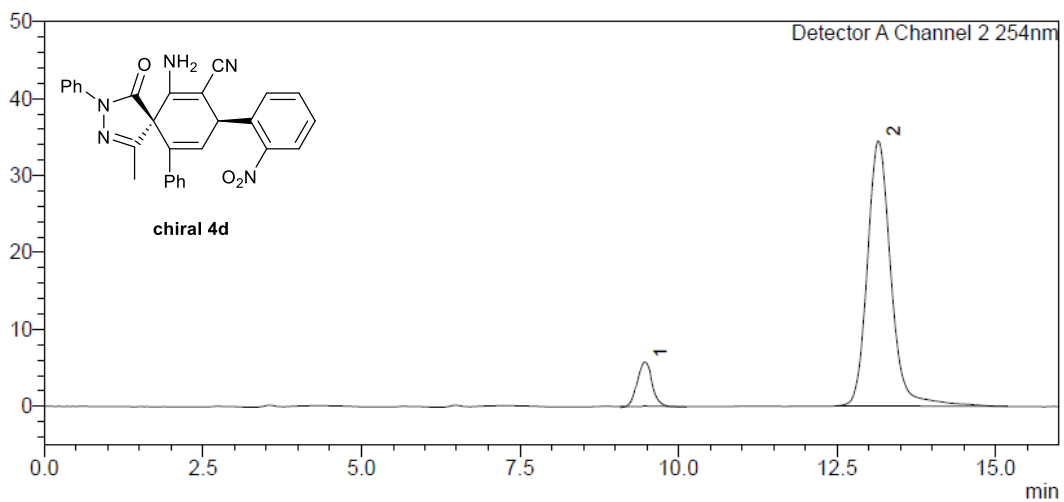




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Detector A Channel 2 254nm

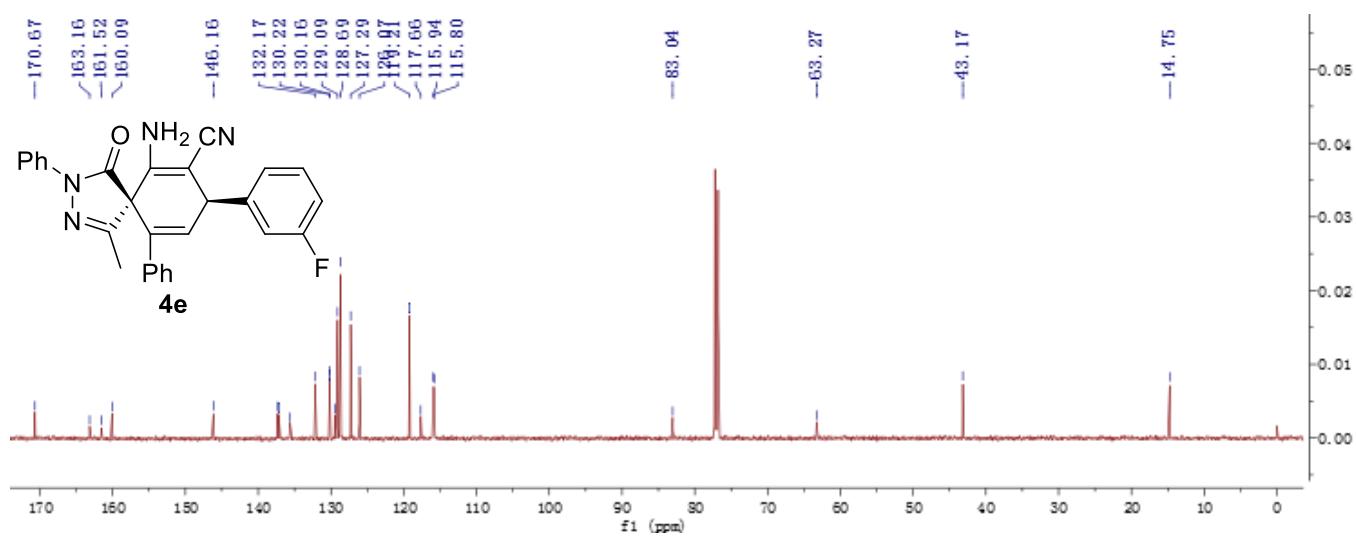
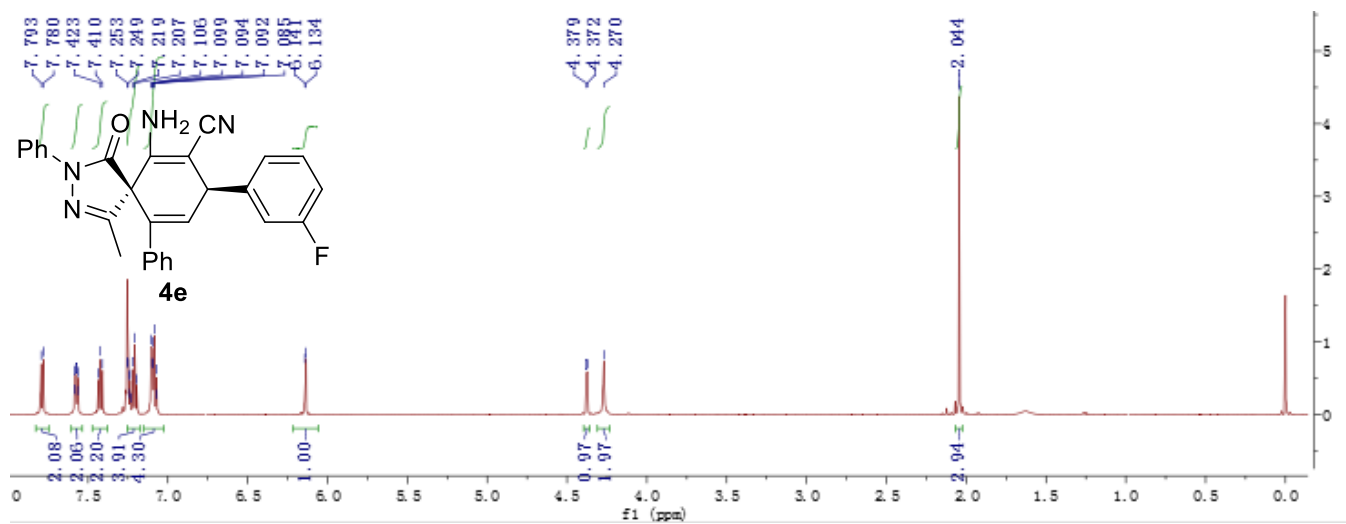
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	9.462	2667202	157005	49.979	49.979
2	13.144	2669416	108842	50.021	50.021
Total		5336618	265847		100.000

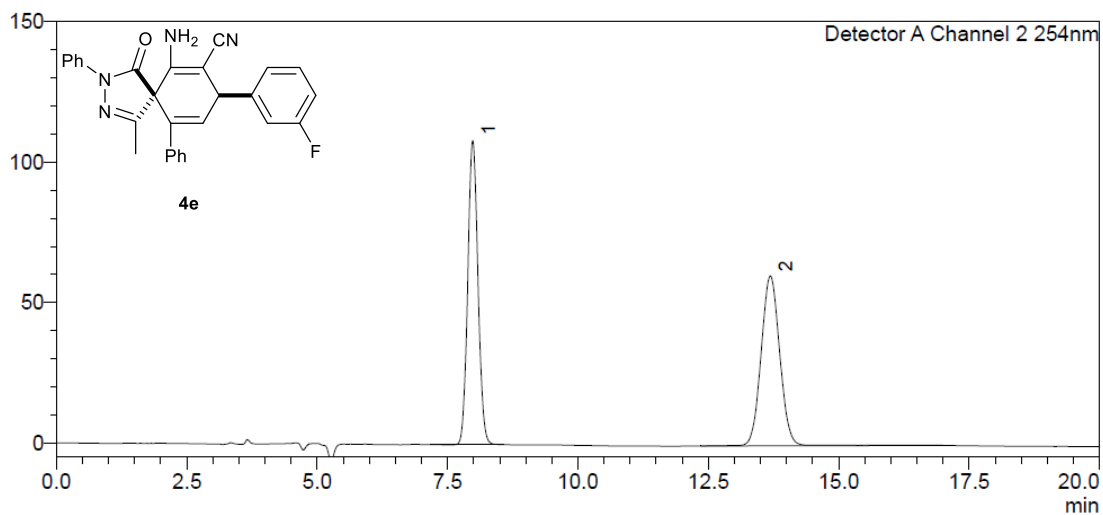


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	9.472	77179	5145	7.962	7.962
2	13.152	892151	34460	92.038	92.038
Total		969330	39605		100.000

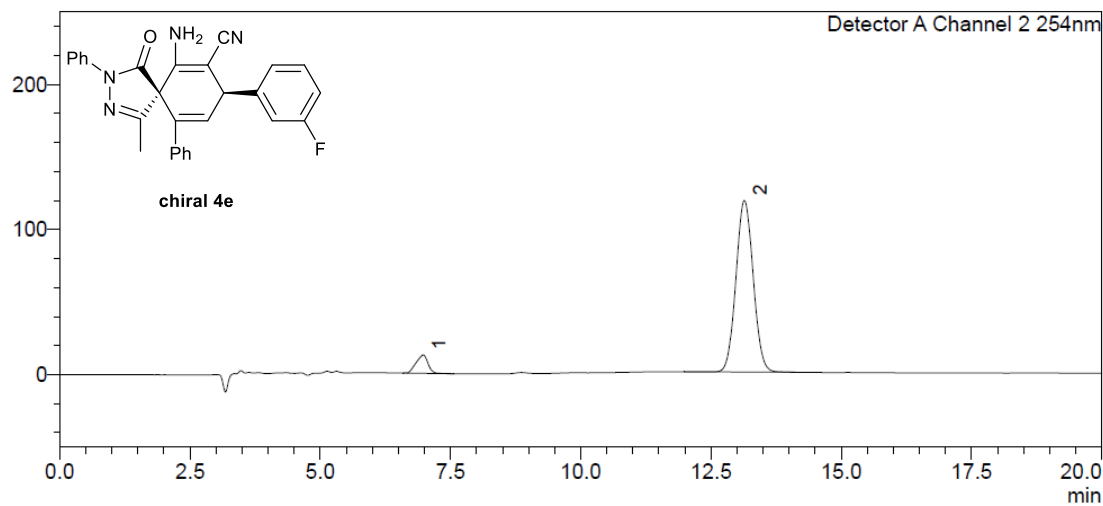




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Detector A Channel 2 254nm

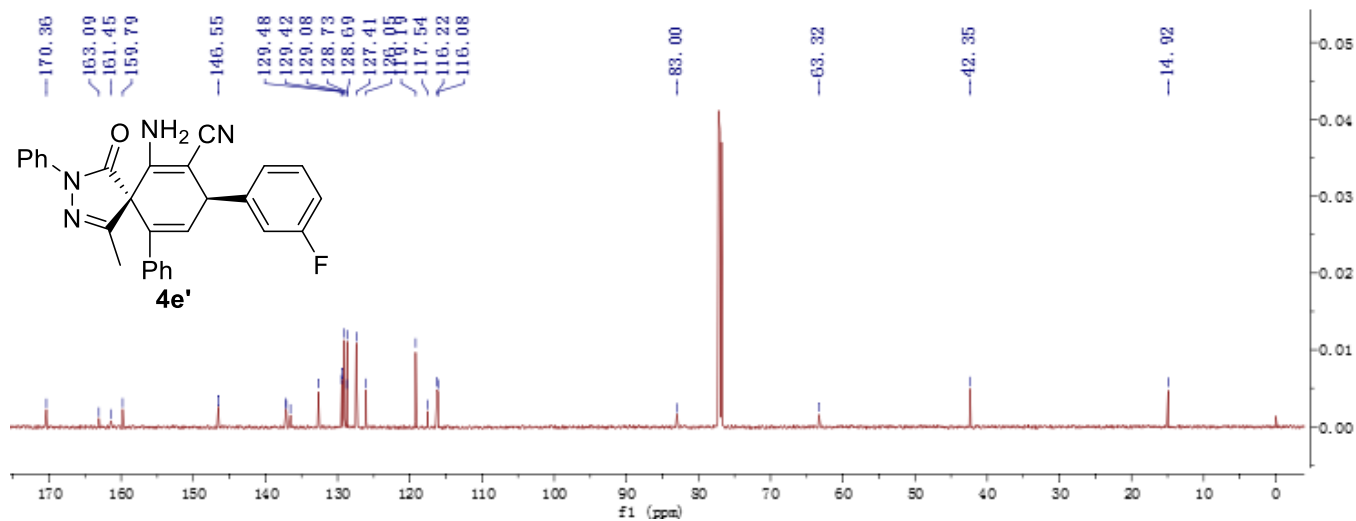
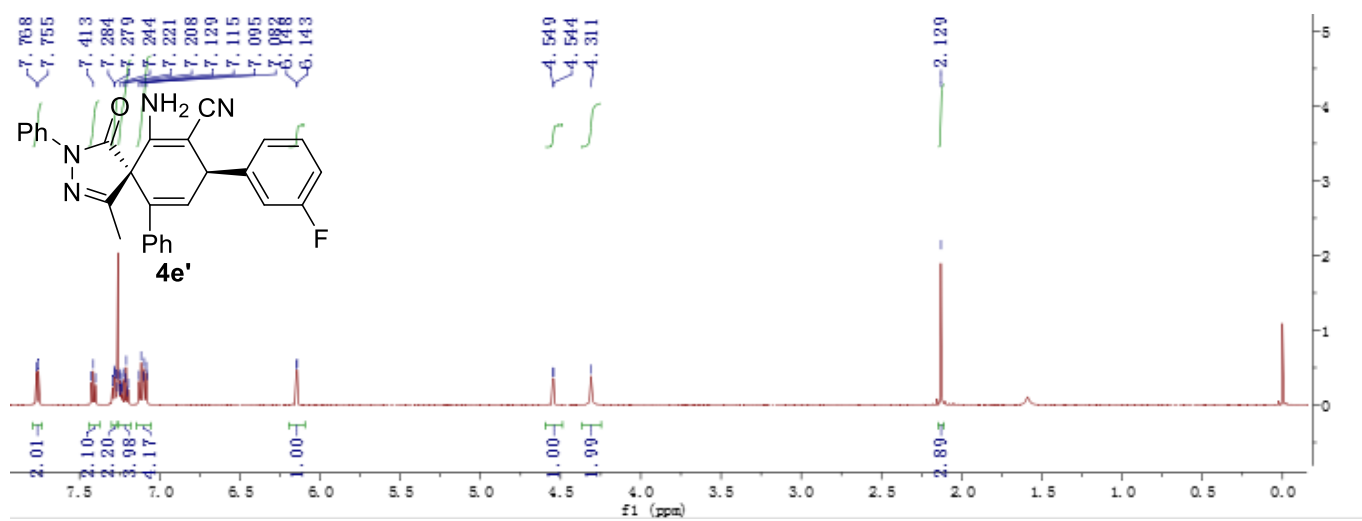
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	7.982	1476125	108382	49.427	49.427
2	13.687	1510330	60490	50.573	50.573
Total		2986455	168871		100.000

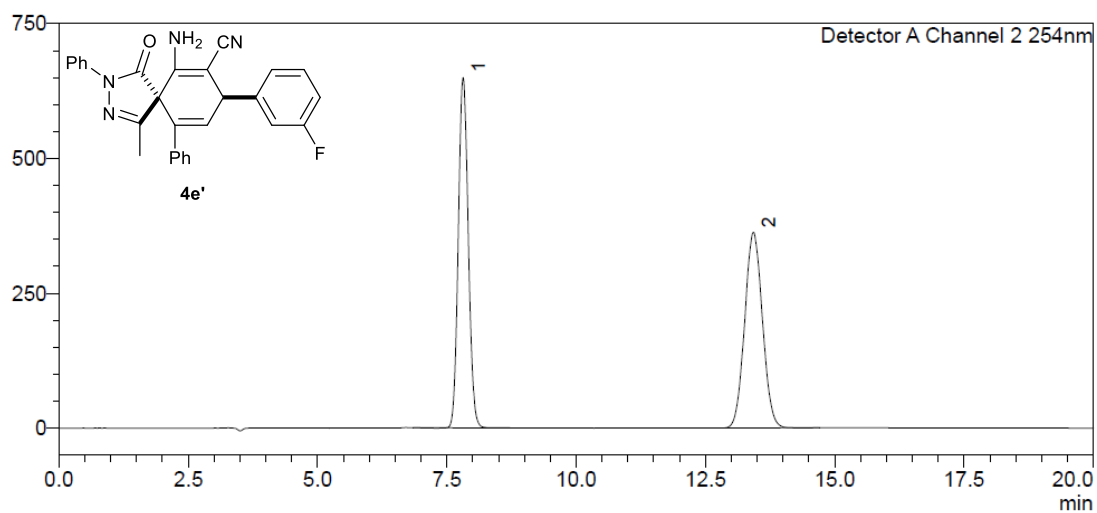


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	6.974	199951	12706	6.814	6.814
2	13.141	2734645	118432	93.186	93.186
Total		2934596	131137		100.000

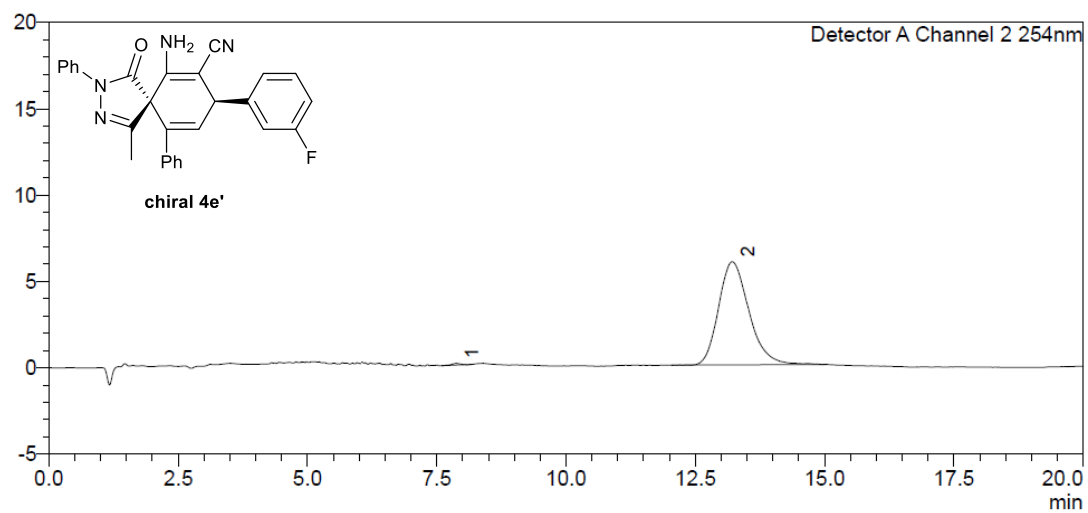




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Detector A Channel 2 254nm

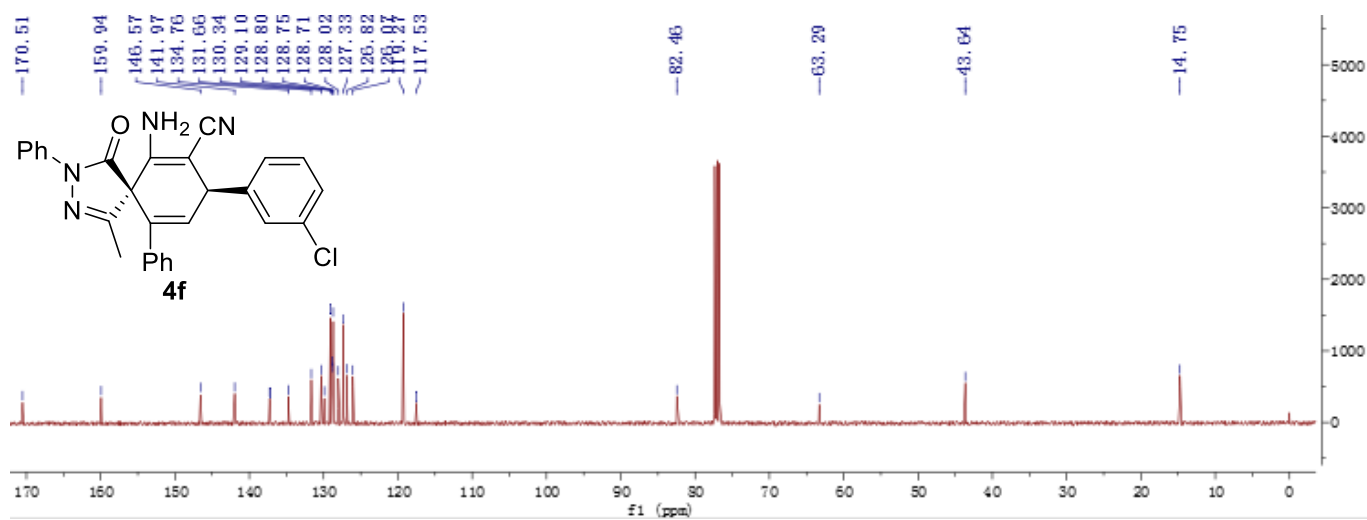
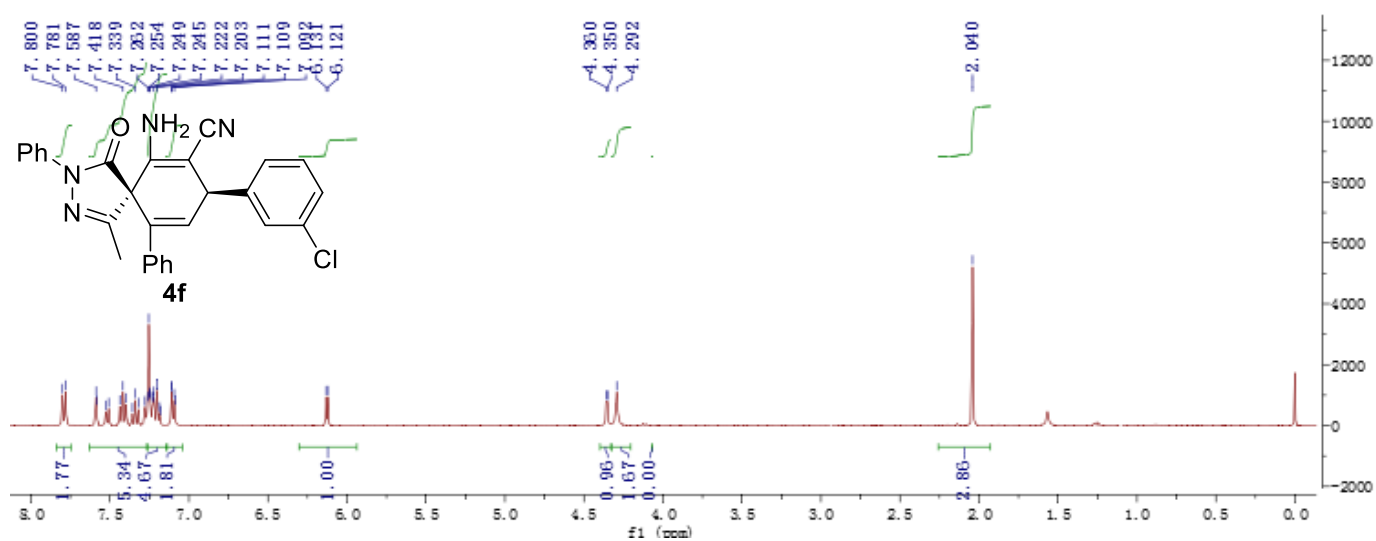
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	7.814	8672651	649737	50.030	50.030
2	13.427	8662290	362904	49.970	49.970
Total		17334941	1012641		100.000

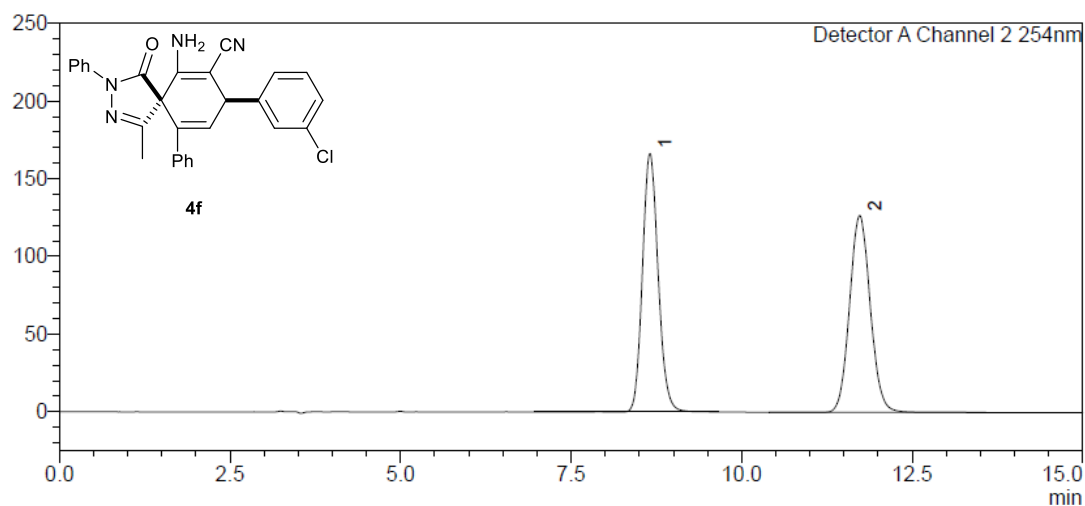


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	7.875	1007	104	0.410	0.410
2	13.211	244291	5971	99.590	99.590
Total		245298	6074		100.000

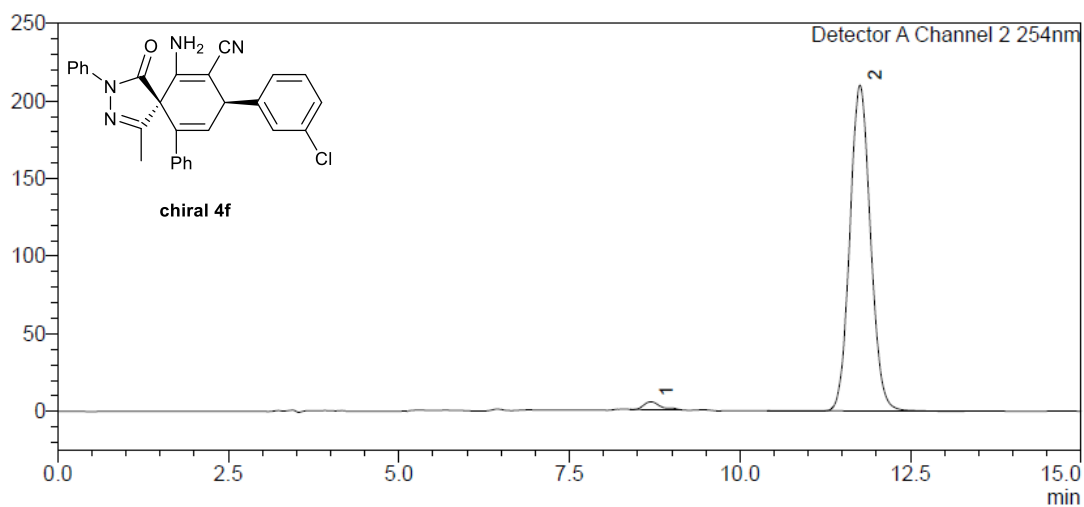




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Detector A Channel 2 254nm

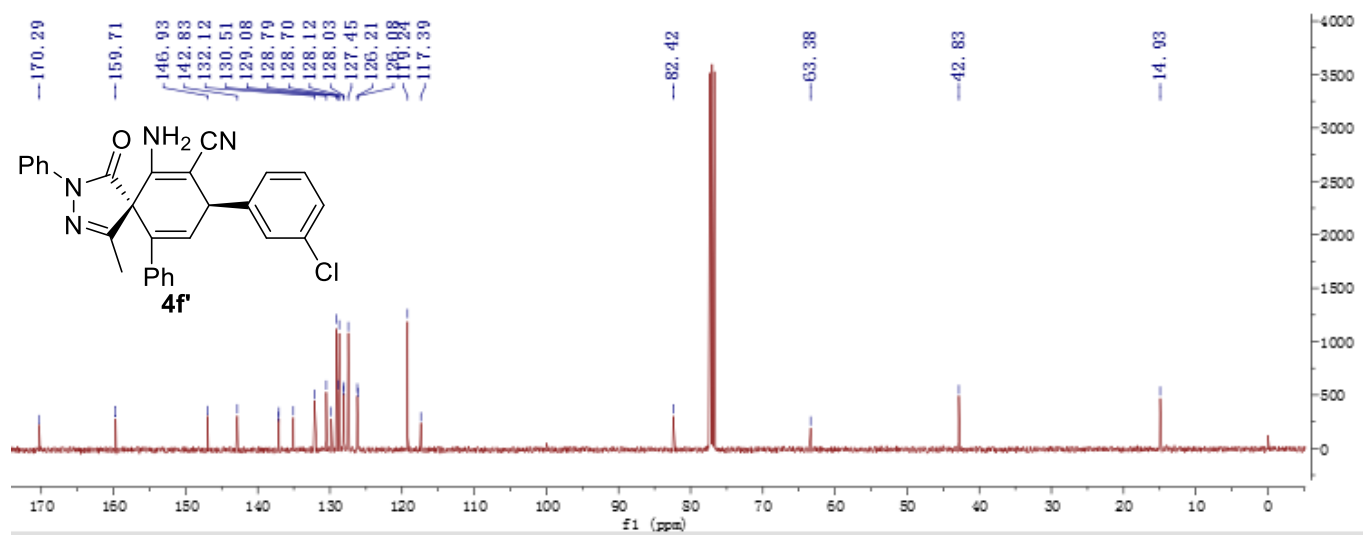
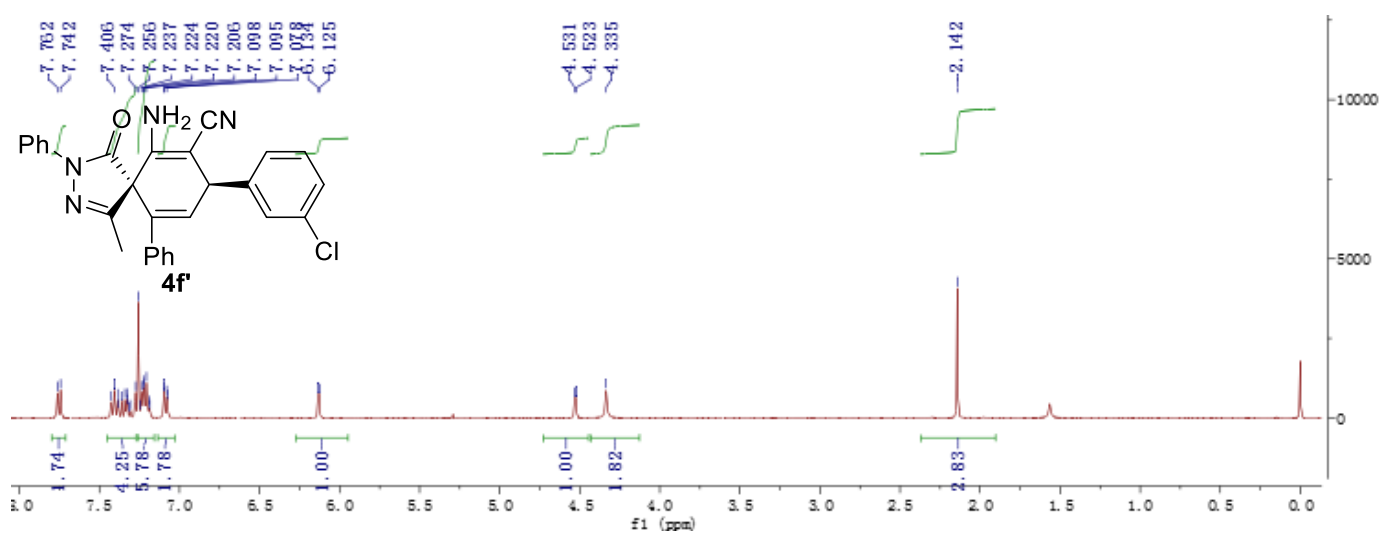
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	8.656	2666037	166408	49.910	49.910
2	11.731	2675623	126727	50.090	50.090
Total		5341660	293135		100.000

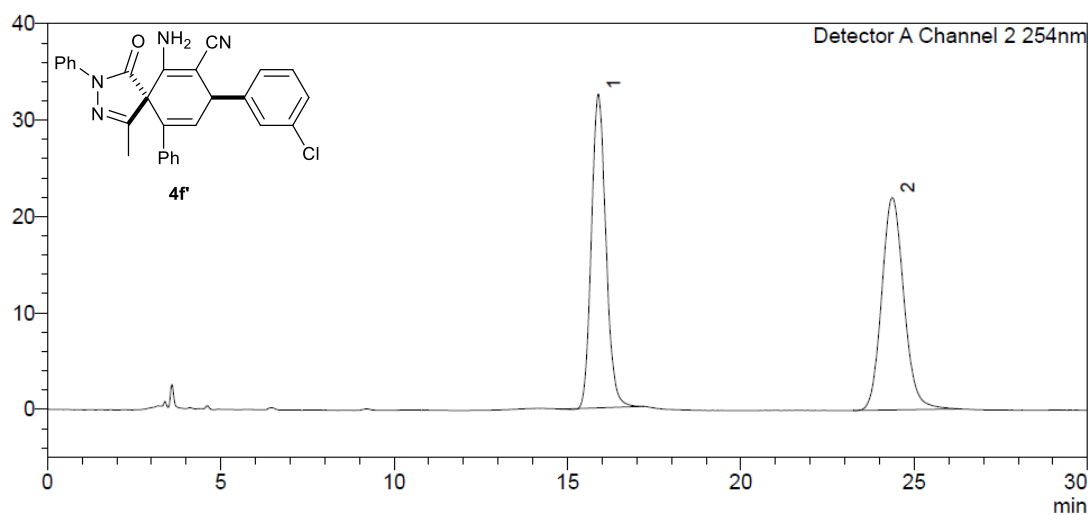


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	8.694	86419	5116	1.920	1.920
2	11.759	4414240	209927	98.080	98.080
Total		4500659	215043		100.000

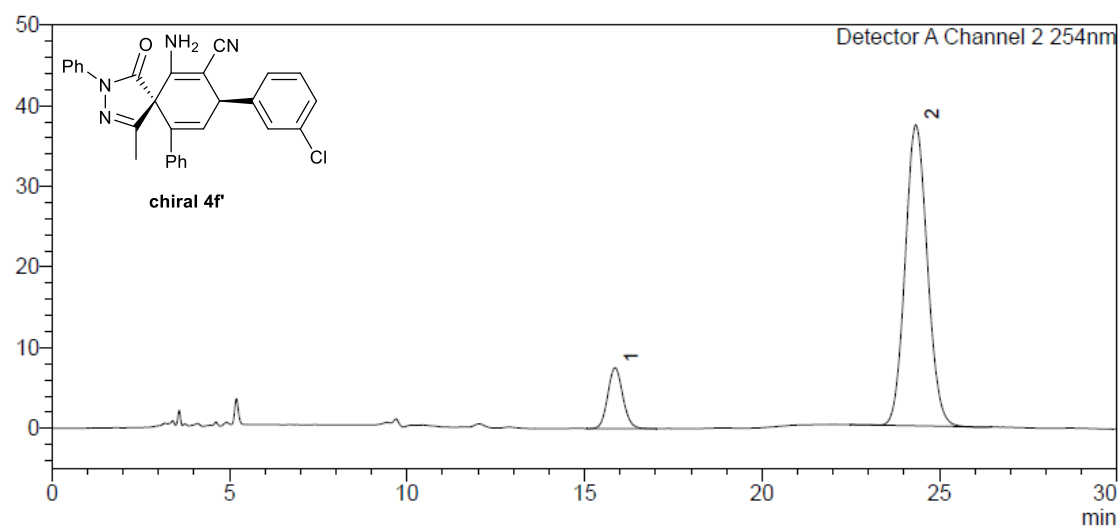




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Detector A Channel 2 254nm

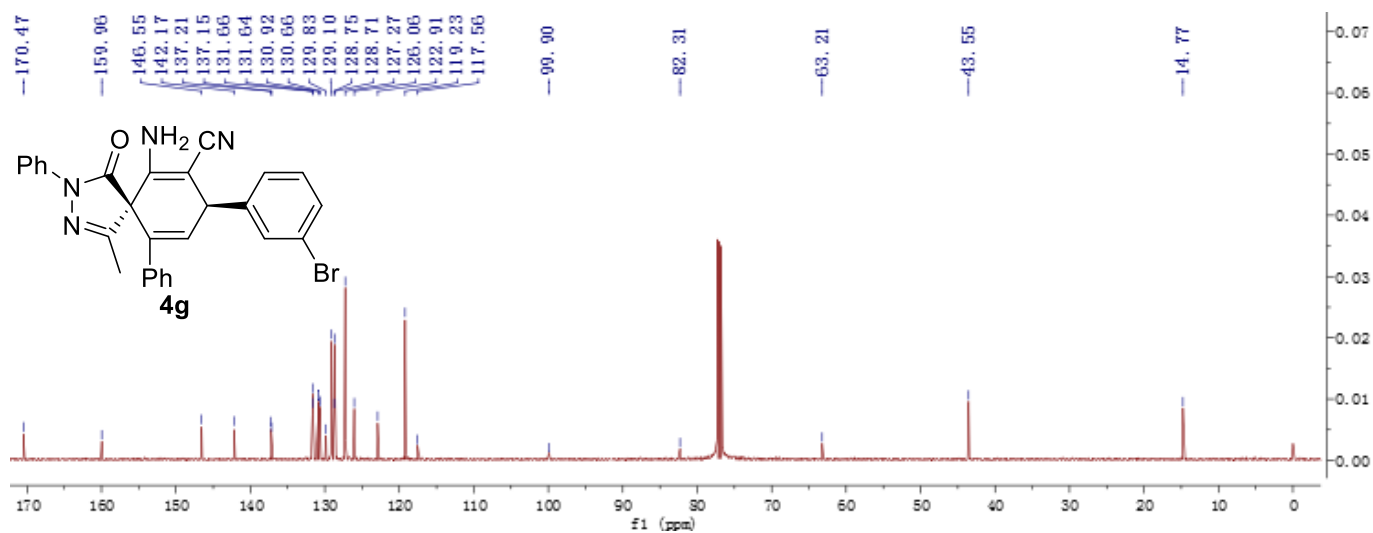
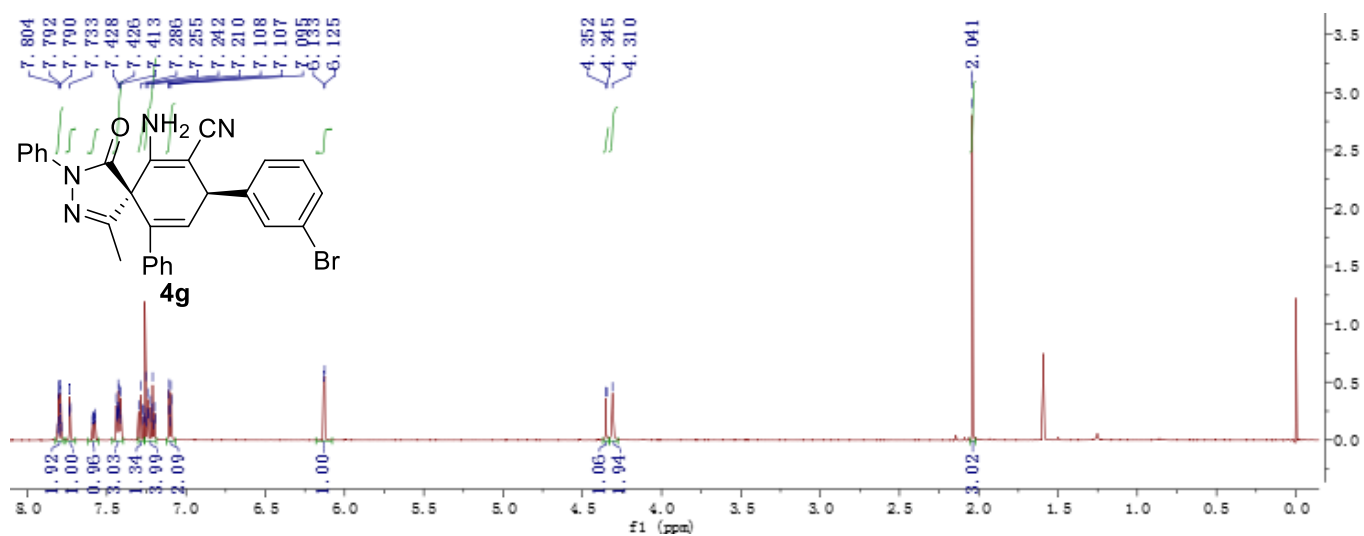
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	15.889	946442	32571	49.074	49.074
2	24.369	982145	22046	50.926	50.926
Total		1928588	54617		100.000

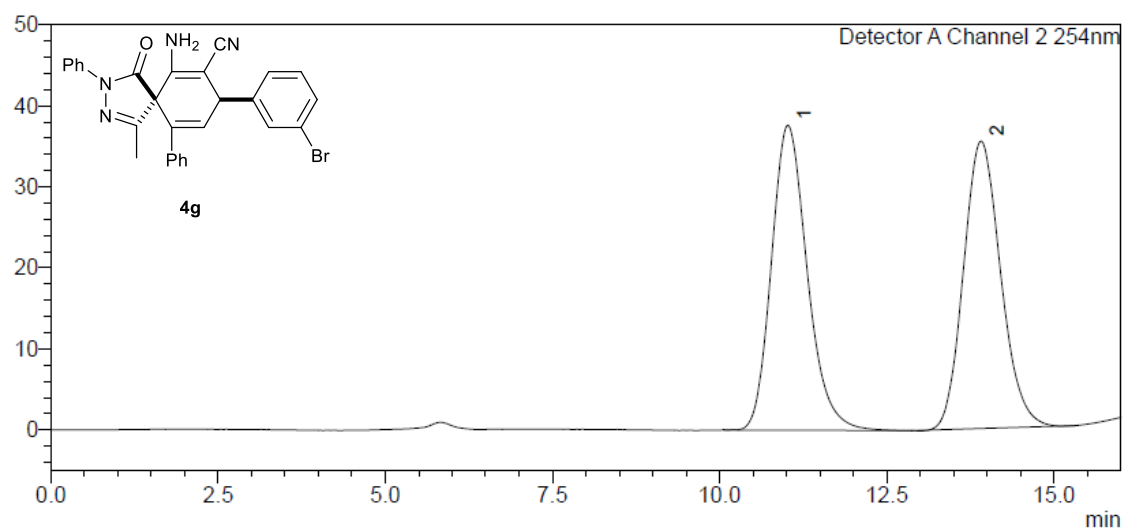


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	15.860	222400	7550	12.071	12.071
2	24.335	1620071	37323	87.929	87.929
Total		1842470	44873		100.000

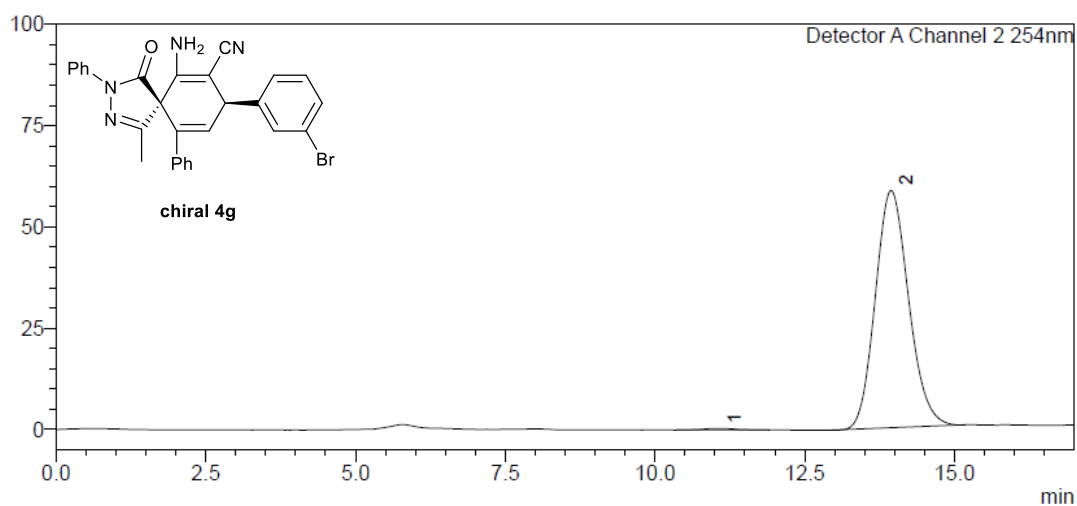




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Detector A Channel 2 254nm

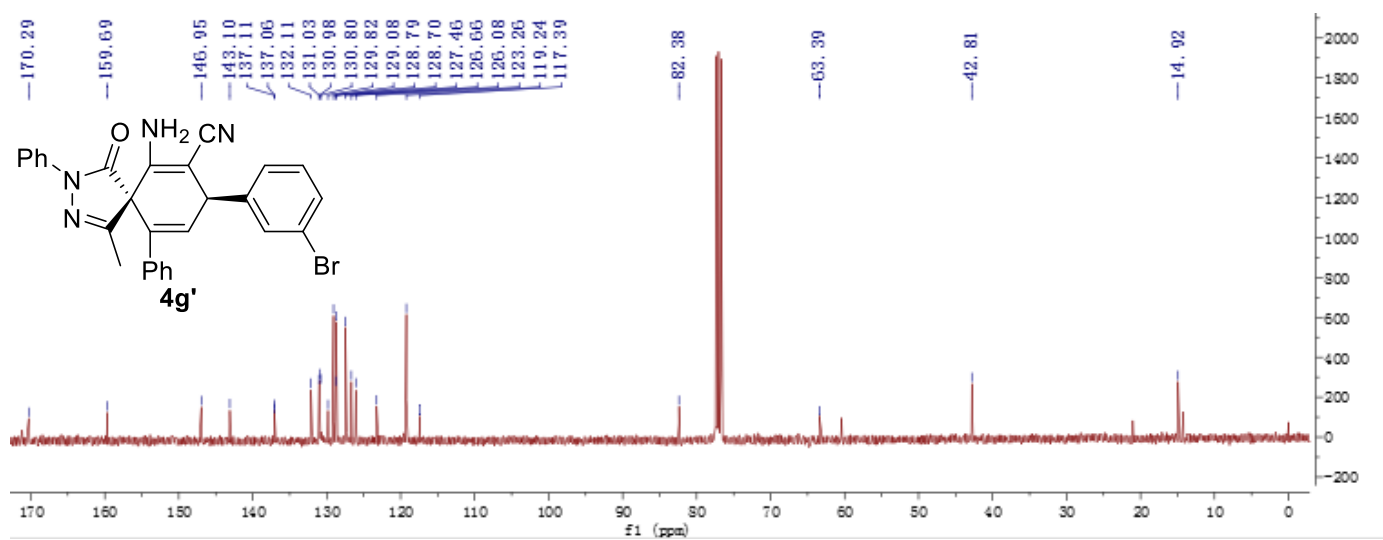
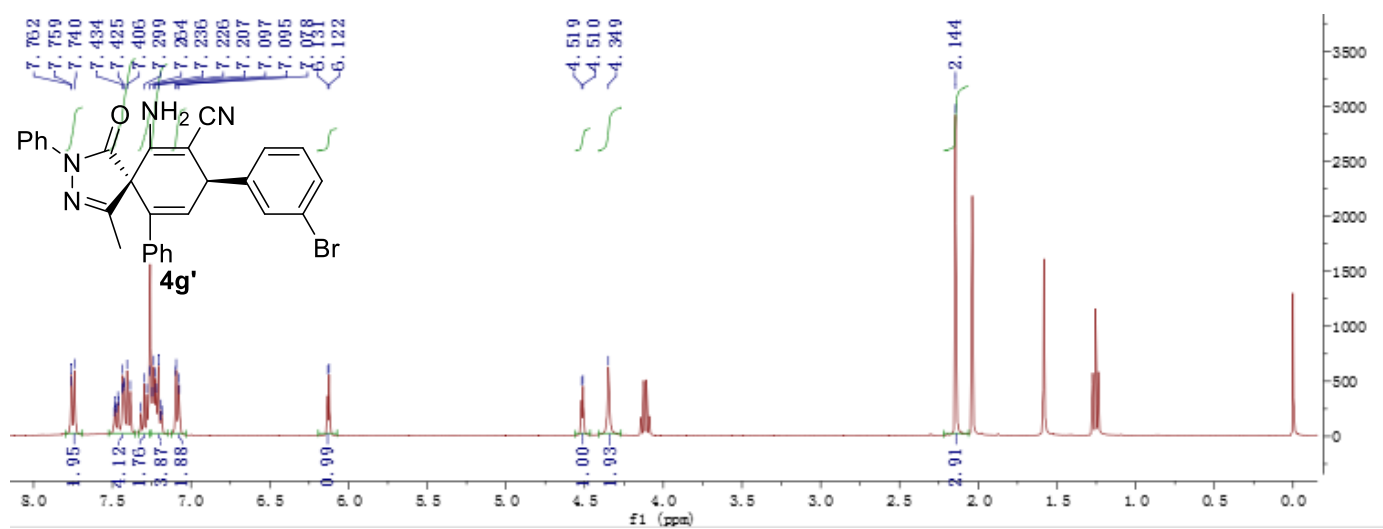
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	11.021	1401330	37623	50.552	50.552
2	13.910	1370735	35468	49.448	49.448
Total		2772064	73091		100.000

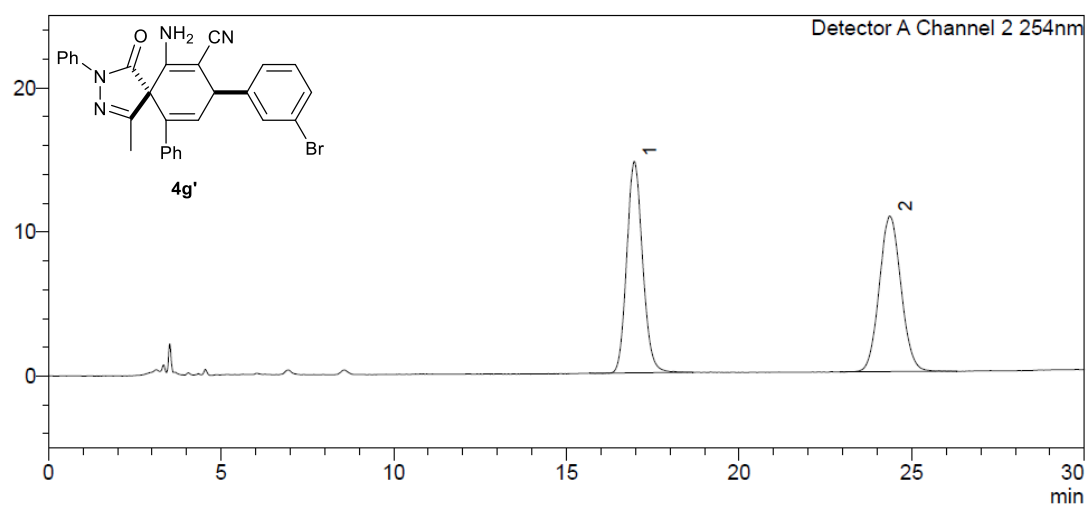


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	11.070	11323	366	0.498	0.498
2	13.941	2261001	58566	99.502	99.502
Total		2272324	58932		100.000

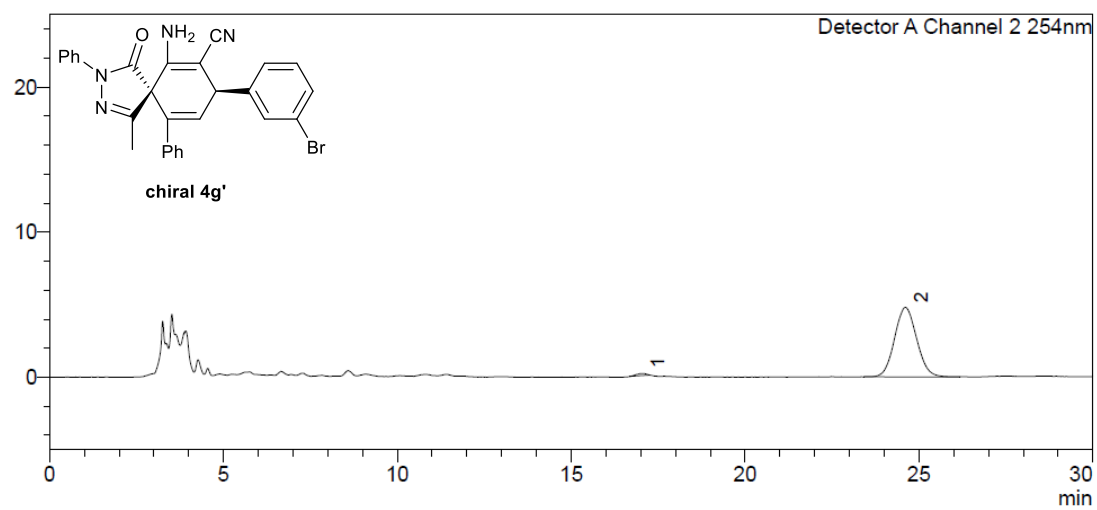




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Detector A Channel 2 254nm

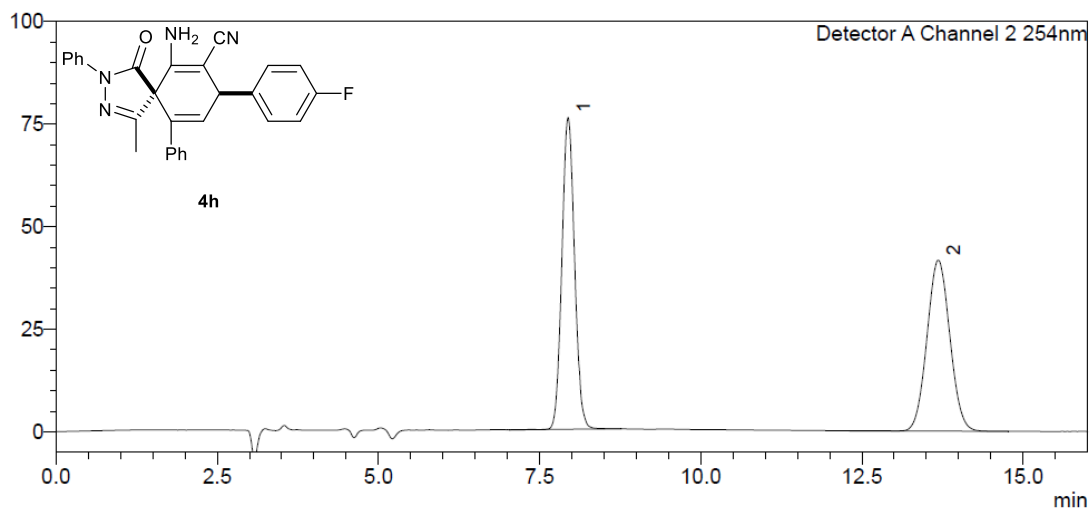
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	16.965	470311	14685	49.873	49.873
2	24.365	472709	10792	50.127	50.127
Total		943021	25477		100.000



<Peak Table>

Detector A Channel 2 254nm

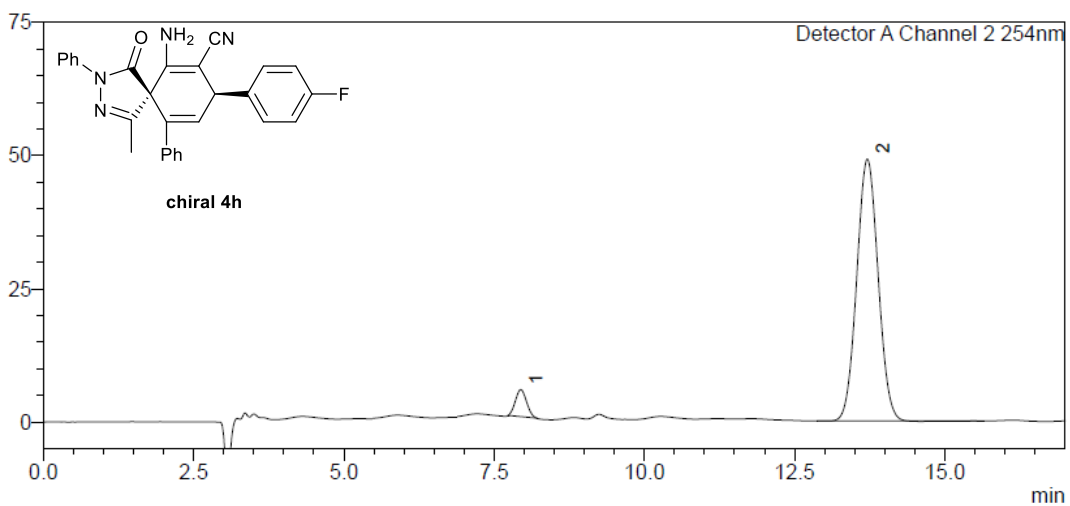
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	17.038	2869	143	1.335	1.335
2	24.611	212020	4799	98.665	98.665
Total		214889	4942		100.000



<Peak Table>

Detector A Channel 2 254nm

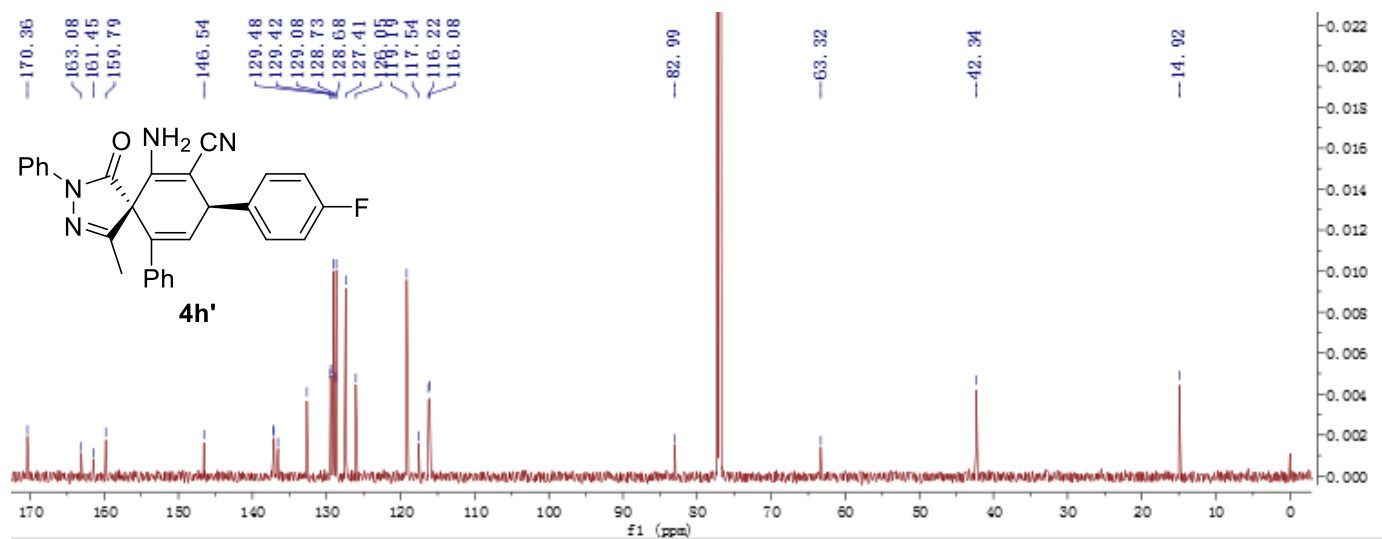
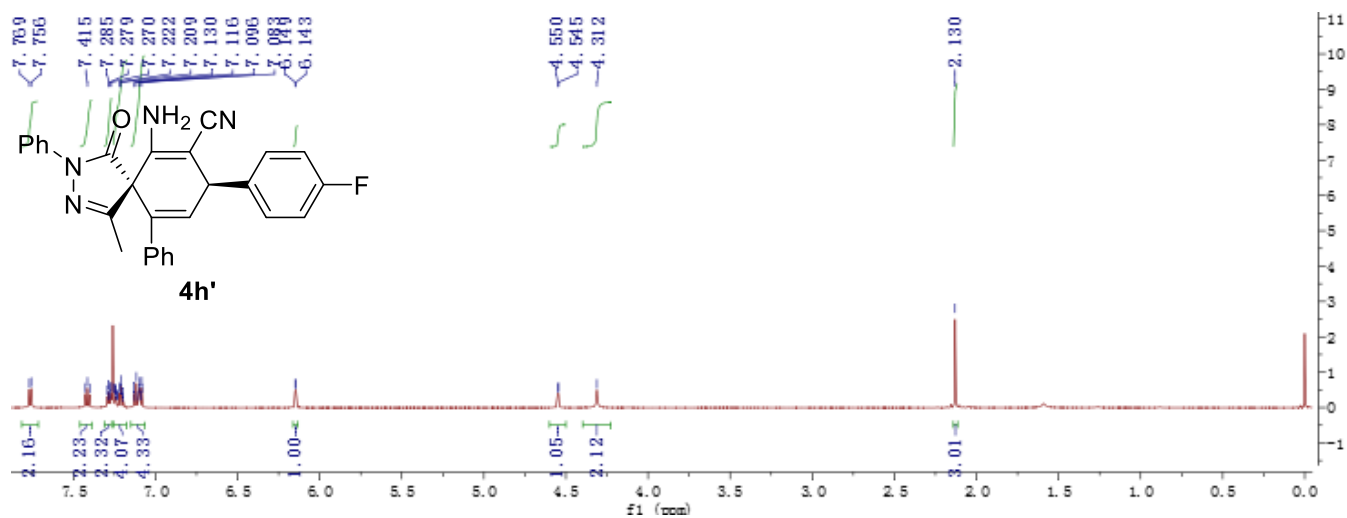
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	7.941	1036573	76087	50.252	50.252
2	13.683	1026190	41681	49.748	49.748
Total		2062763	117768		100.000

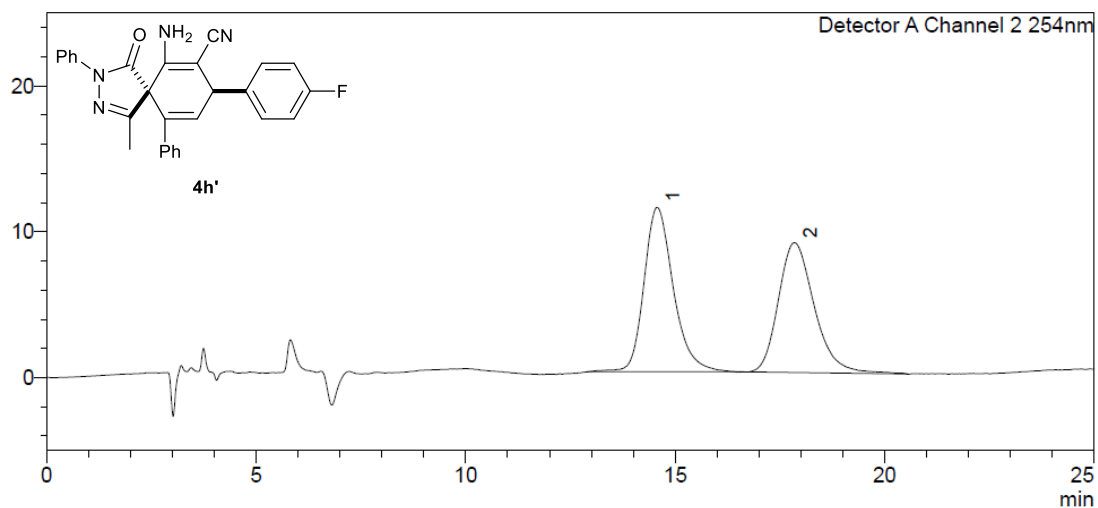


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	7.943	65401	5076	5.114	5.114
2	13.710	1213444	49167	94.886	94.886
Total		1278845	54242		100.000

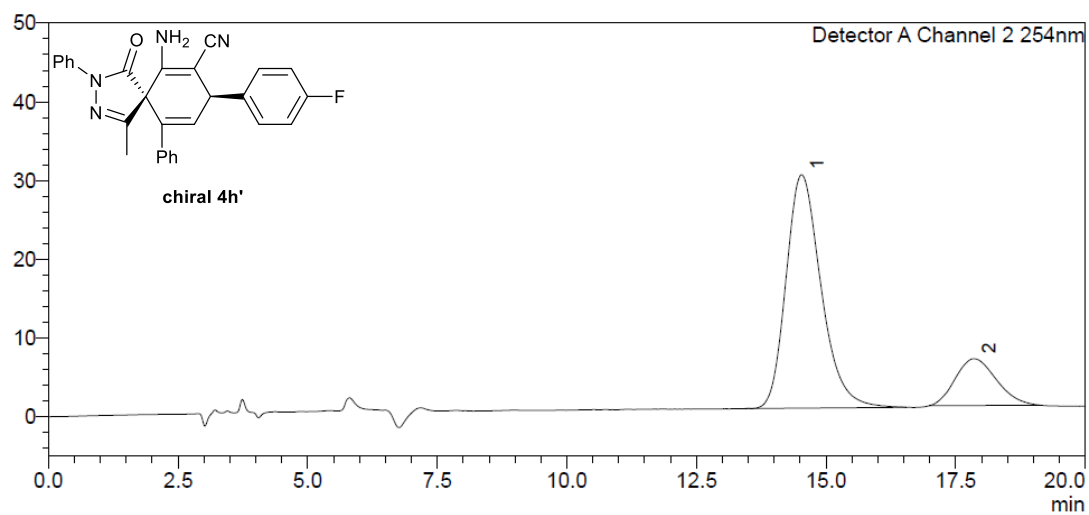




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Detector A Channel 2 254nm

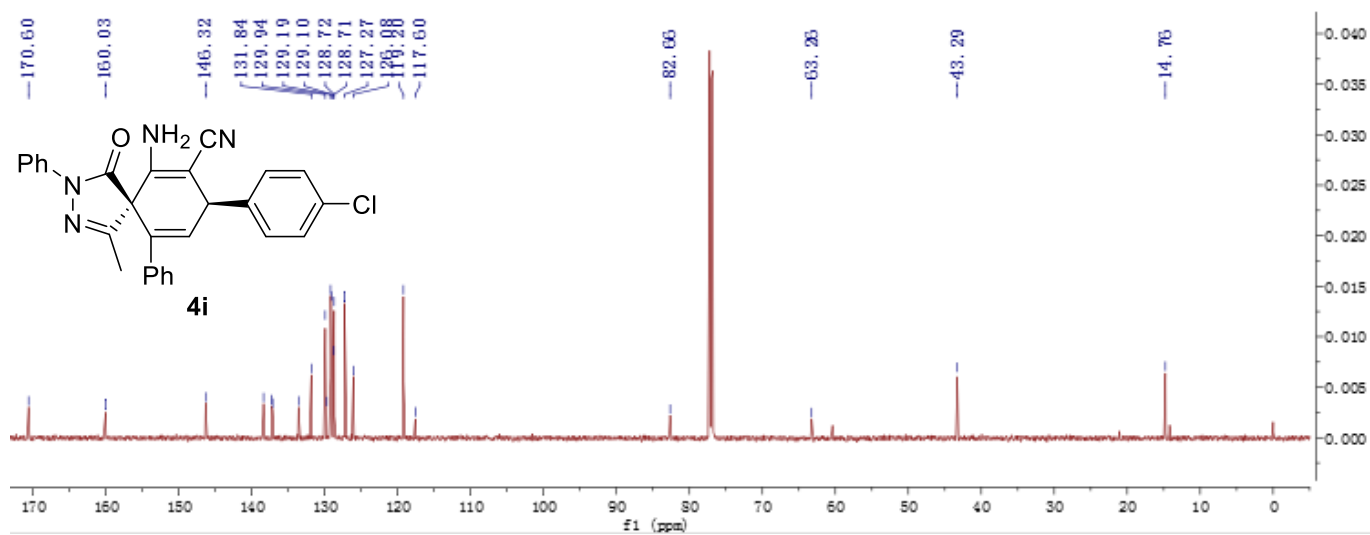
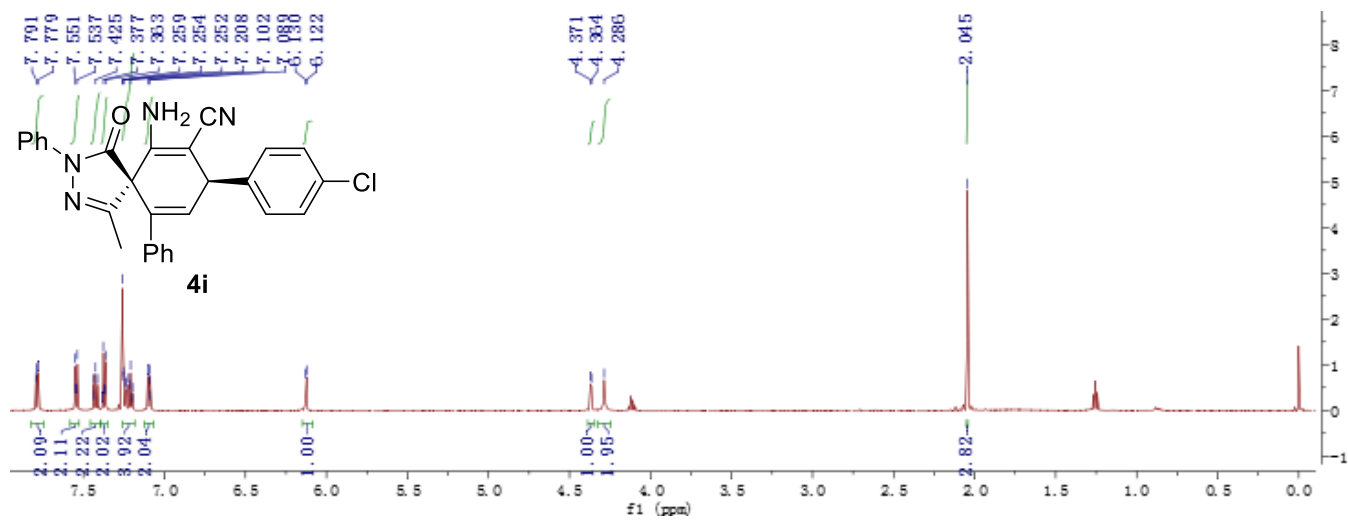
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	14.569	551442	11286	50.893	50.893
2	17.849	532081	8908	49.107	49.107
Total		1083522	20194		100.000

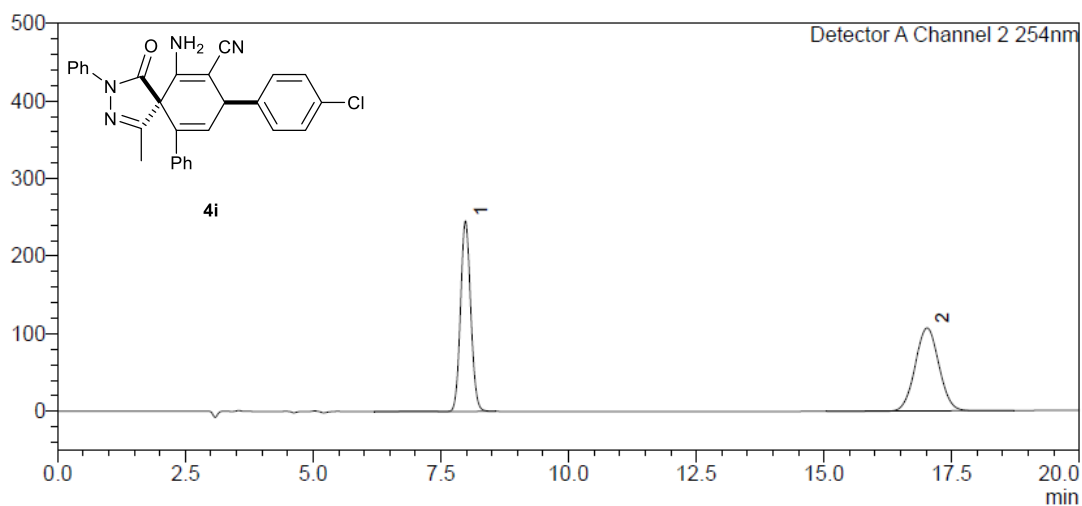


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	14.528	1393699	29653	80.948	80.948
2	17.840	328014	5920	19.052	19.052
Total		1721713	35573		100.000

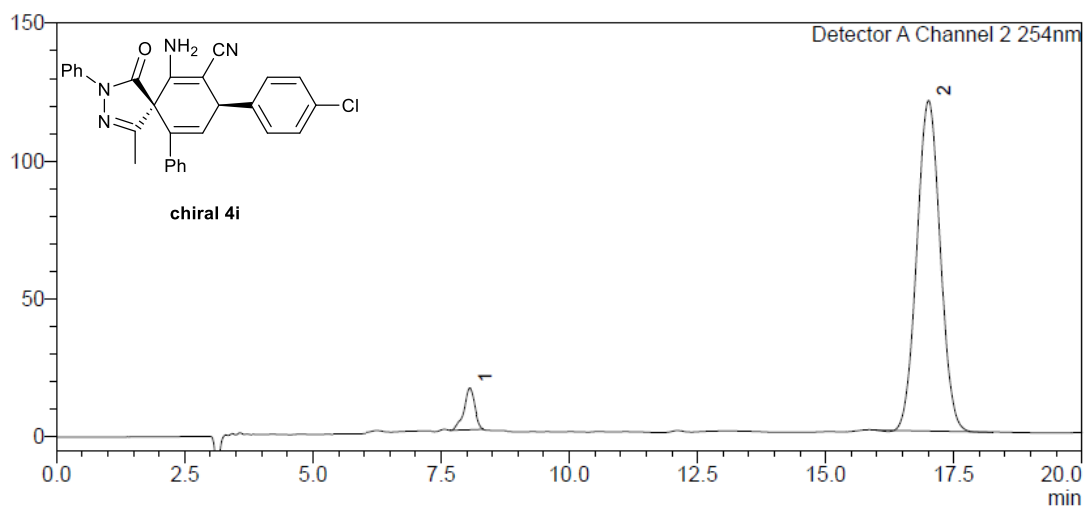




<Peak Table>

Detector A Channel 2 254nm

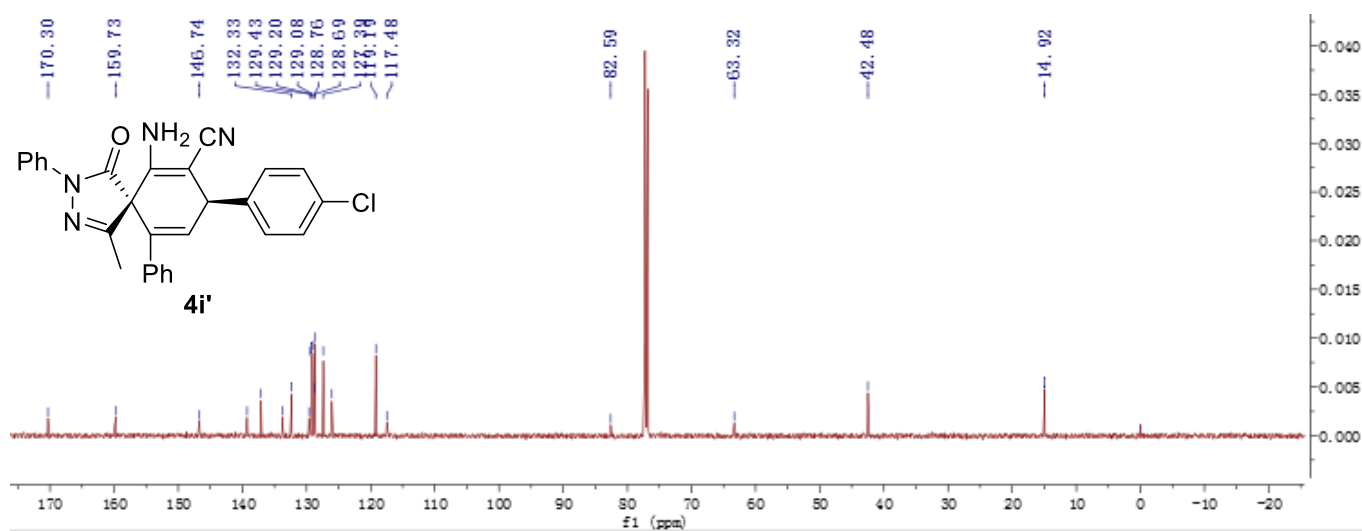
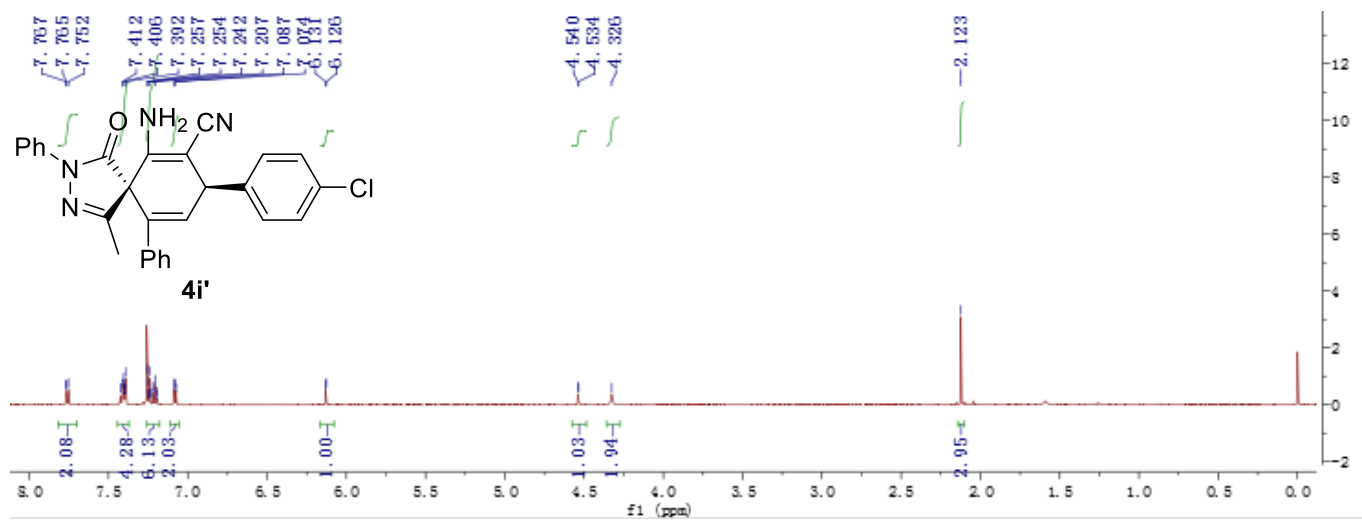
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	7.982	3441669	245478	50.110	50.110
2	17.018	3426609	106953	49.890	49.890
Total		6868278	352431		100.000

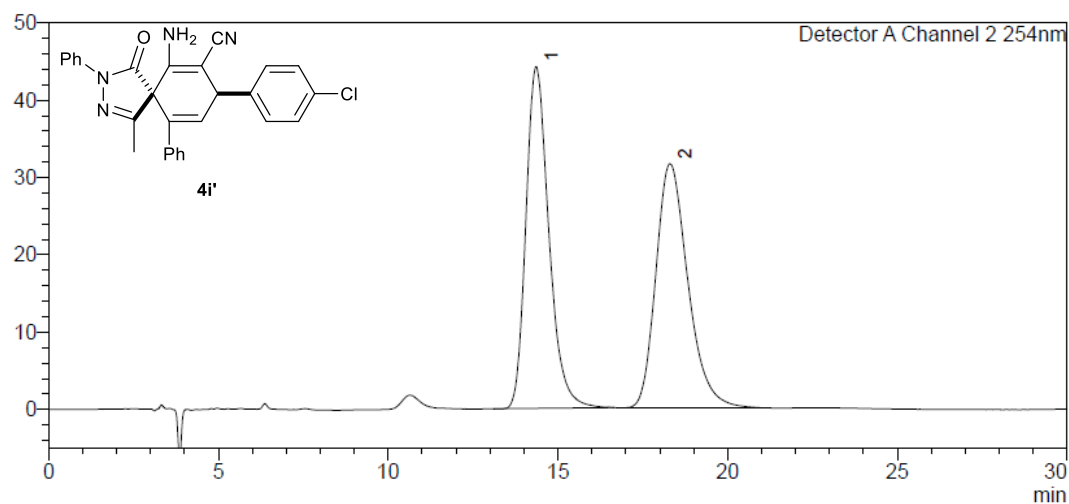


<Peak Table>

Detector A Channel 2 254nm

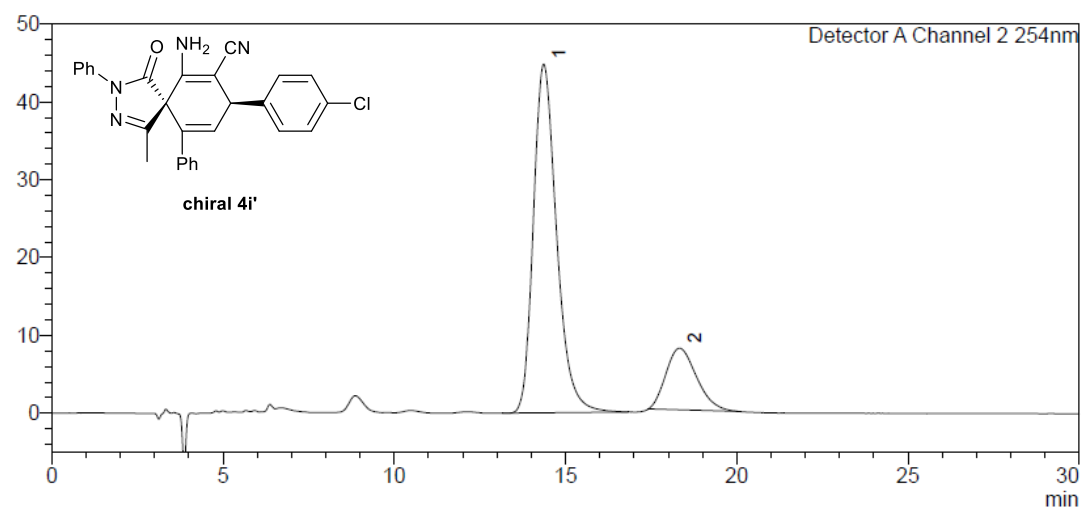
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	8.062	246634	15174	6.103	6.103
2	17.008	3794477	119824	93.897	93.897
Total		4041111	134998		100.000





Detector A Channel 2 254nm

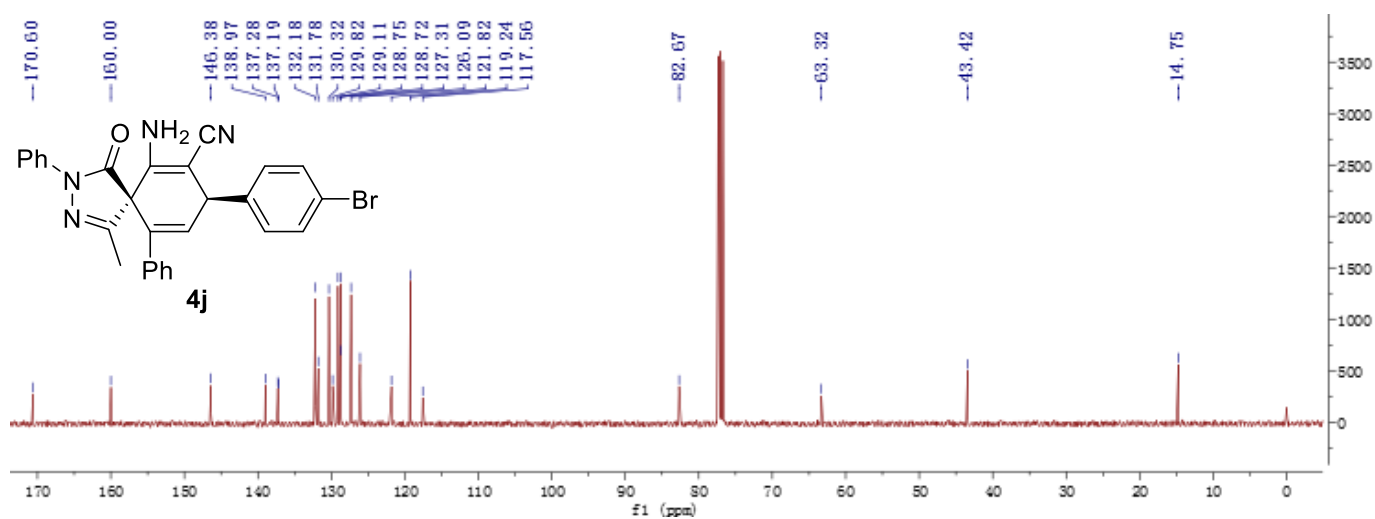
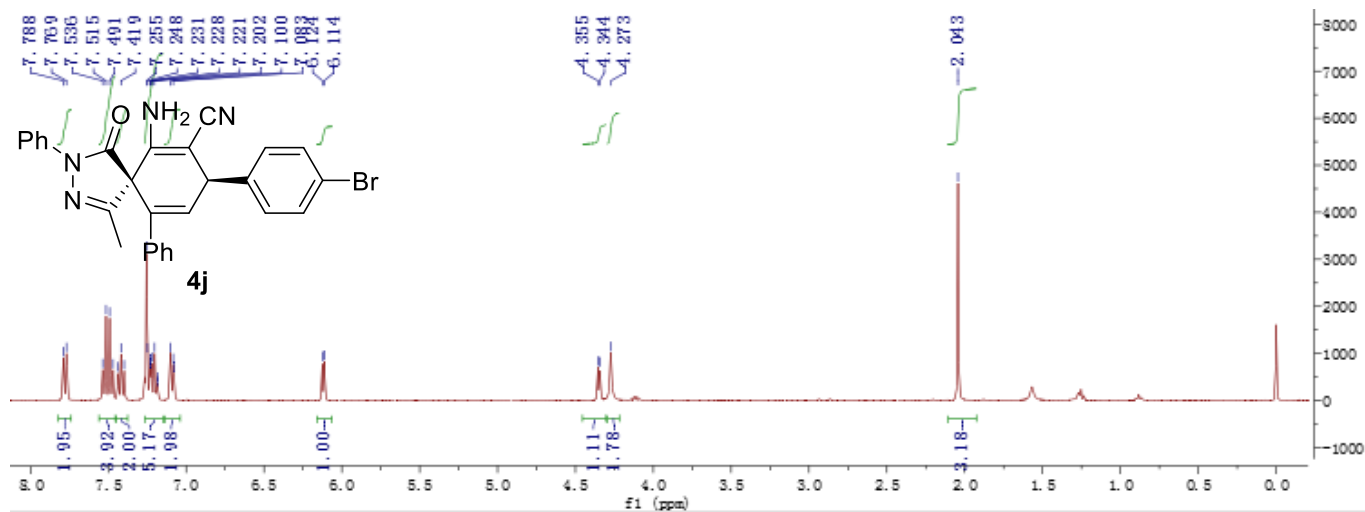
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	14.358	2075120	44206	50.099	50.099
2	18.303	2066915	31577	49.901	49.901
Total		4142035	75783		100.000

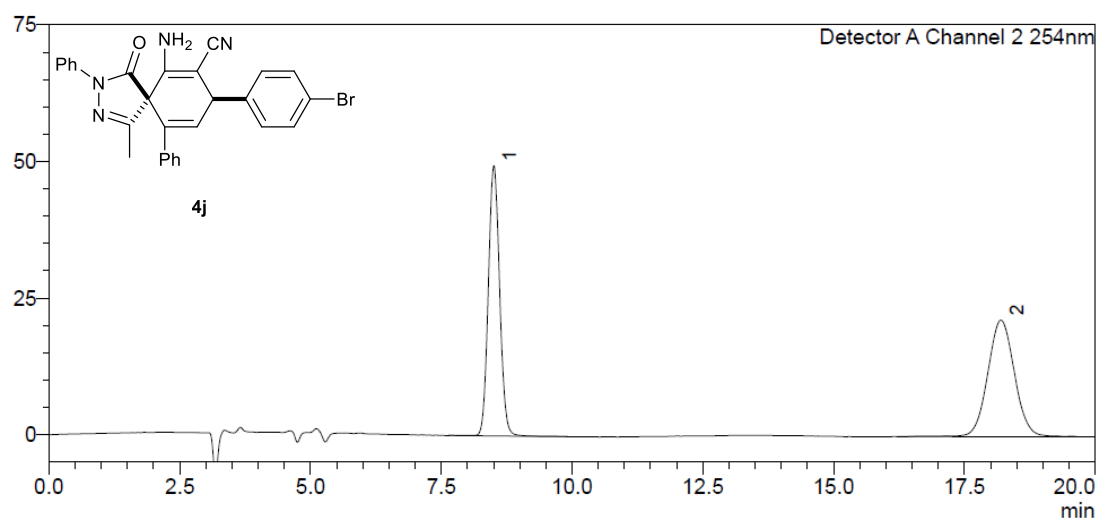


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	14.362	2111585	44789	81.069	81.069
2	18.325	493098	7906	18.931	18.931
Total		2604682	52696		100.000

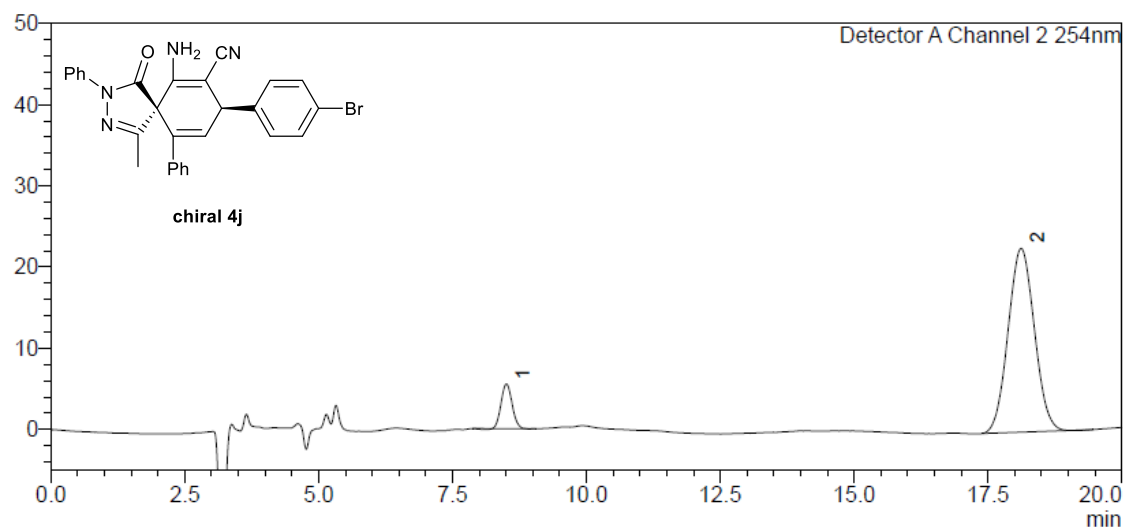




<Peak Table>

Detector A Channel 2 254nm

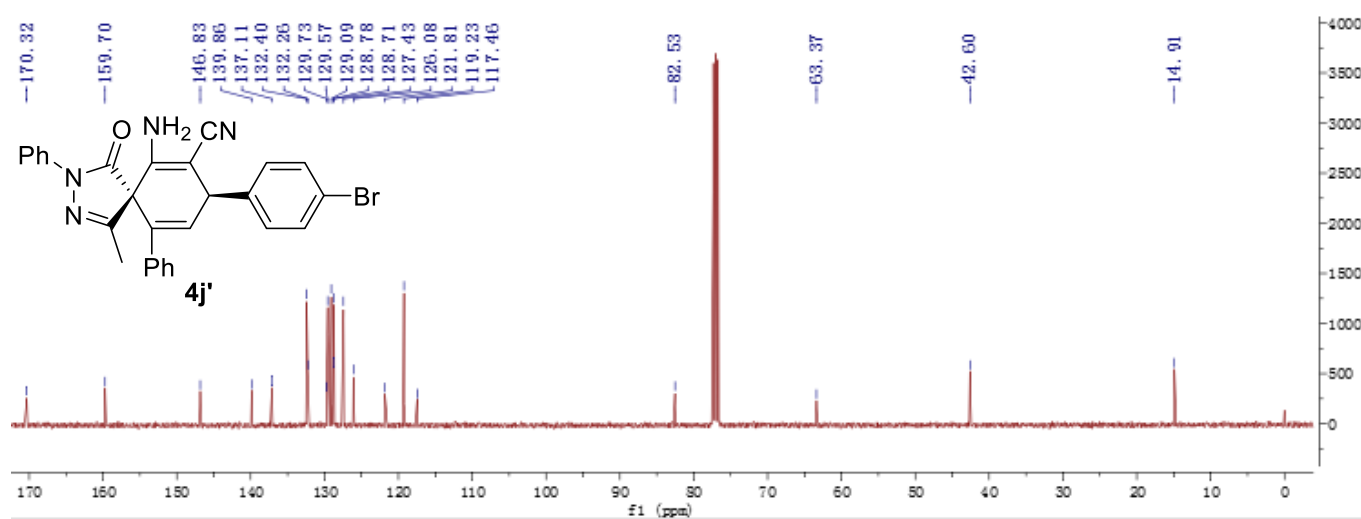
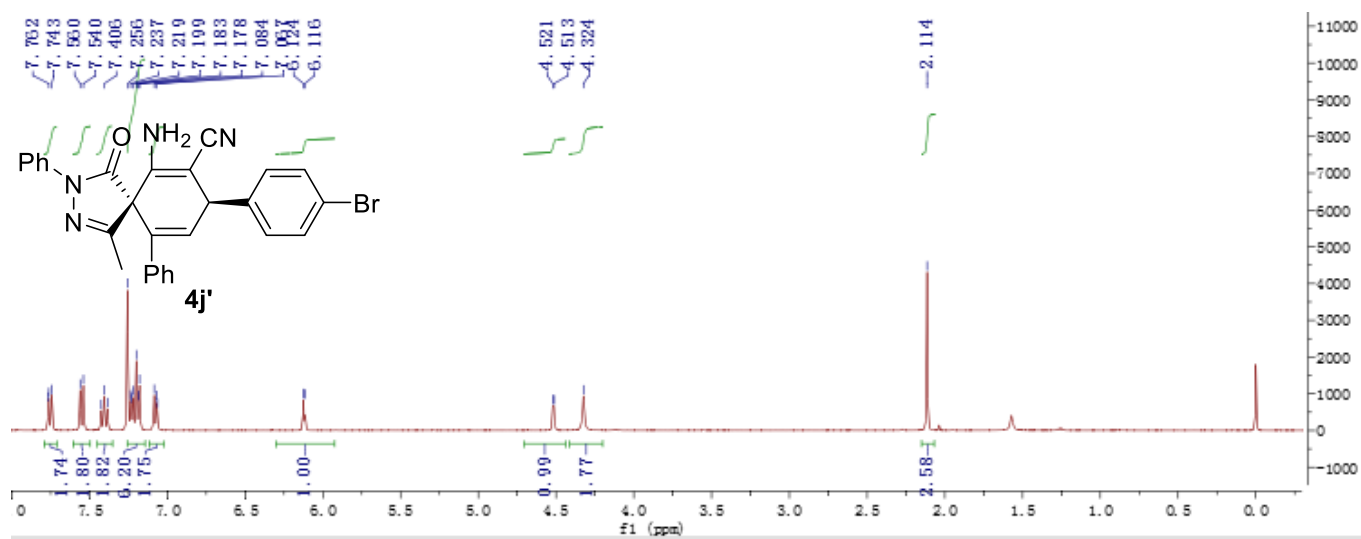
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	8.504	741991	49450	49.445	49.445
2	18.199	758636	21343	50.555	50.555
Total		1500627	70793		100.000

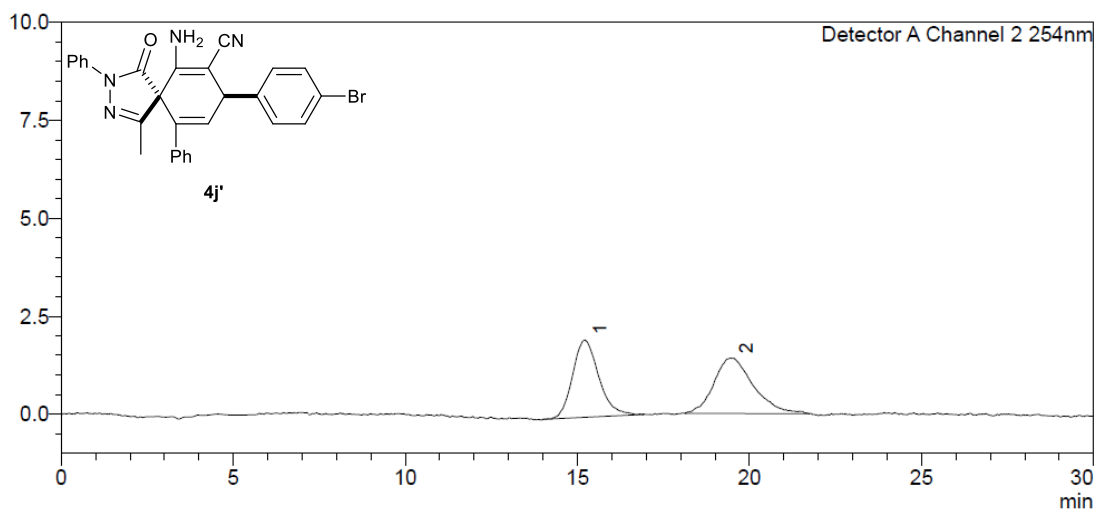


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	8.506	77213	5502	9.096	9.096
2	18.120	771670	22632	90.904	90.904
Total		848883	28134		100.000

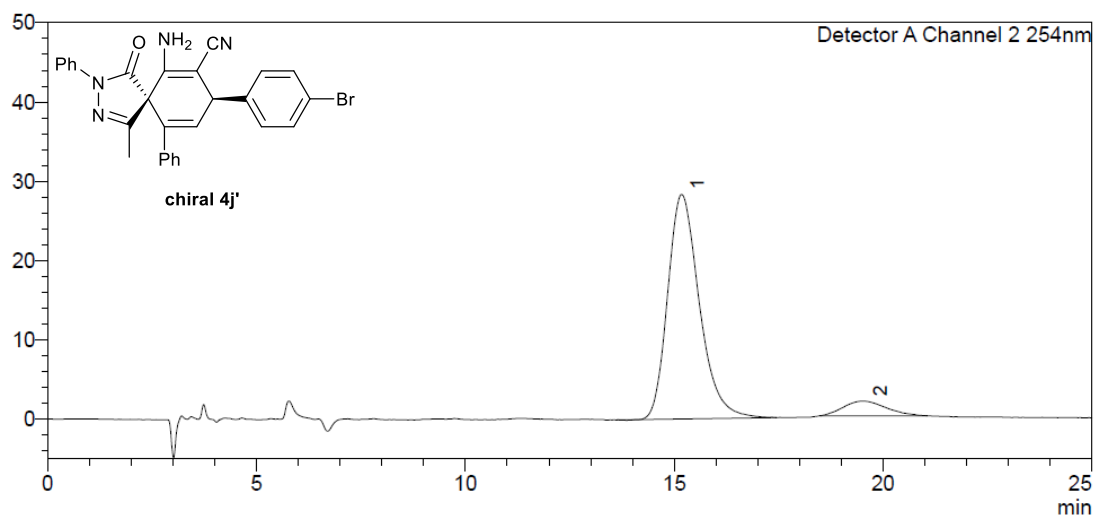




<Peak Table>

Detector A Channel 2 254nm

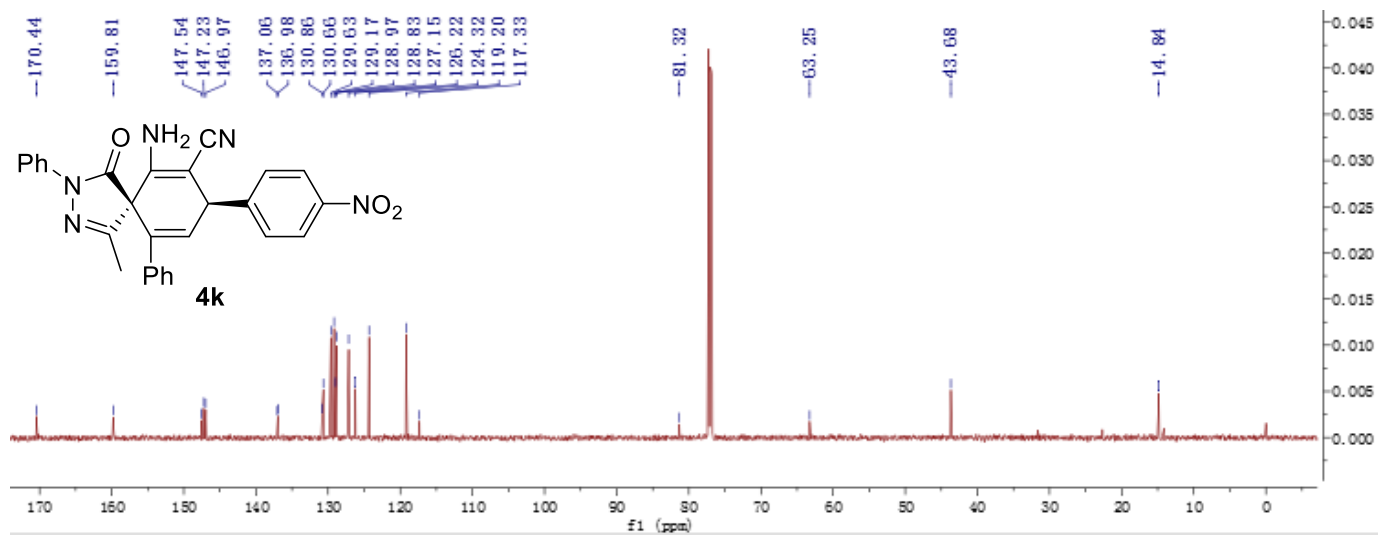
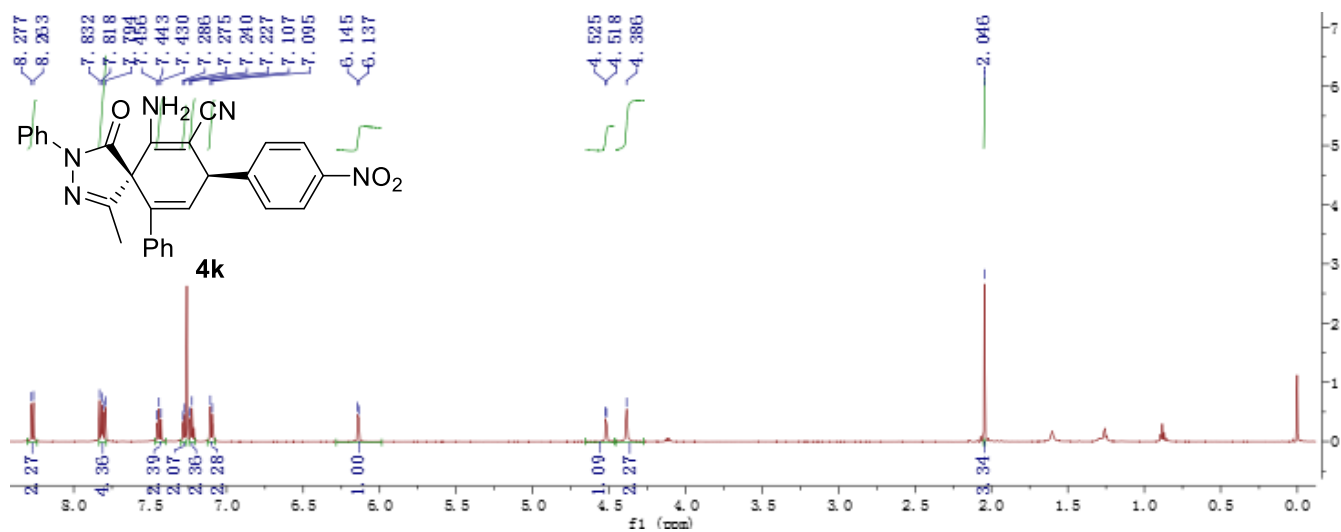
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	15.207	104267	1975	48.000	48.000
2	19.456	112957	1412	52.000	52.000
Total		217224	3388		100.000

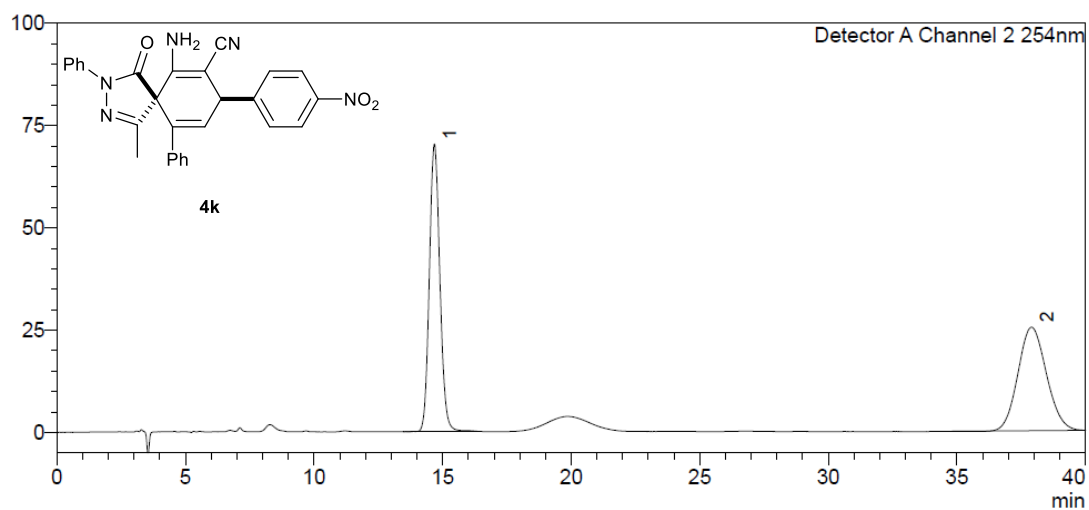


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	15.177	1494597	28325	91.850	91.850
2	19.560	132619	1858	8.150	8.150
Total		1627216	30183		100.000

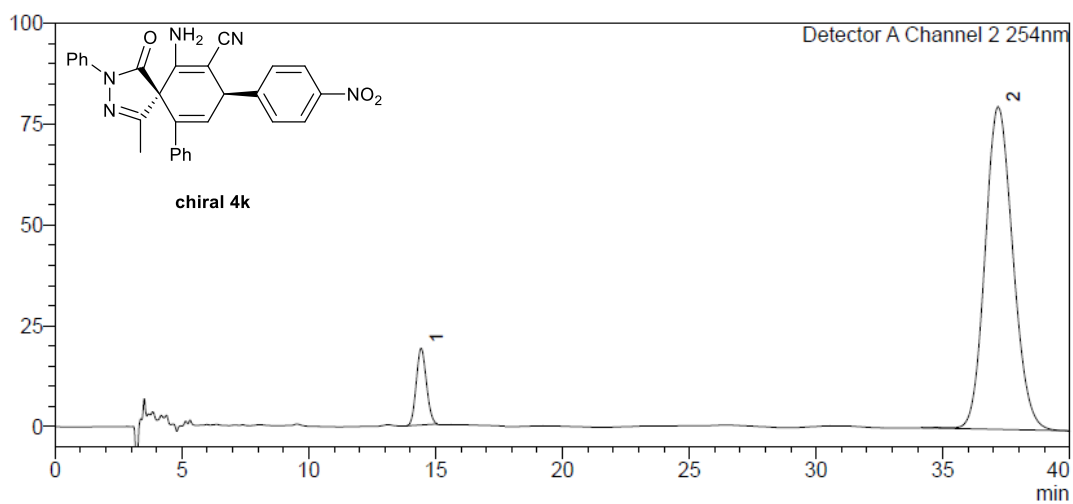




<Peak Table>

Detector A Channel 2 254nm

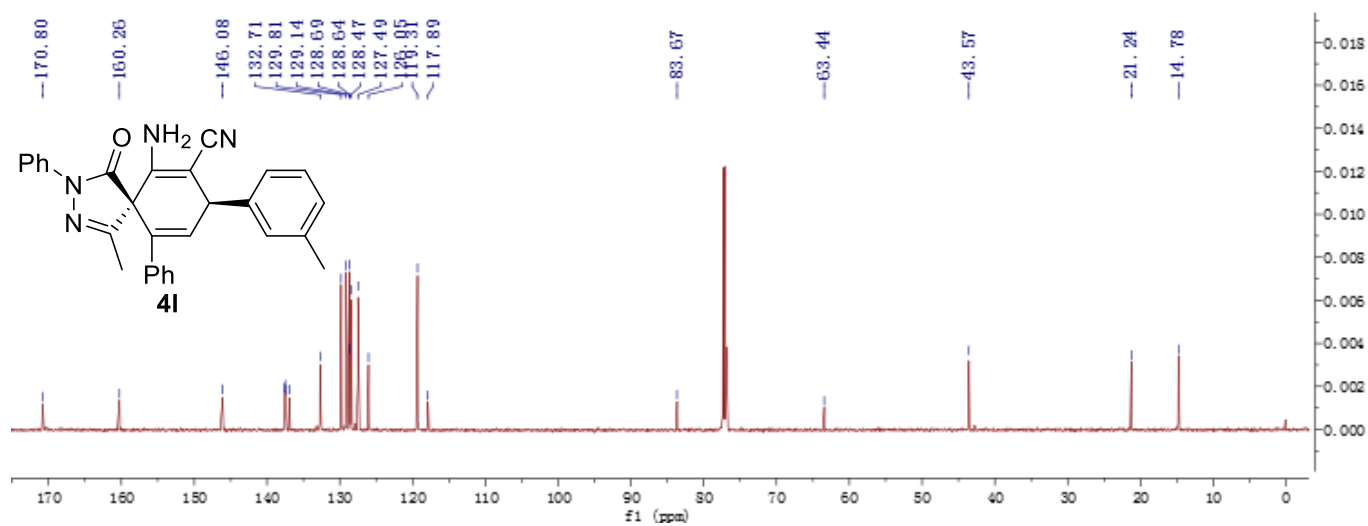
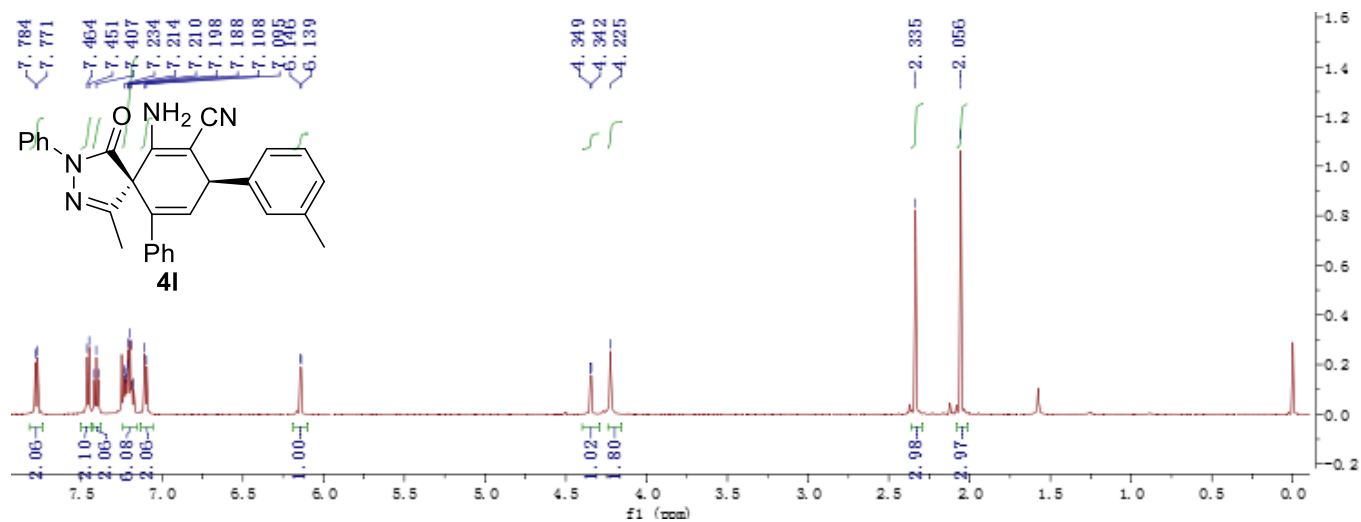
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	14.678	1998294	70273	50.711	50.711
2	37.902	1942291	25325	49.289	49.289
Total		3940585	95598		100.000

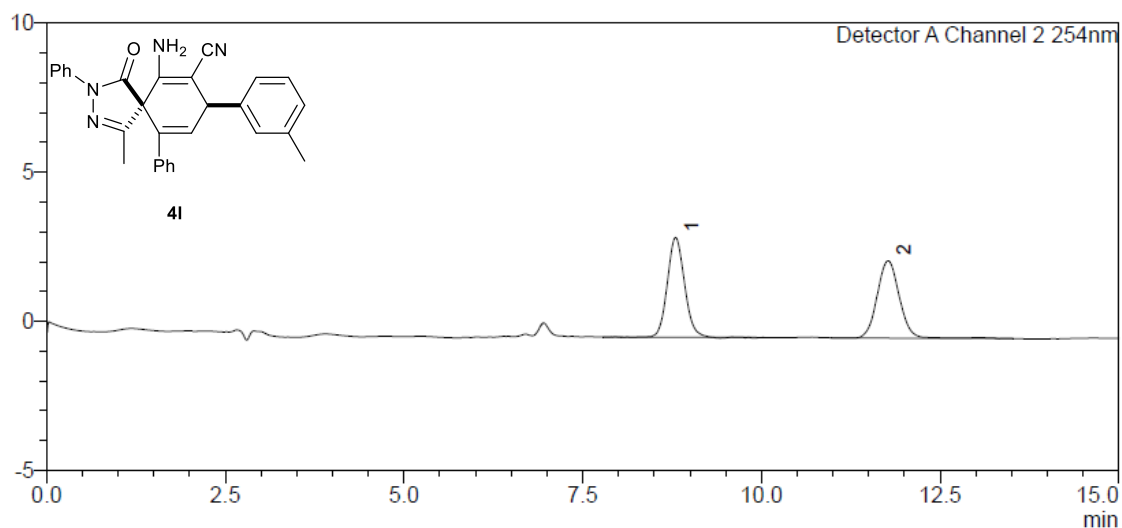


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	14.427	529302	19040	7.977	7.977
2	37.177	6106047	80125	92.023	92.023
Total		6635348	99165		100.000

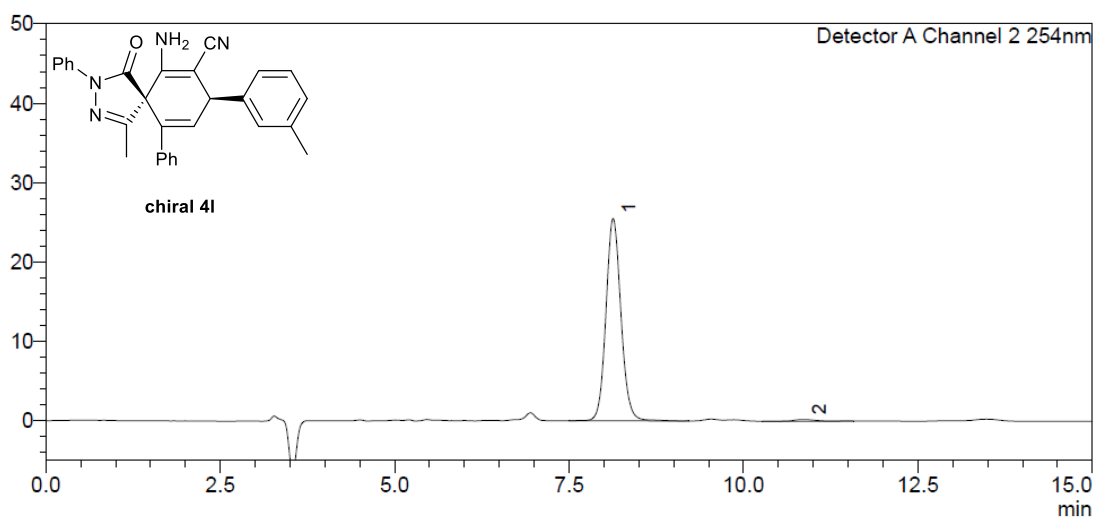




<Peak Table>

Detector A Channel 2 254nm

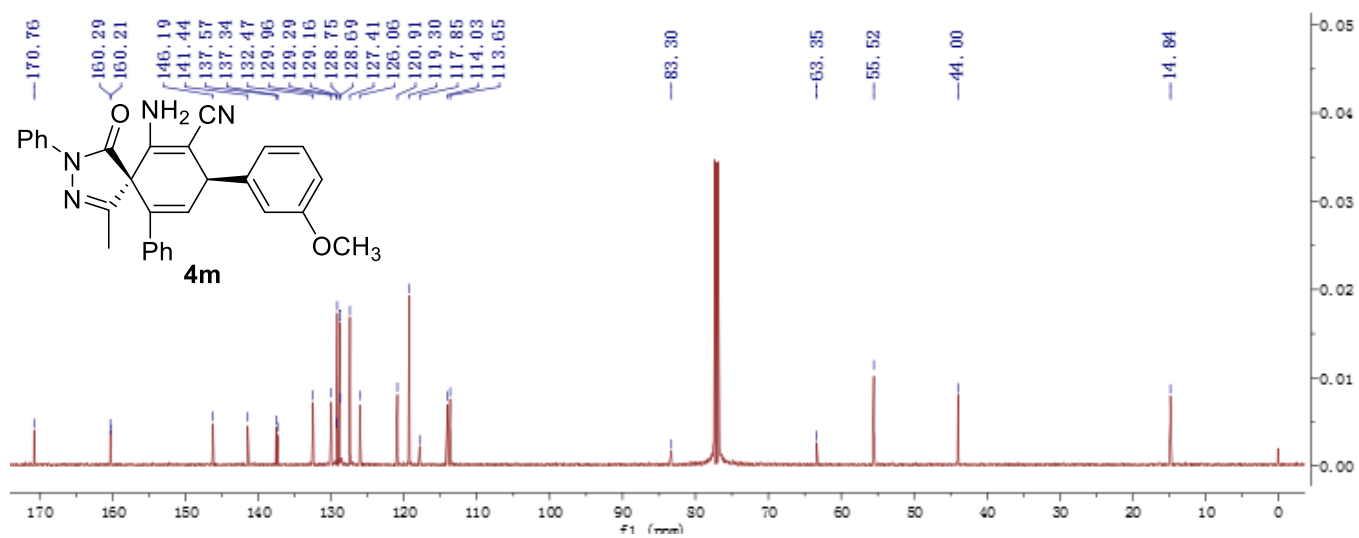
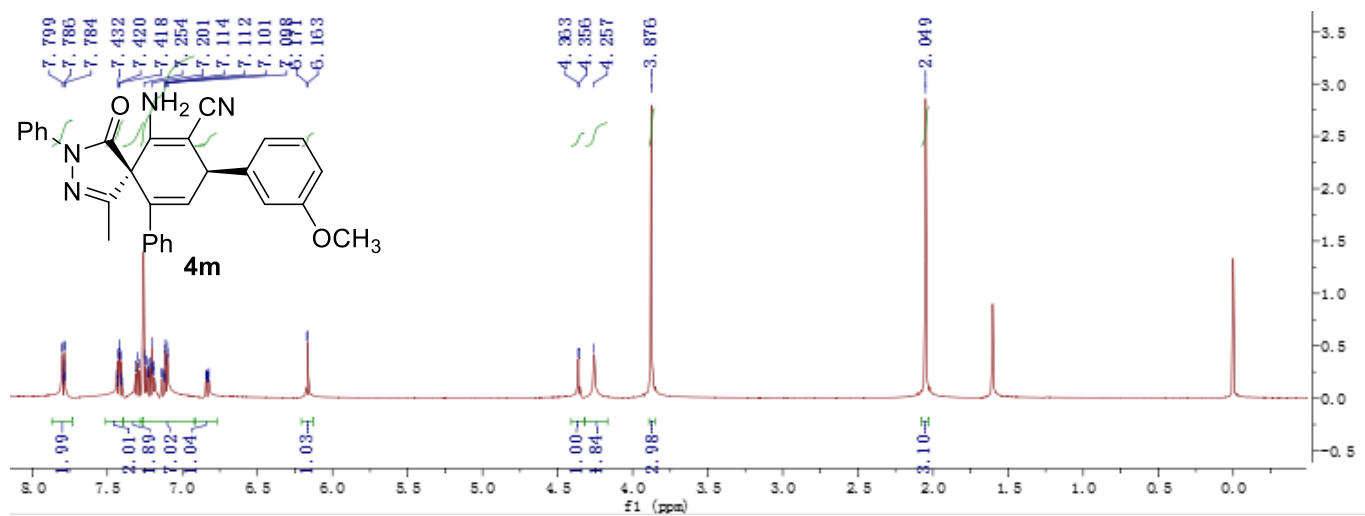
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	8.799	55494	3346	49.583	49.583
2	11.771	56427	2594	50.417	50.417
Total		111922	5941		100.000

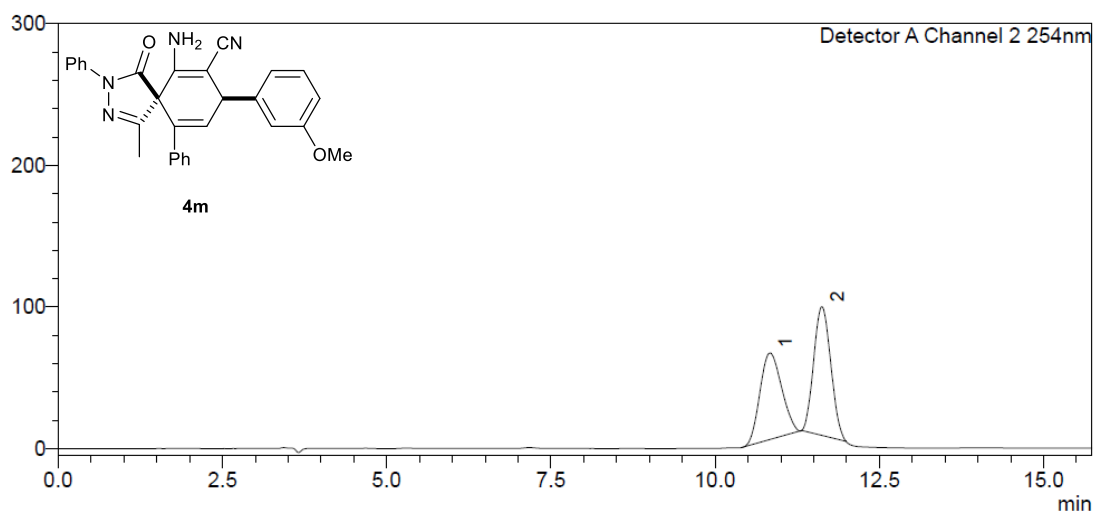


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	8.131	381785	25503	98.827	98.827
2	10.865	4532	229	1.173	1.173
Total		386318	25732		100.000

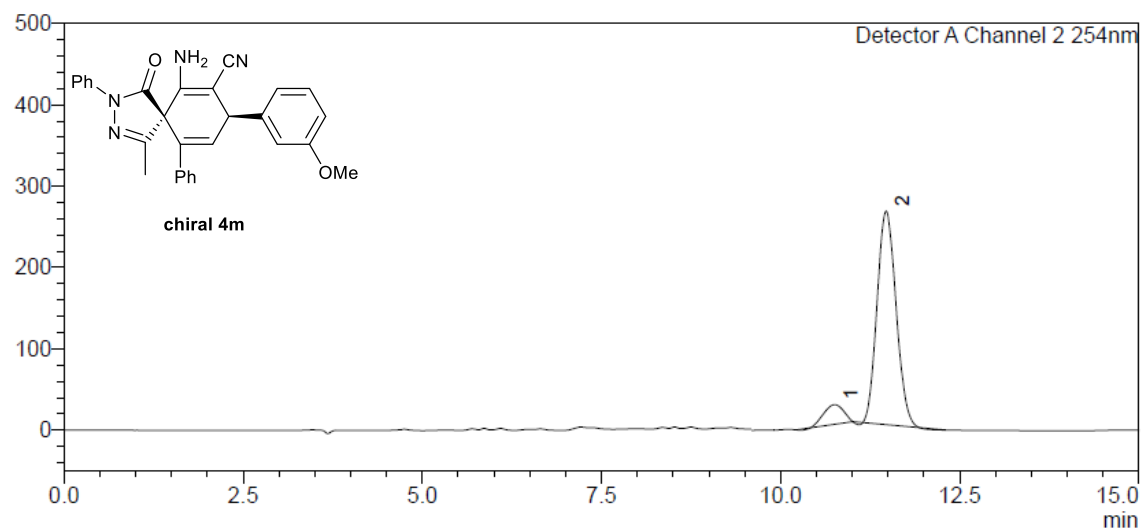




<Peak Table>

Detector A Channel 2 254nm

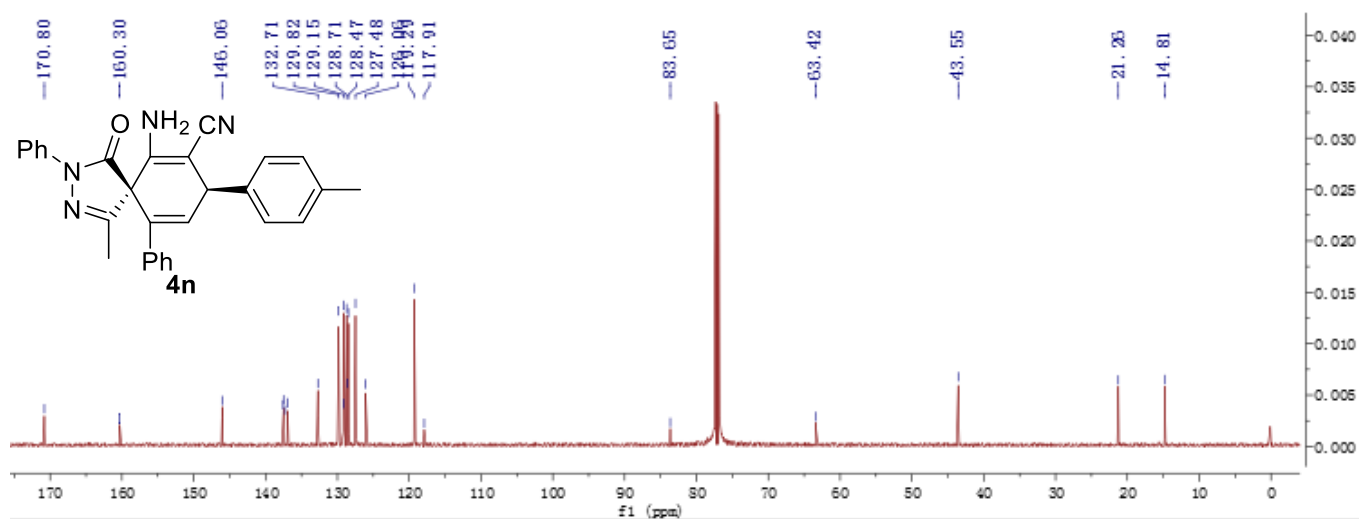
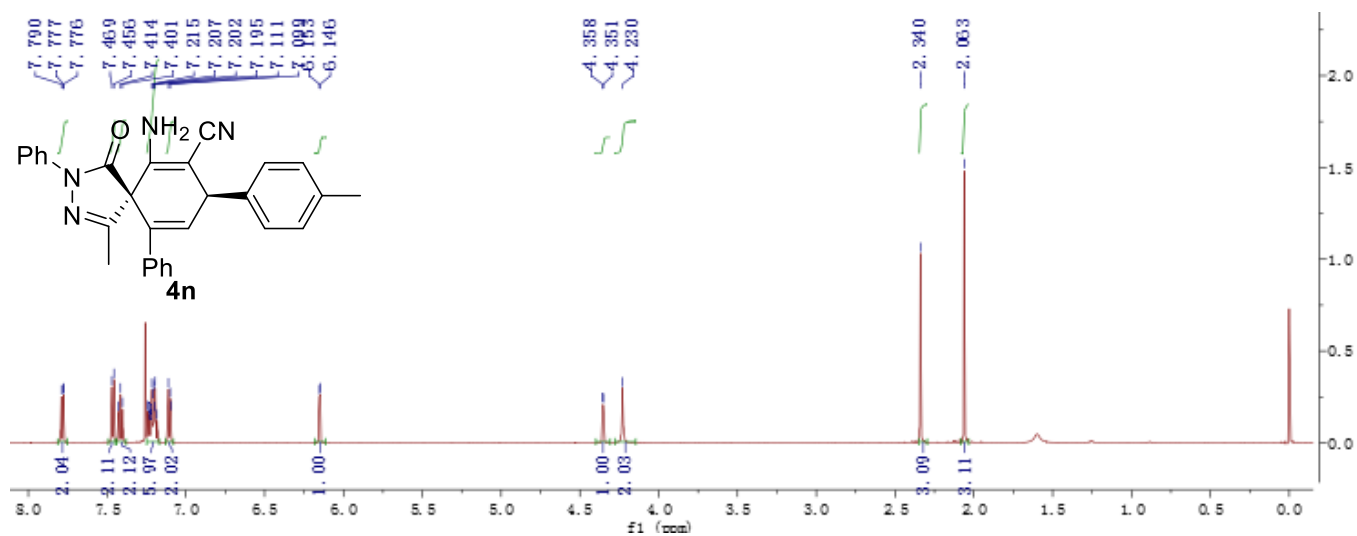
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	10.835	1401725	61208	46.031	46.031
2	11.624	1643443	90927	53.969	53.969
Total		3045168	152135		100.000

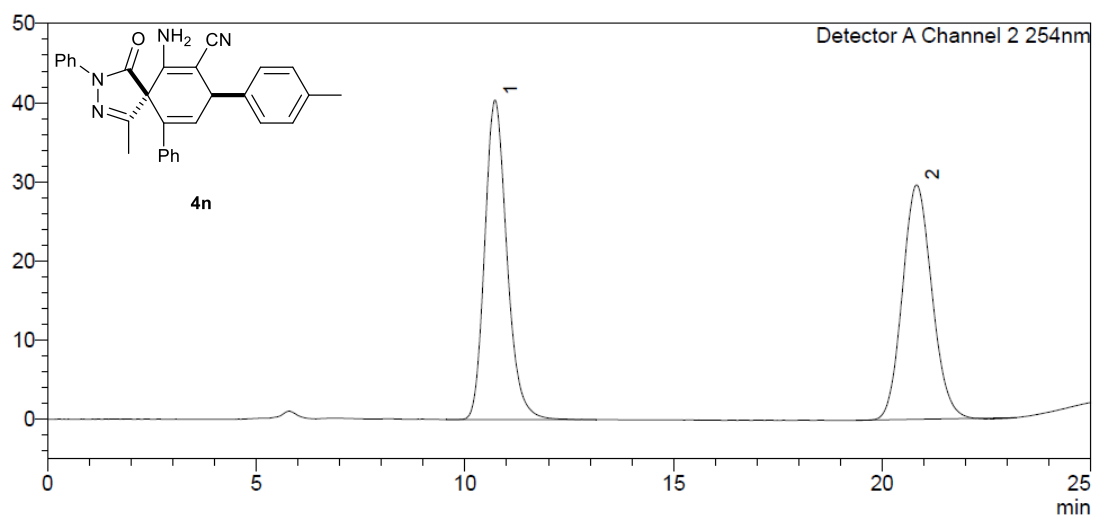


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	10.757	464041	24128	8.778	8.778
2	11.474	4822387	262614	91.222	91.222
Total		5286428	286742		100.000

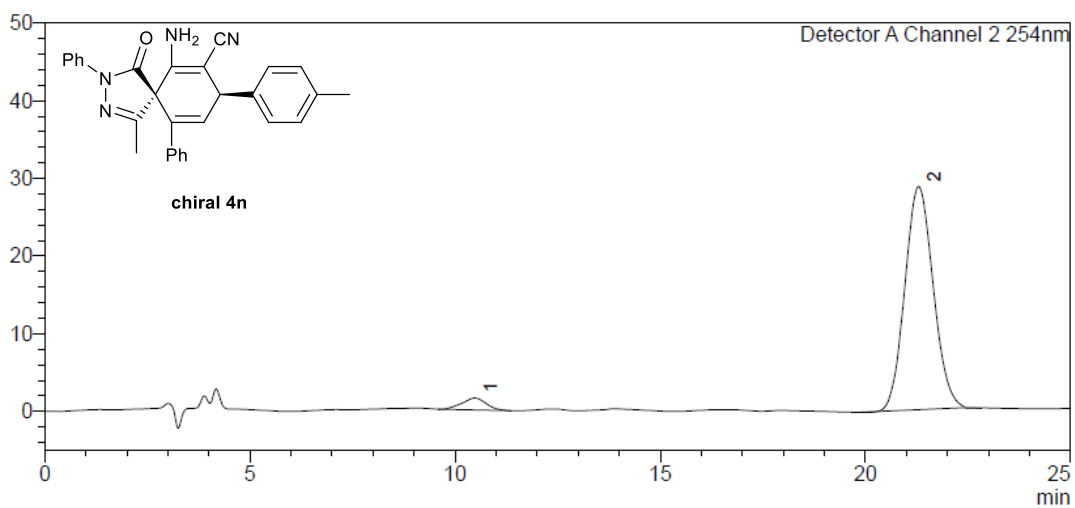




<Peak Table>

Detector A Channel 2 254nm

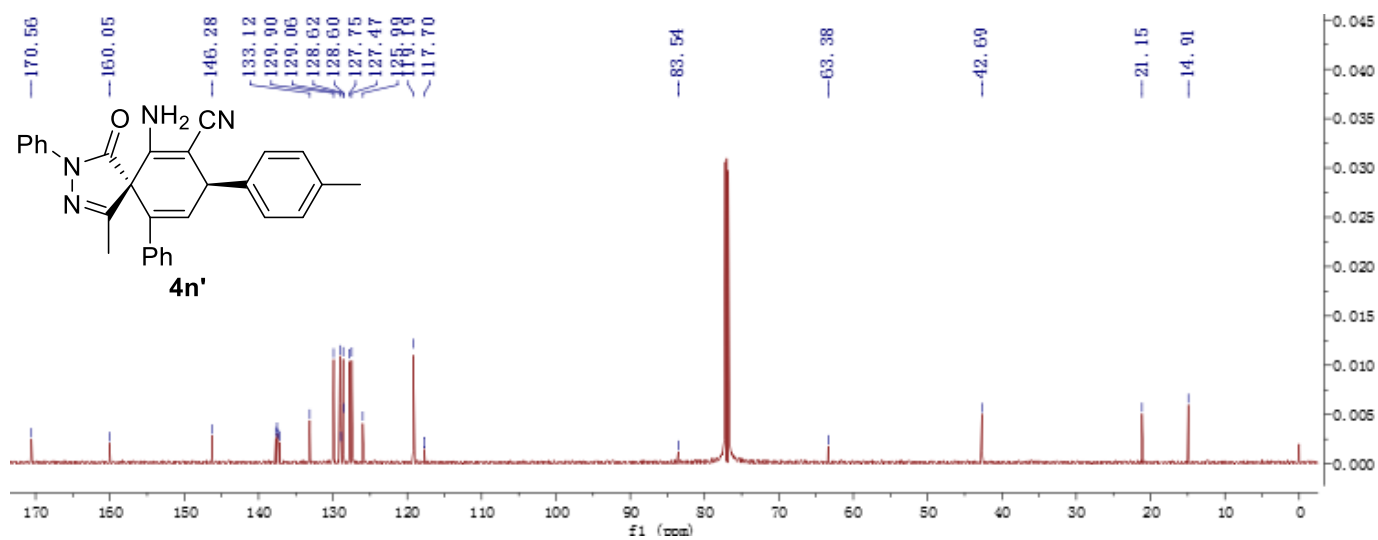
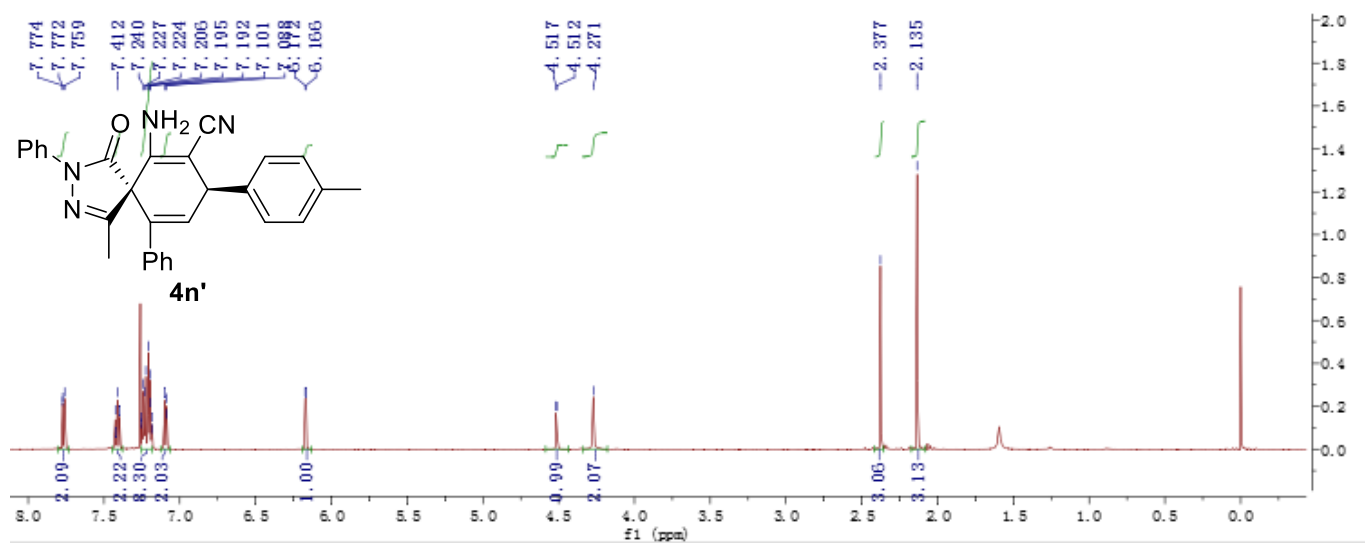
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	10.720	1493282	40392	50.716	50.716
2	20.825	1451146	29625	49.284	49.284
Total		2944427	70016		100.000

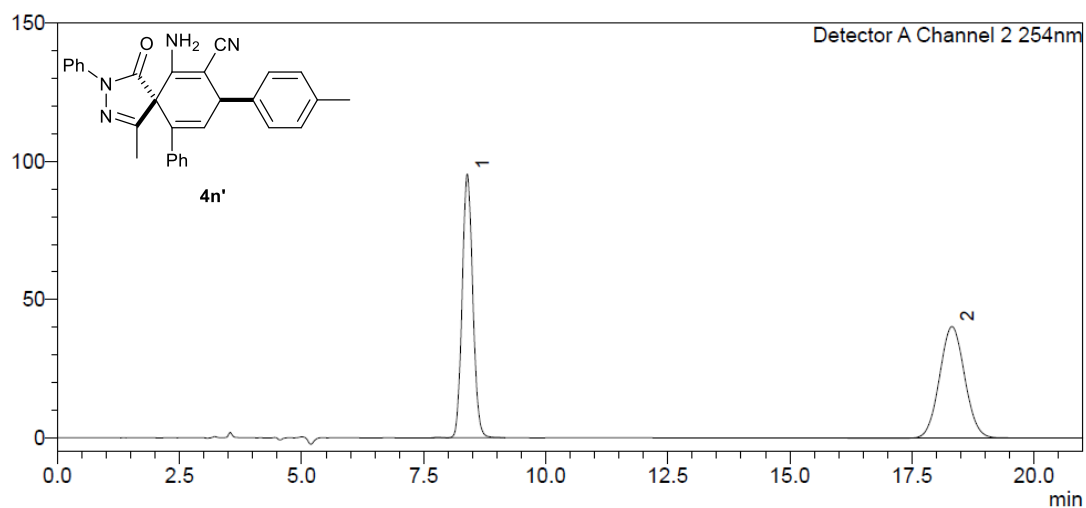


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	10.488	63806	1548	4.418	4.418
2	21.298	1380375	28739	95.582	95.582
Total		1444181	30287		100.000

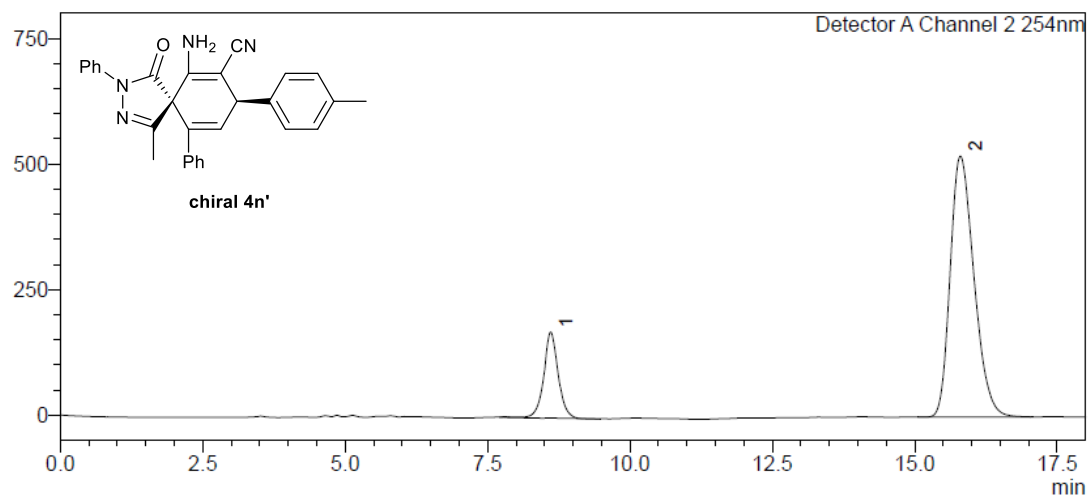




<Peak Table>

Detector A Channel 2 254nm

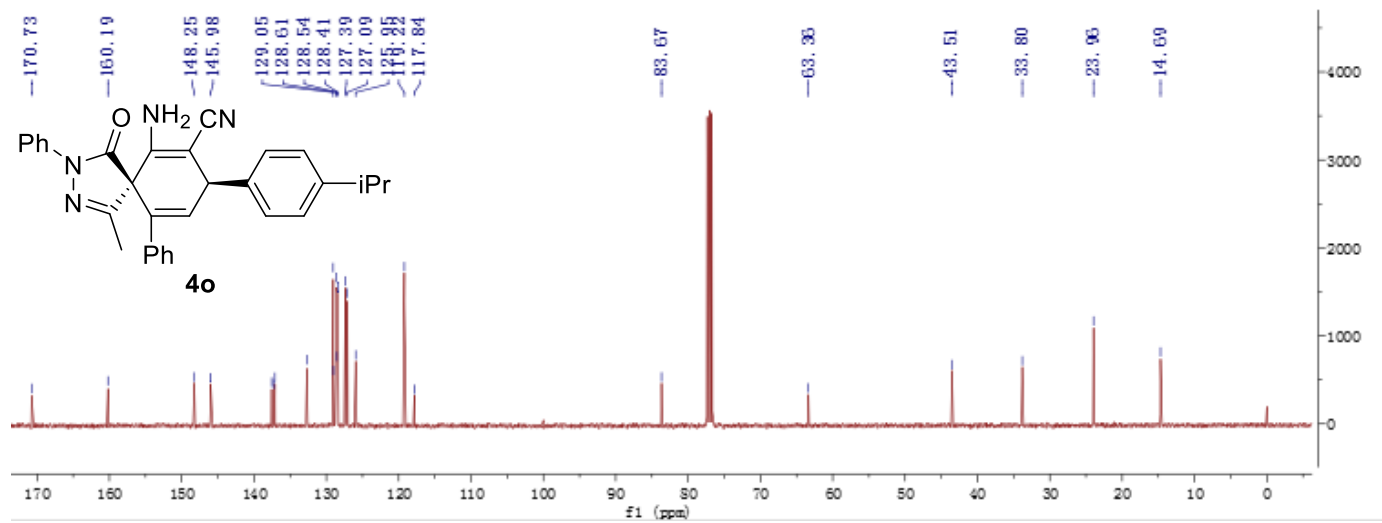
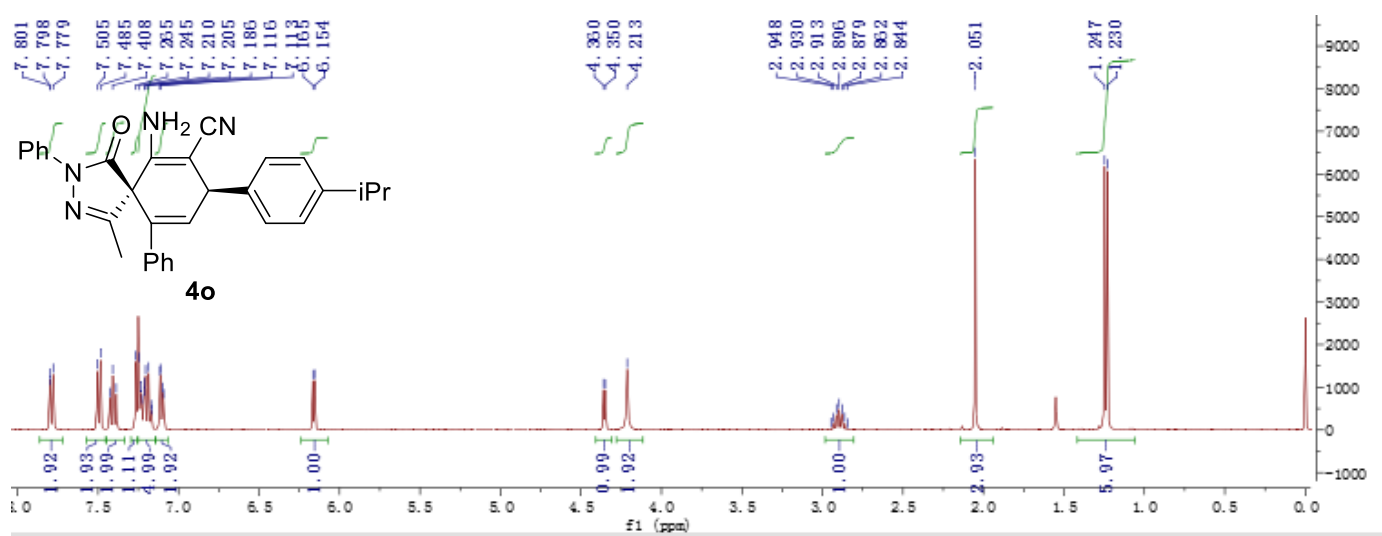
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	8.395	1443150	95441	50.081	50.081
2	18.321	1438489	40348	49.919	49.919
Total		2881639	135789		100.000

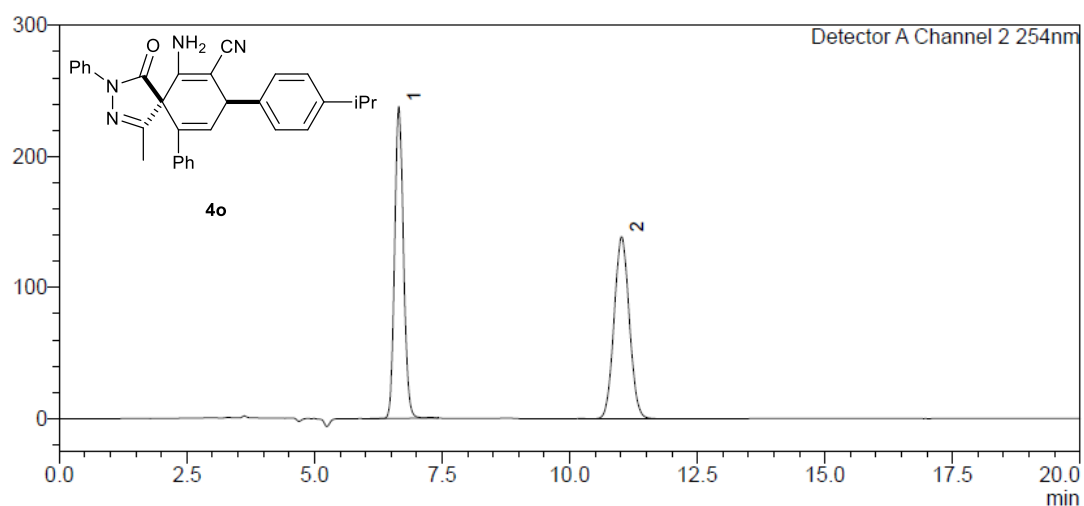


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	8.608	2258809	155008	13.024	13.024
2	15.802	15084074	519382	86.976	86.976
Total		17342883	674391		100.000

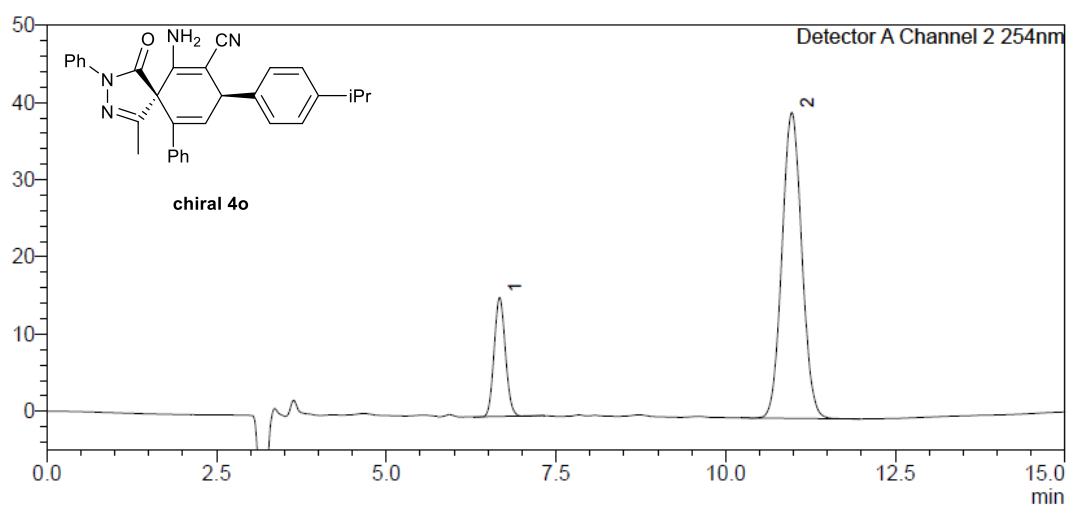




<Peak Table>

Detector A Channel 2 254nm

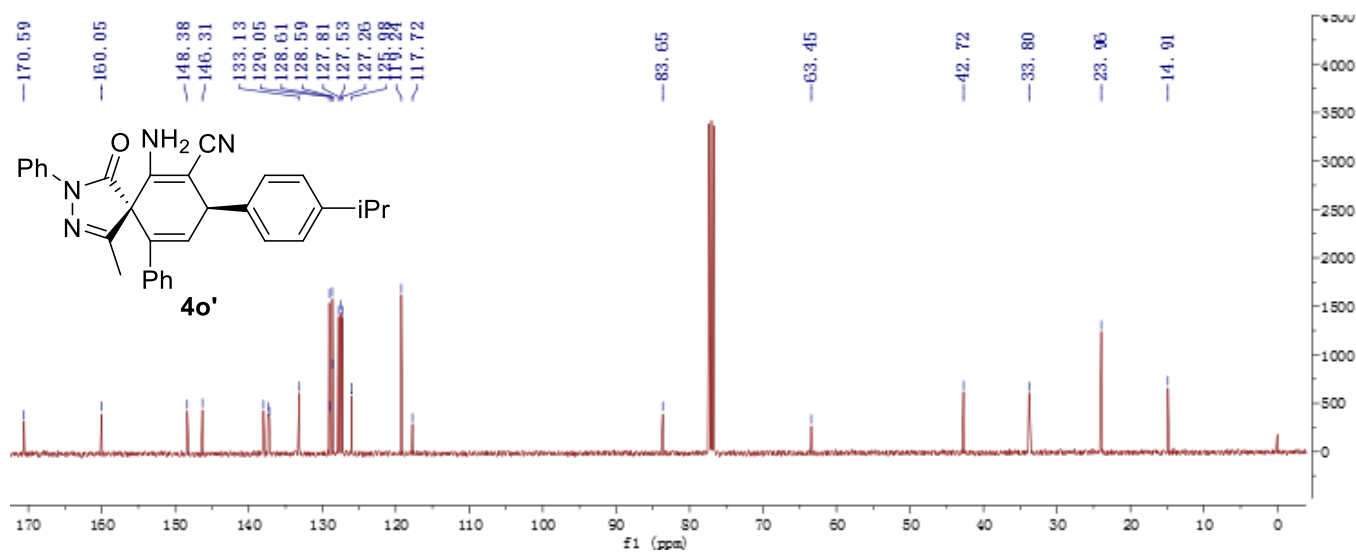
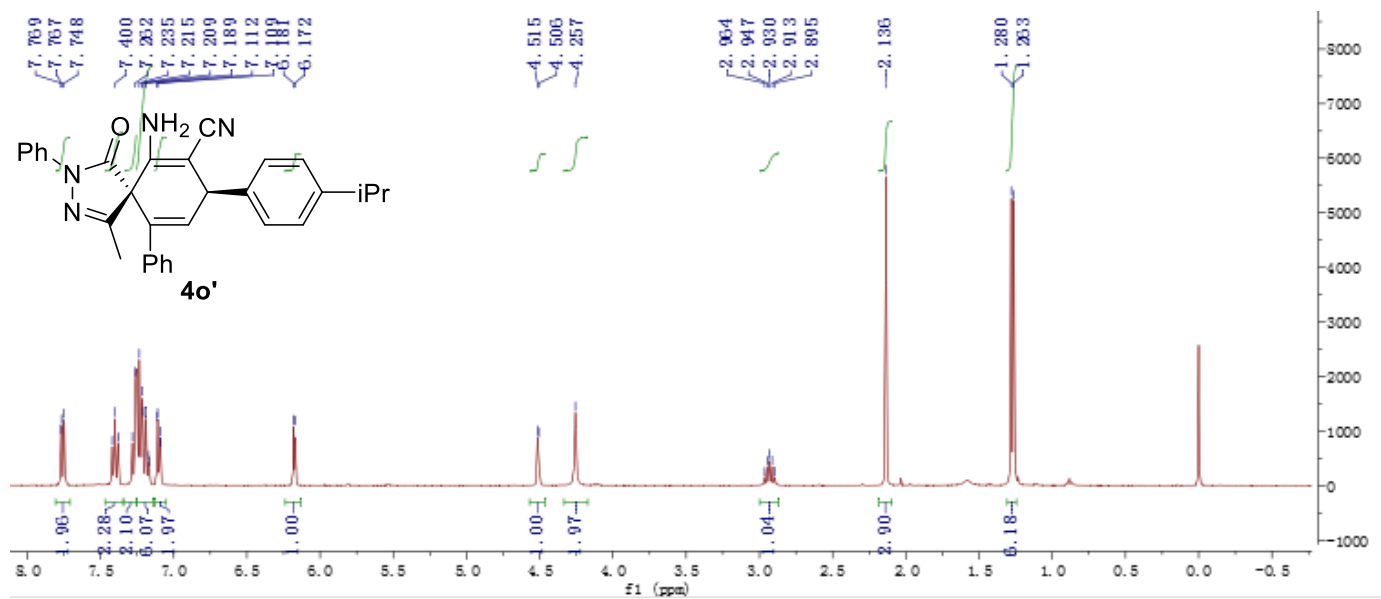
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	6.652	2887182	238036	49.999	49.999
2	11.017	2887326	138992	50.001	50.001
Total		5774508	377027		100.000

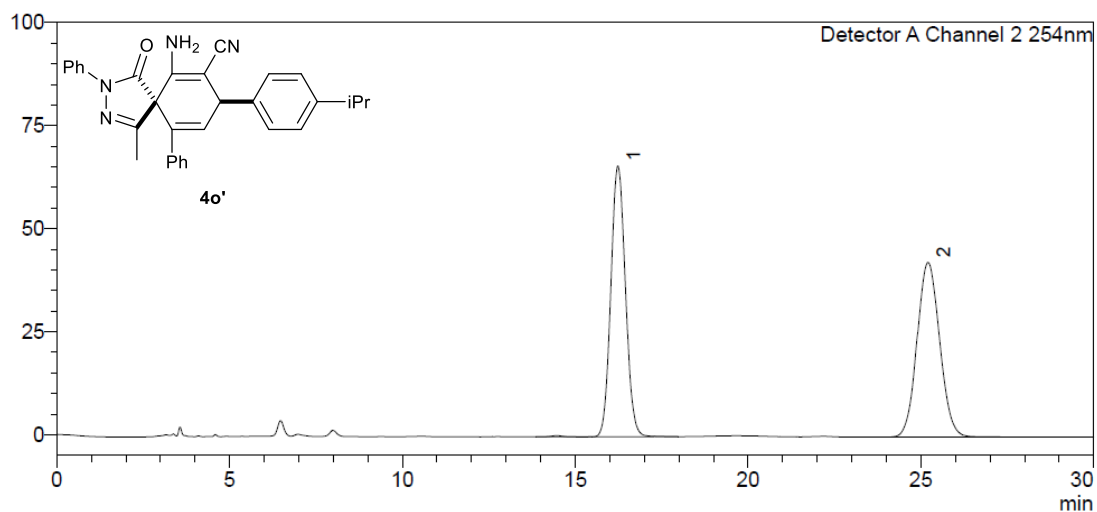


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	6.670	177881	15423	18.107	18.107
2	10.975	804491	39630	81.893	81.893
Total		982371	55053		100.000

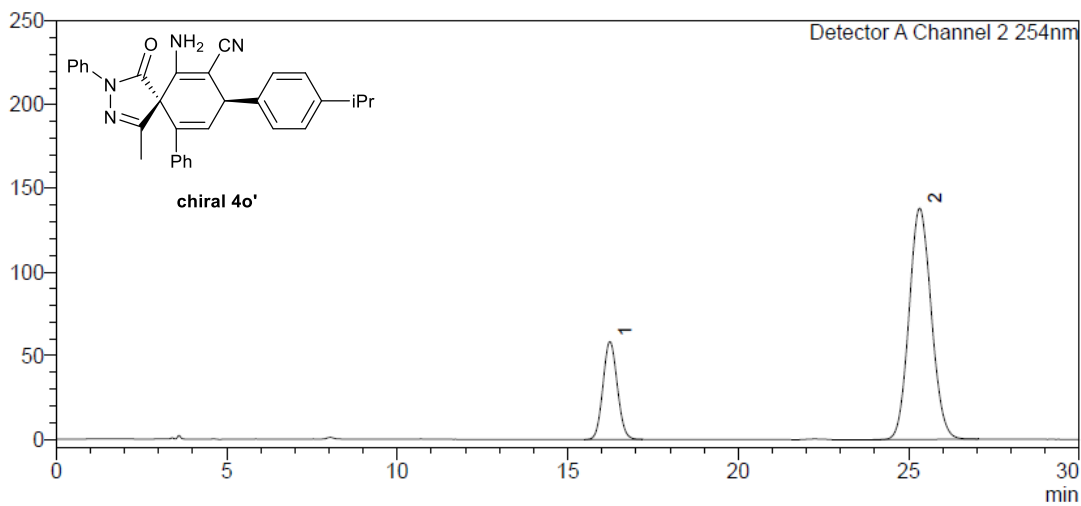




<Peak Table>

Detector A Channel 2 254nm

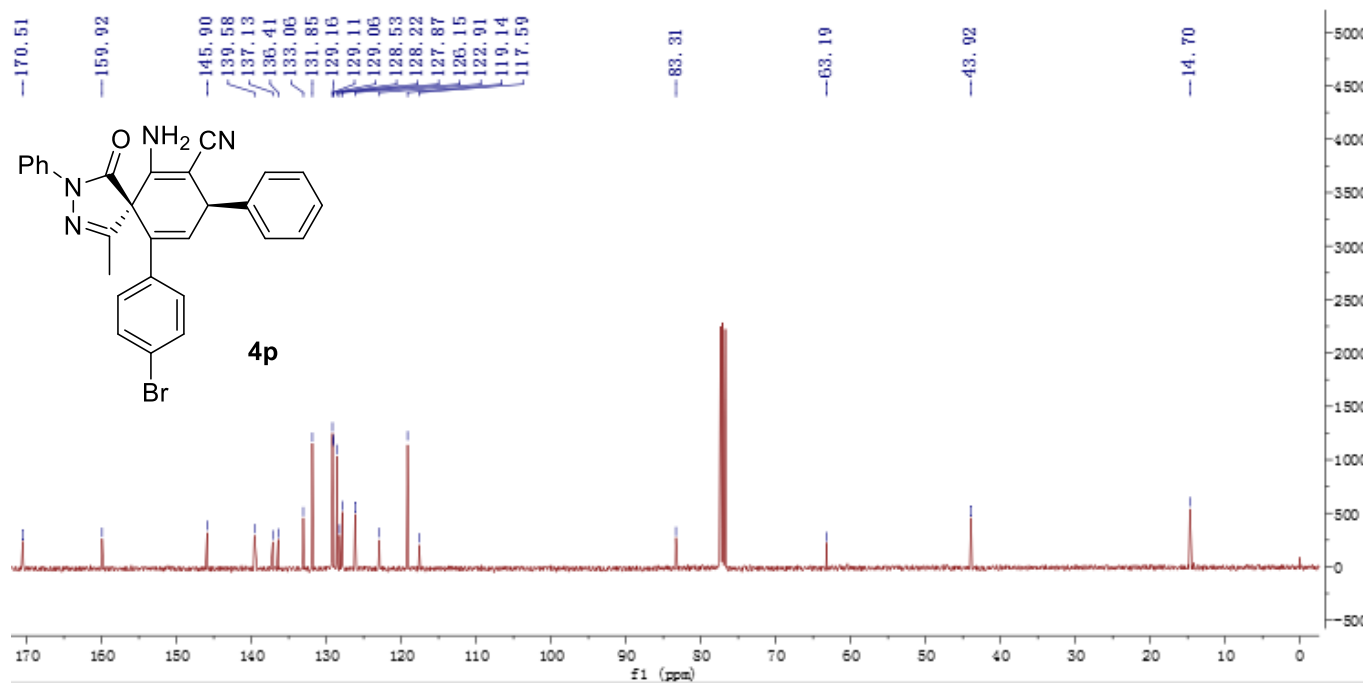
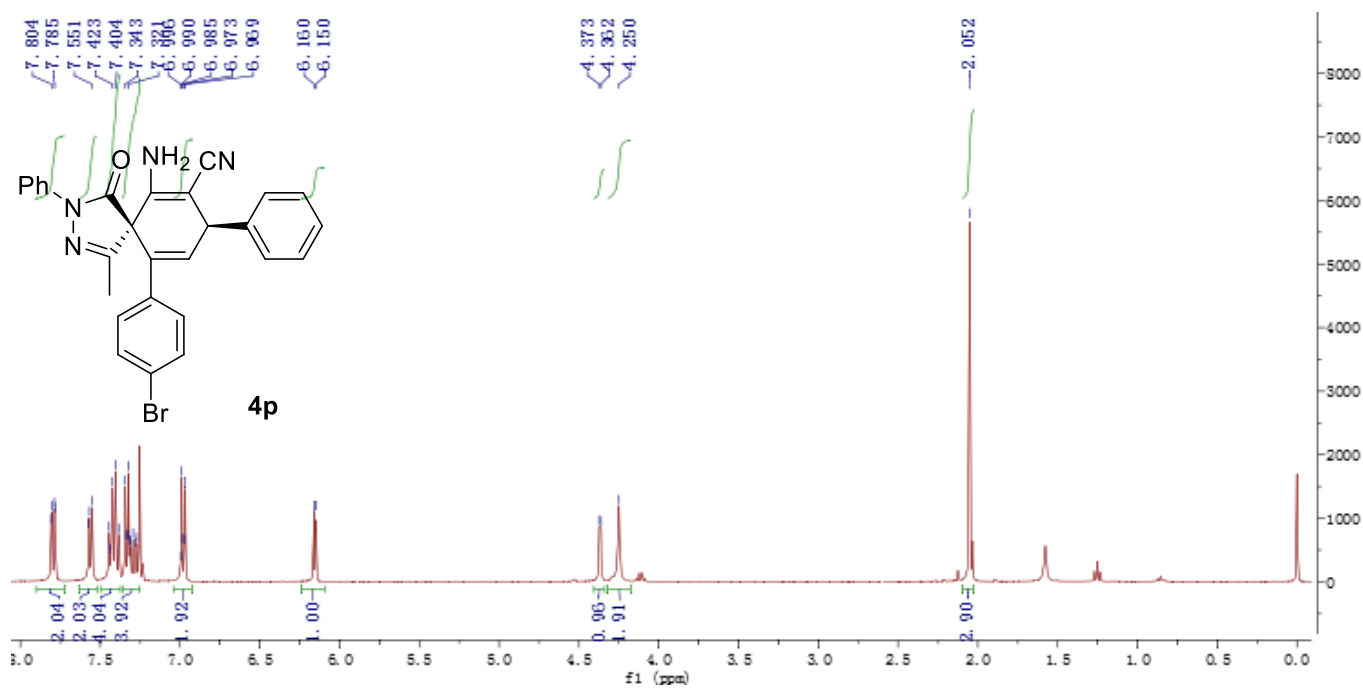
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	16.234	1933705	65760	50.084	50.084
2	25.208	1927193	42386	49.916	49.916
Total		3860898	108146		100.000

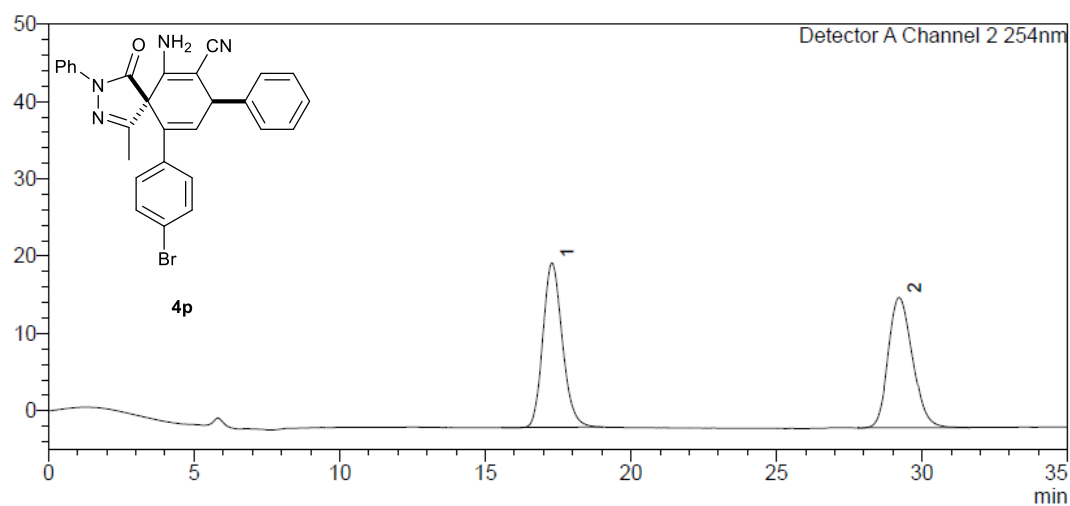


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	16.237	1712754	58496	21.454	21.454
2	25.324	6270755	138247	78.546	78.546
Total		7983509	196743		100.000

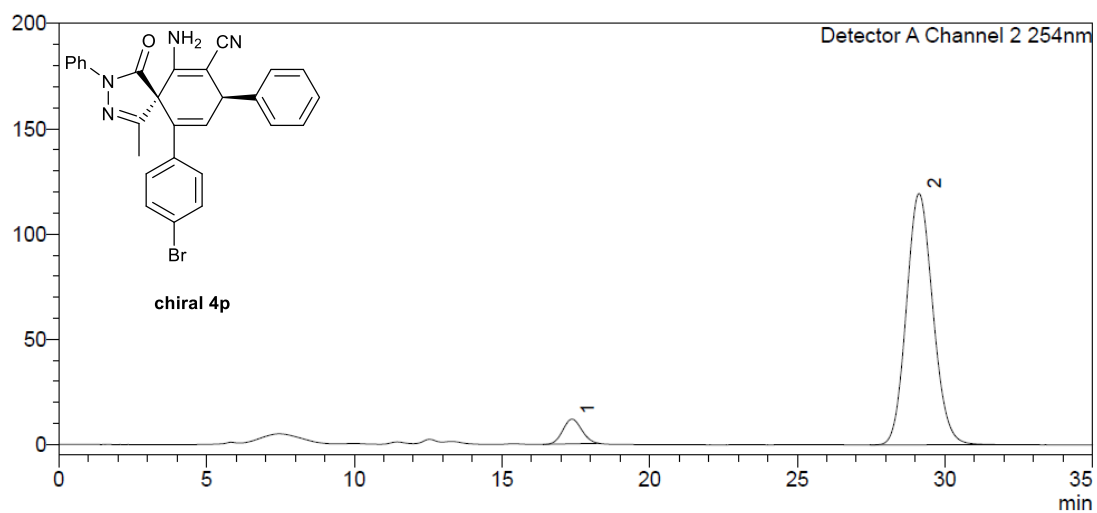




<Peak Table>

Detector A Channel 2 254nm

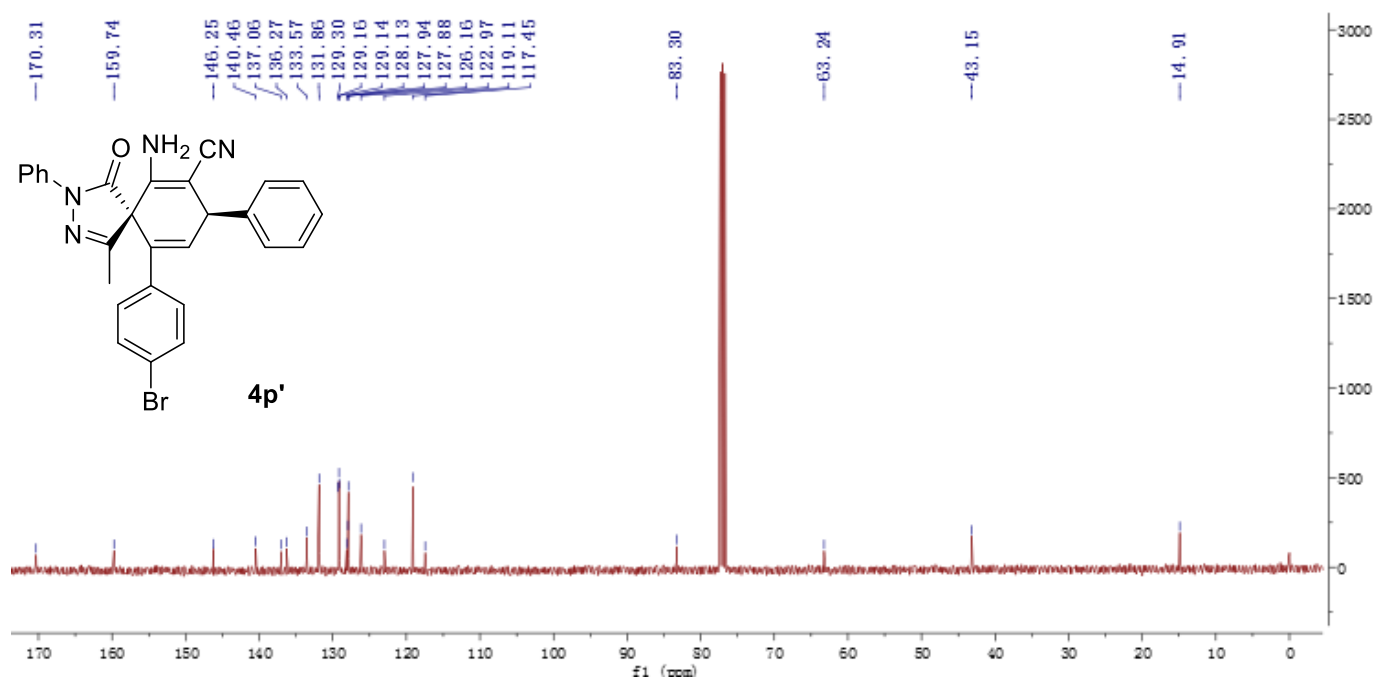
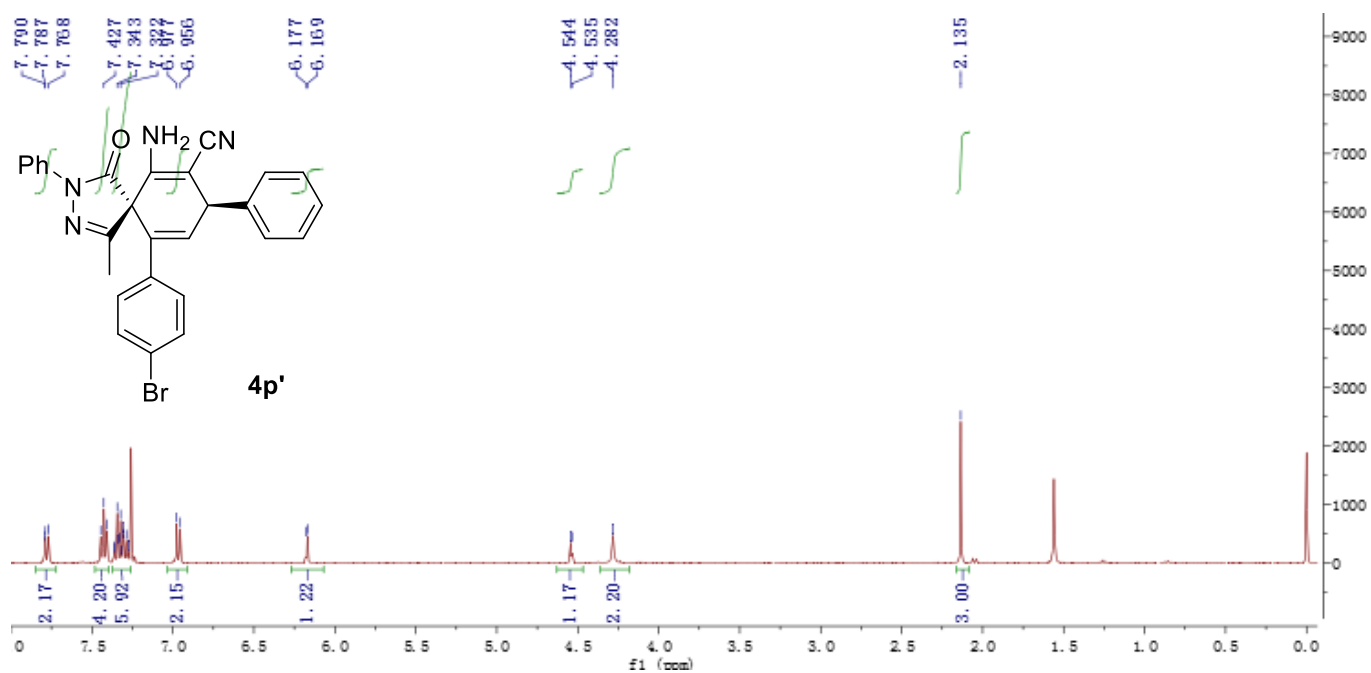
Peak#	Ret. Time	Area	Height	Conc.	Area%
1	17.289	985075	21275	49.019	49.019
2	29.212	1024486	17959	50.981	50.981
Total		2009561	39234		100.000

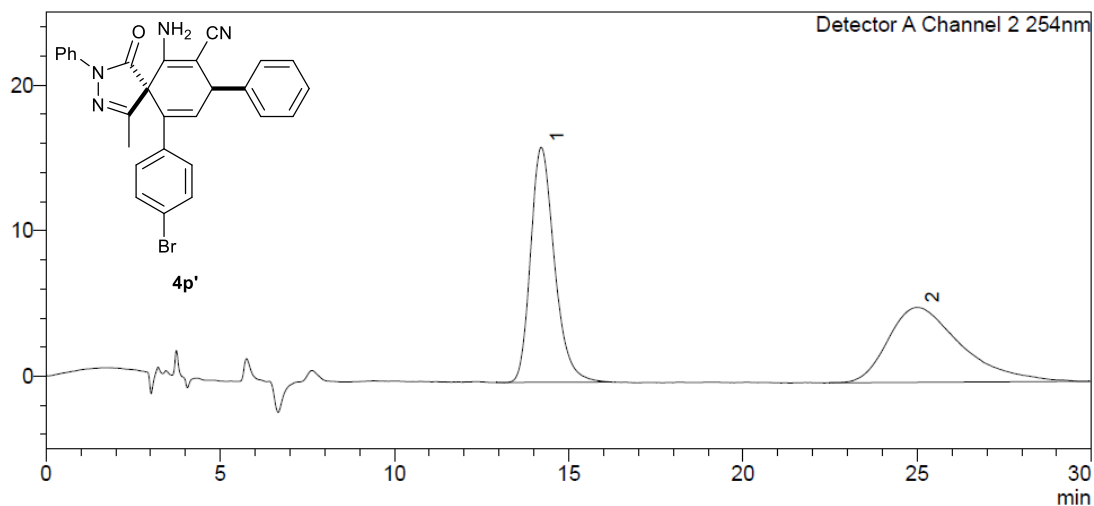


<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	17.370	521126	11726	6.547	6.547
2	29.123	7439252	119434	93.453	93.453
Total		7960379	131161		100.000

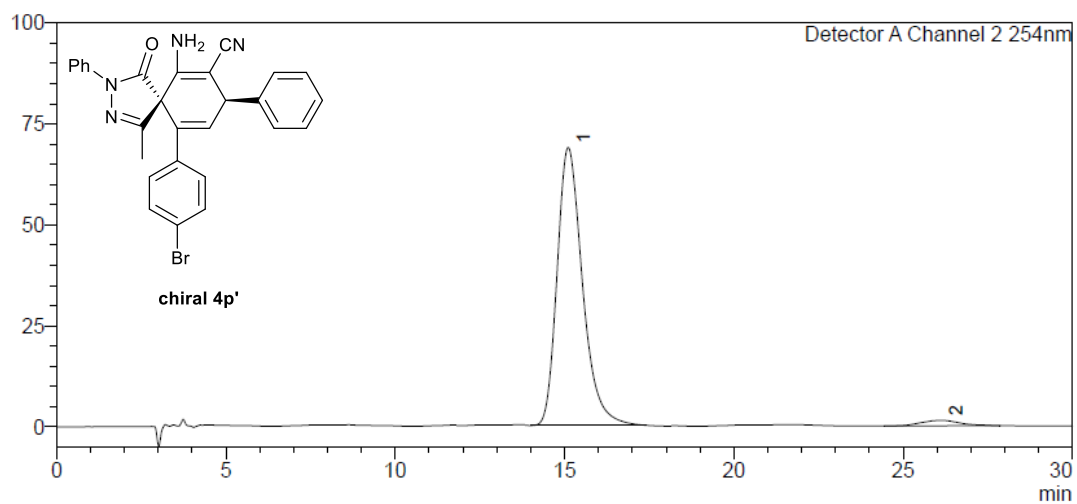




<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	14.210	769482	16158	50.687	50.687
2	24.983	748615	5150	49.313	49.313
Total		1518098	21308		100.000



<Peak Table>

Detector A Channel 2 254nm

Peak#	Ret. Time	Area	Height	Conc.	Area%
1	15.108	3588733	68826	97.148	97.148
2	26.110	105360	1357	2.852	2.852
Total		3694092	70183		100.000