

Electronic Supplementary Information

Iminonaphthofuranone Synthesis via Multiple- Component Cyclization of 2-Naphthols Using Only Molecular Oxygen

Jianyu Dong,^{‡a} Shaofeng Wu,^{‡*,a,b} Long Liu,^c Dan Zhou,^b Min Mo,^b Yadong Gao,^b Lebin Su,^b
Shuang-Feng Yin,^b and Yongbo Zhou^{*,b}

^aSchool of Physics and Chemistry, Hunan First Normal University, Changsha 410205, China.

^bAdvanced Catalytic Engineering Research Center of the Ministry of Education, College of Chemistry and Chemical Engineering, Hunan University, Changsha 410082, China.

^cKey Laboratory of Ministry of Education for Advanced Materials in Tropical Island Resources, School of Chemical Engineering and Technology, Hainan University, Haikou 570228, China.

E-mail: zhouyb@hnu.edu.cn; shallfar@hnu.edu.cn.

Table of contents

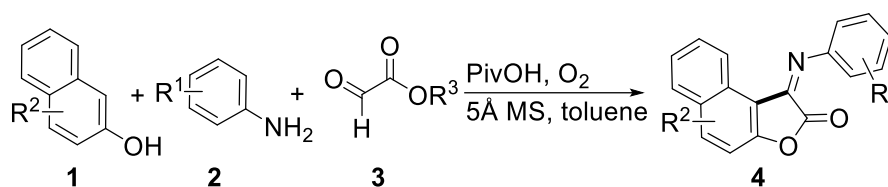
1. General Information	S2
2. Experimental Procedure	S2
3. X-ray Crystallographic Data of the Product (4m)	S3
4. The UV Absorption Detection and Solution Photographs of Products	S5
5. Investigations of the Reaction Mechanism	S12
6. References	S23
7. Characterization Data for the Products	S23
8. Copies of ¹H and ¹³C NMR Spectra of the Products	S37

1. General Information

The reactions were carried out in Schlenk tubes of 25 mL under N₂ atmosphere. The heat source is IKA magnetic stirrer with RCT Basic. Reagents were used as received unless otherwise noted, and solvents were purified according to standard operation procedure. Column chromatography was performed using Silica Gel 60 (300–400 mesh). The reactions were monitored by GC and GC-MS, GC-MS results were recorded on GC-MS QP2010, and GC analysis was performed on GC 2010 plus. The ¹H and ¹³C NMR spectra were recorded on a Bruker ADVANCE III spectrometer at 400 MHz and 101 MHz, respectively, and chemical shifts were reported in parts per million (ppm). The HRMS measurements were recorded on MAT95XP high resolution mass spectrometer by the electron ionization (EI) method, and the mass analyzer type is TOF for EI. EPR spectra were recorded on JES-FA 200 electron spin resonance spectrometer. The single-crystal X-ray diffraction was conducted on the D8 Quest X-ray single crystal diffractometer. All solvents and reagents were purchased from Energy Chemical, Bide Pharmatech Ltd., Alfa Aesar, and Aladdin.

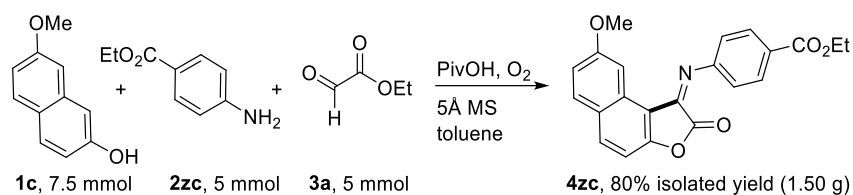
2. Experimental Procedure

2.1 General Experimental Procedure for the Synthesis of Iminonaphthofuranones



In an oven dried 25 mL Schlenk tube was charged with 2-naphthols **1** (0.3 mmol), anilines **2** (0.2 mmol), trimethylacetic acid (0.4 mmol), and 5Å molecular sieve (200 mg), after charging oxygen for three times, the ester glyoxylate solution **3** (0.2 mmol), and toluene (4 mL) were added. The reaction mixture was reacted at 100 °C for 14 h. After completion of the reaction, the reaction mixture was filtered and the filtrate was concentrated under vacuum. The desired product was isolated by column chromatography over silica gen (300-400 mesh) using petroleum ether-ethyl acetate as eluent.

2.2 Preparation of ethyl (*Z*)-4-((8-methoxy-2-oxonaphtho[2,1-*b*]furan-1(2*H*)-ylidene)amino)benzoate (**4zc**) at 5 mmol scale



In an oven dried 250 mL Schlenk tube was charged with 7-methoxy-2-naphthol (**1c**, 7.5 mmol), ethyl *p*-aminobenzoate (**2zc**, 5 mmol), trimethylacetic acid (10 mmol), and 5 Å molecular sieve (2 g), after charging oxygen for three times, the ester glyoxylate solution **3a** (5 mmol), and toluene (100 mL) were added. The reaction mixture was reacted at 100 °C for 14 h. After completion of the reaction, the reaction solution was cooled to room temperature and left undisturbed for at least one hour, during which red solids precipitated and deposited on the walls of the container. The solids were then filtered and washed to obtain the product. The remaining solution was subjected to solvent removal, and the residue was further purified by column chromatography over silica gen (300-400 mesh) using petroleum ether-ethyl acetate as eluent.

3. X-ray Crystallographic Data of the Product (**4m**): Thermal Ellipsoid Plot Drawn with 50% Probability.

Crystal of product **4m** was obtained after slow evaporation of DCM, layered by methanol at room temperature. Crystals suited for single crystal X-Ray diffraction measurements were mounted on a glass fiber. Geometry and intensity data were collected with a Bruker D8 Quest X-ray single crystal diffractometer equipped with a PHOTON II detector using graphite monochromated Mo-K α radiation ($\lambda = 0.71073 \text{ \AA}$). The crystal was kept at 150(2) K during data collection. The structure was solved by direct methods and refined on F^2 with SHELXL-97 using Olex-2 software.

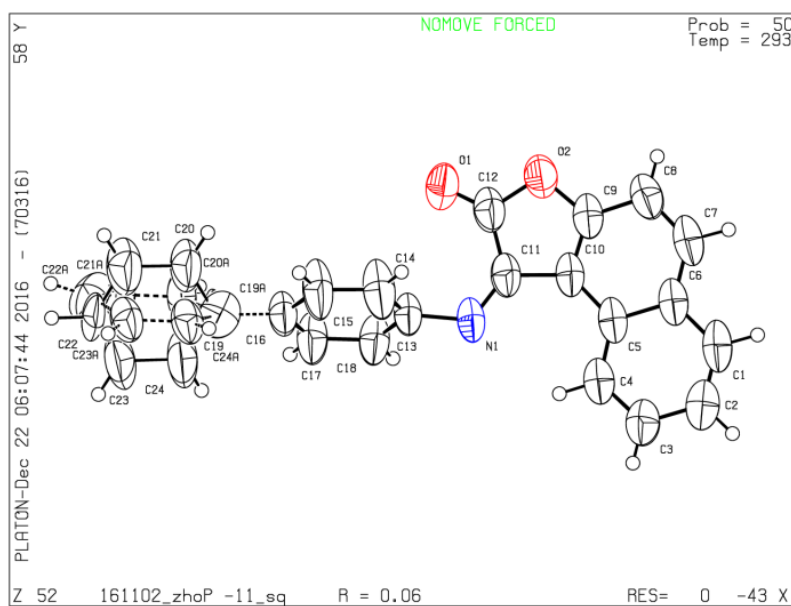


Figure S1. X-ray crystal structure of compound **4m** (CCDC number: 2182610).

Table S2. Summary of X-ray crystallographic data for compound 4m.

formula	C ₂₄ H ₁₅ NO ₂
Fw.	349.37
crystal system	triclinic
space group	P-1
<i>a</i> /Å	4.5878(8)
<i>b</i> /Å	13.936(3)
<i>c</i> /Å	16.303(3)
<i>α</i> /deg	111.653(19)
<i>β</i> /deg	95.183(16)
<i>γ</i> /deg	99.427(16)
<i>V</i> /Å ³	942.7(3)
<i>Z</i>	2
<i>D</i> /g cm ⁻³	1.231
cryst size/mm	0.4 × 0.26 × 0.23
reflns collected	5885
ind reflns, <i>R</i> _{int}	3464, 0.0259
goodness-of-fit on <i>F</i> ²	1.010
<i>R</i> ₁ , <i>wR</i> ₂ [<i>I</i> > 2σ(<i>I</i>)]	0.0646, 0.1707
<i>R</i> ₁ , <i>wR</i> ₂ (all data)	0.1323, 0.2150

4. The UV Absorption Detection and Solution Photographs of Products

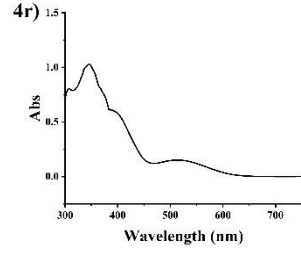
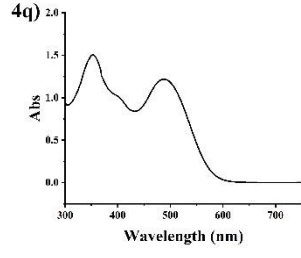
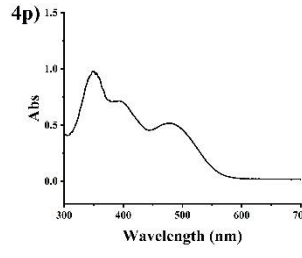
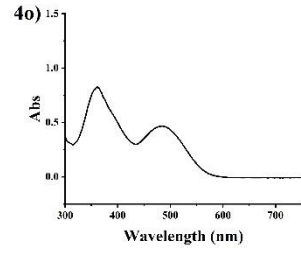
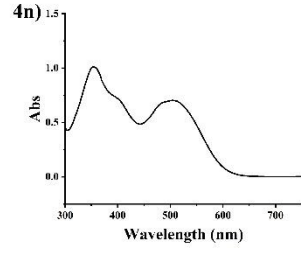
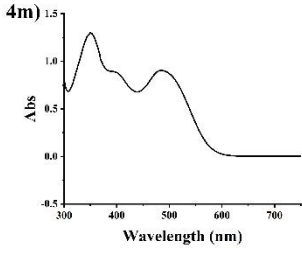
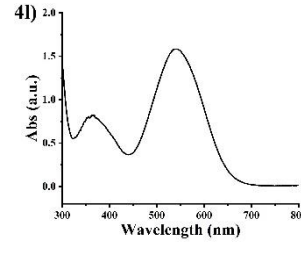
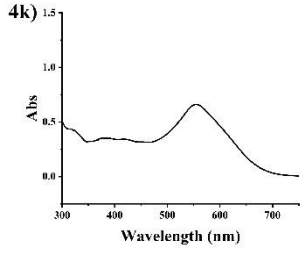
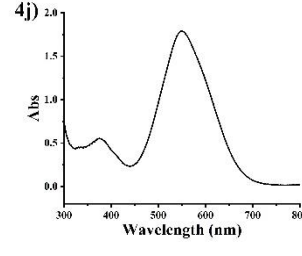
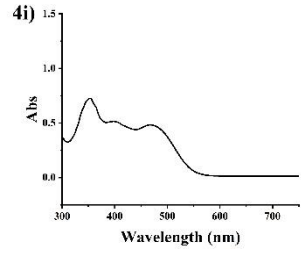
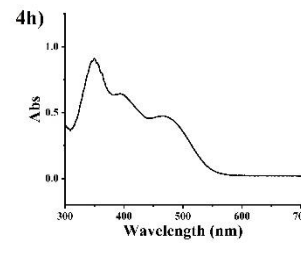
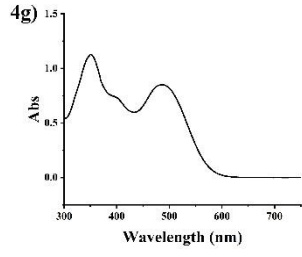
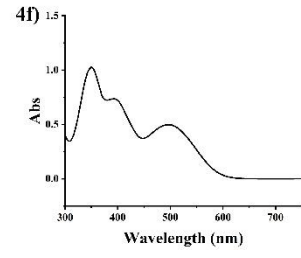
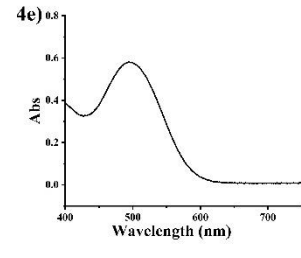
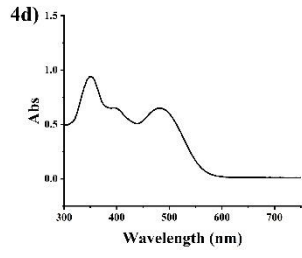
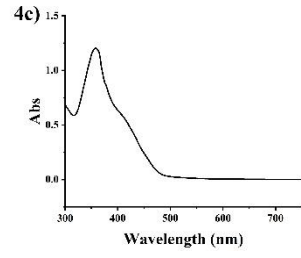
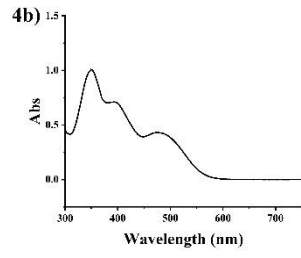
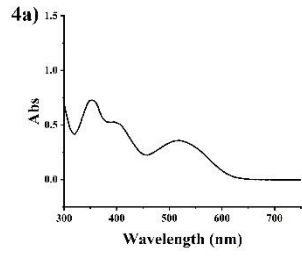
4.1 The UV Absorption Detection of Products

Table S3. The λ_{max} and ε values of compounds

Compound	4a	4b	4c	4d	4e	4f	4g	4h	4i	4j	4k	4l
λ _{max} /nm	353	349	359	348	494	349	349	349	353	549	555	540
ε (× 10 ⁵)	0.727	1.009	1.203	0.939	0.582	1.027	1.128	0.909	0.726	1.792	0.661	1.586

Compound	4m	4n	4o	4p	4q	4r	4s	4t	4u	4v	4w	4x
λ _{max} /nm	350	353	361	349	353	345	349	394	354	349	349	395
ε (× 10 ⁵)	1.298	1.013	0.826	0.987	1.504	1.032	0.982	1.212	1.014	1.144	0.689	1.054

Compound	4y	4z	4za	4zb	4zc	4zd
λ _{max} /nm	400	399	399	403	405	401



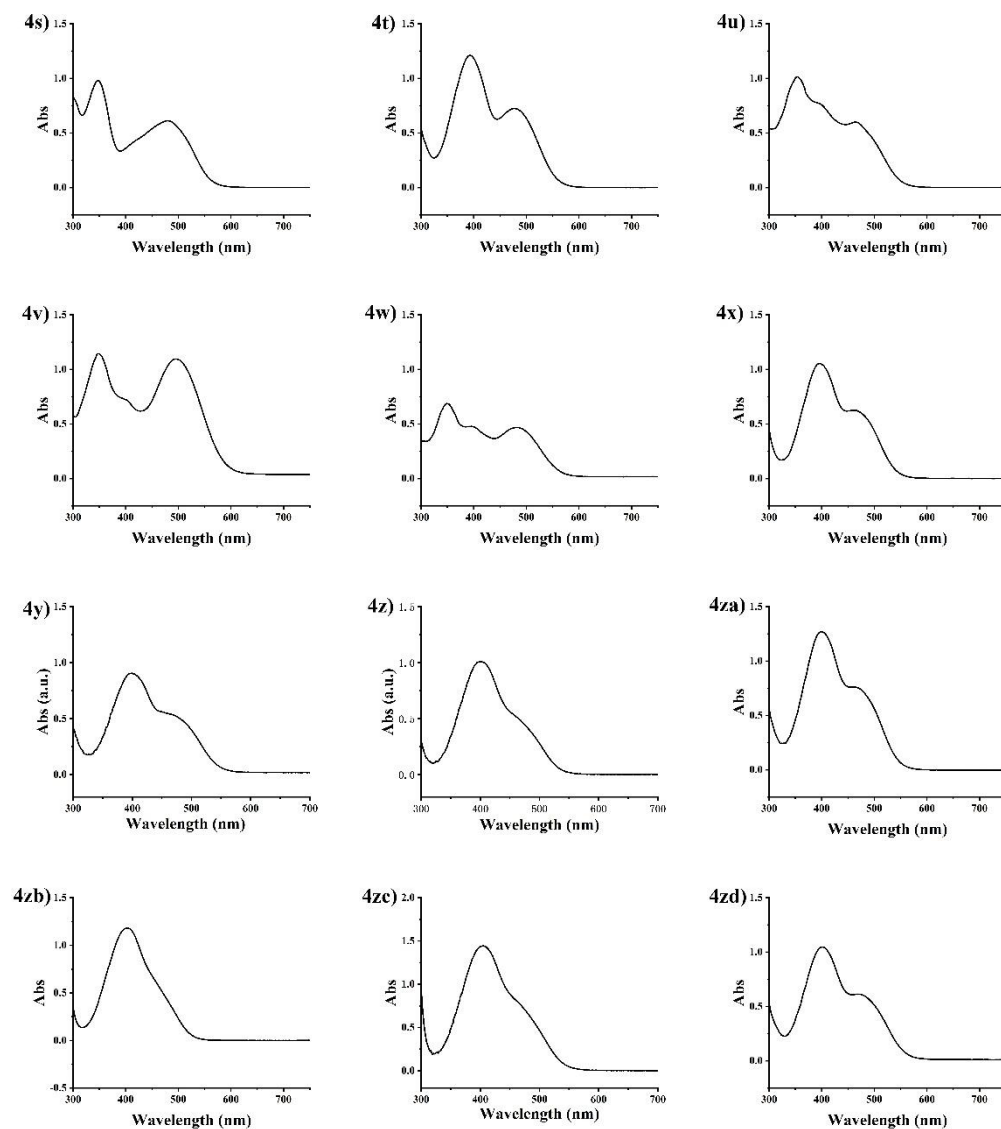


Figure S2. The UV absorption spectra of **4a–4zd** in DCM solution (1×10^{-5} mol/L) at 25 °C.

4.2 The Solution Photographs of Products

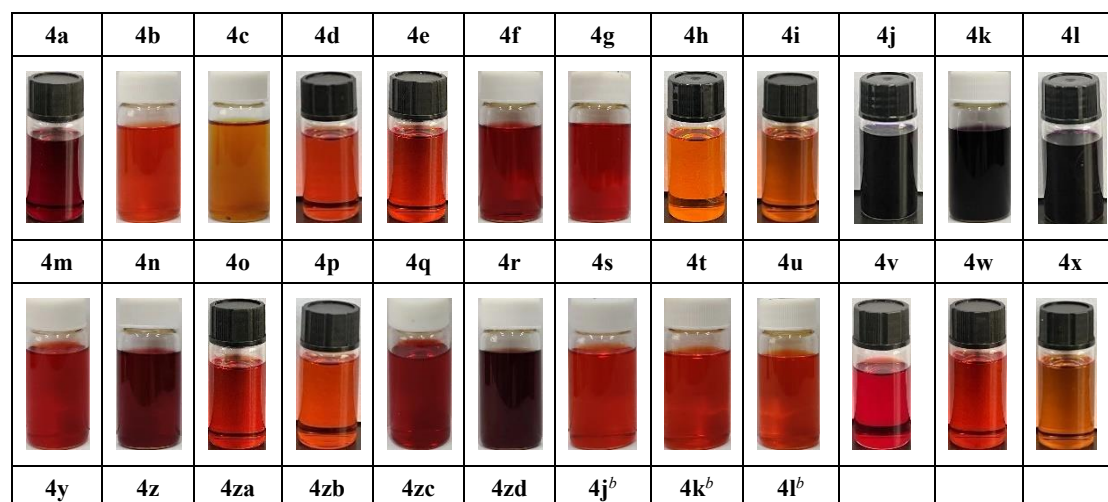
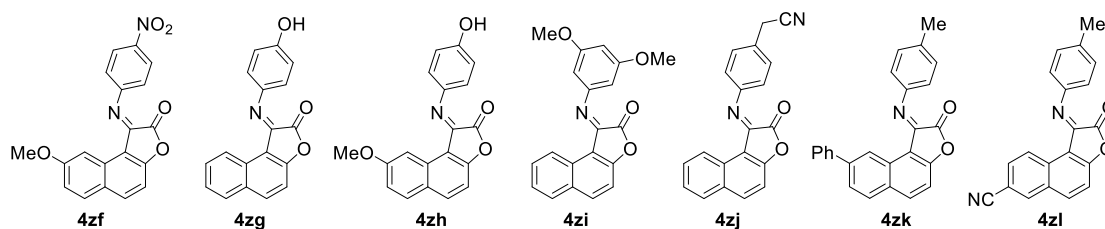




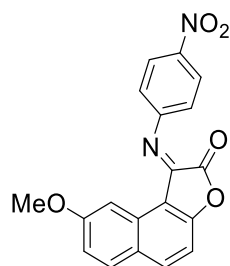
Figure S3. The Solution Photographs of **4a–4zd** in DCM solution (1×10^{-4} mol/L); ^bThe corresponding DCM solution (1×10^{-5} mol/L).

4.3 The HRMS Spectra, UV Absorption Detection, and Solution Photographs of Other Products

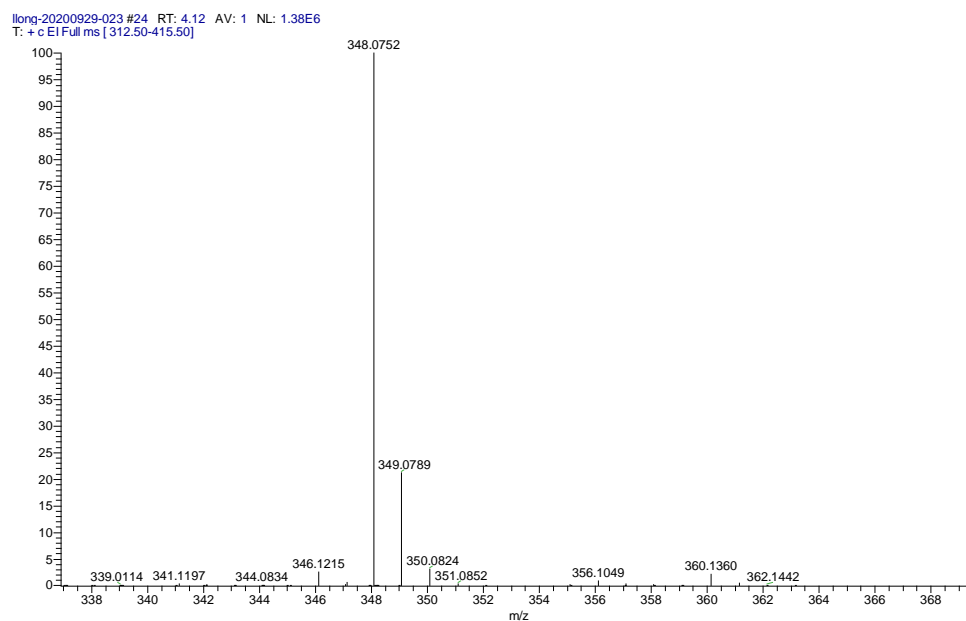


4.3.1 The HRMS Spectra of 4zf–4zl

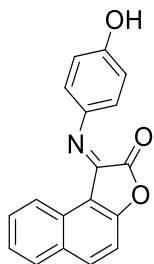
(*Z*)-8-methoxy-1-((4-nitrophenyl)imino)naphtho[2,1-*b*]furan-2(1*H*)-one (4zf)



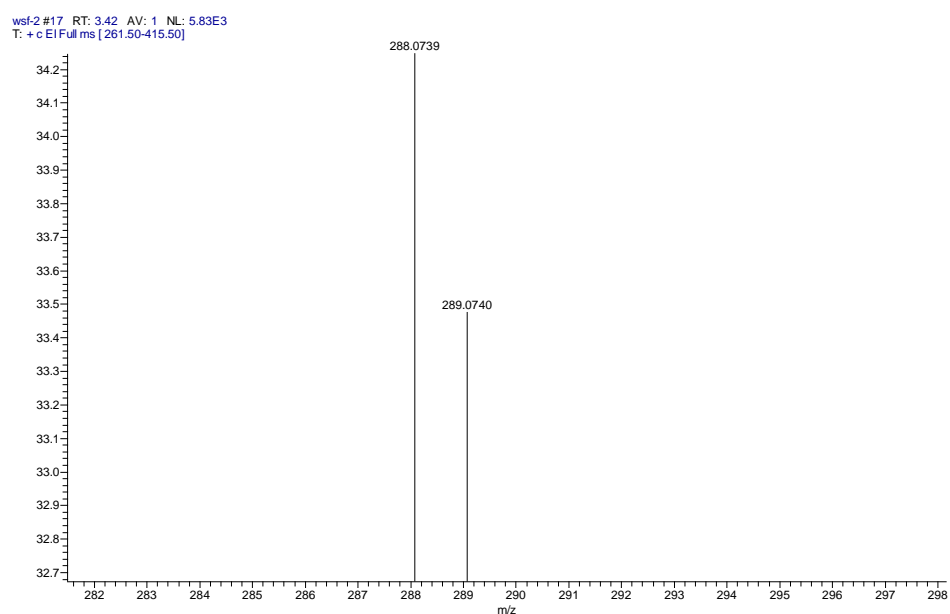
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (5/1) to afford a red solid; HRMS (EI) m/z : $[M]^+$ calcd. for $C_{19}H_{12}N_2O_5$: 348.0746; found: 348.0752.



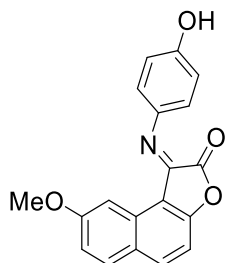
(Z)-1-((4-hydroxyphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4zg)



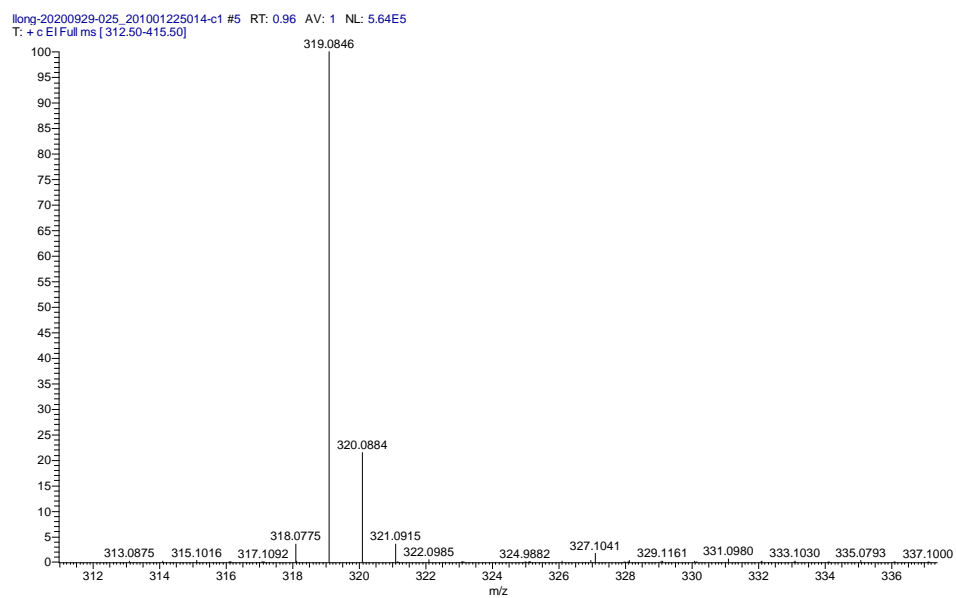
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (10/1) to afford a red solid; HRMS (EI) m/z: $[M]^+$ calcd. for $C_{18}H_{11}NO_3$: 289.0739; found: 289.0740.



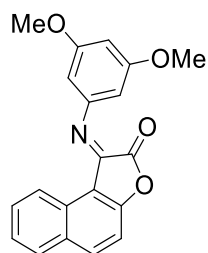
(Z)-1-((4-hydroxyphenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4zh)



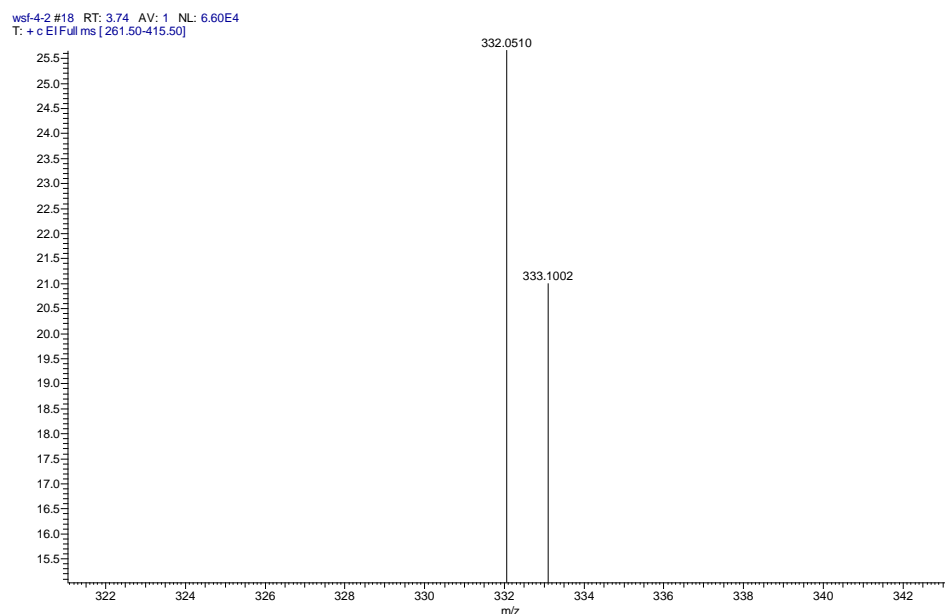
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (10/1) to afford a red solid; HRMS (EI) m/z: $[M]^+$ calcd. for $C_{19}H_{13}NO_4$: 319.0845; found: 319.0846.



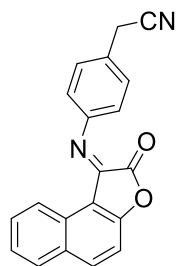
(Z)-1-((3,5-dimethoxyphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4zi)



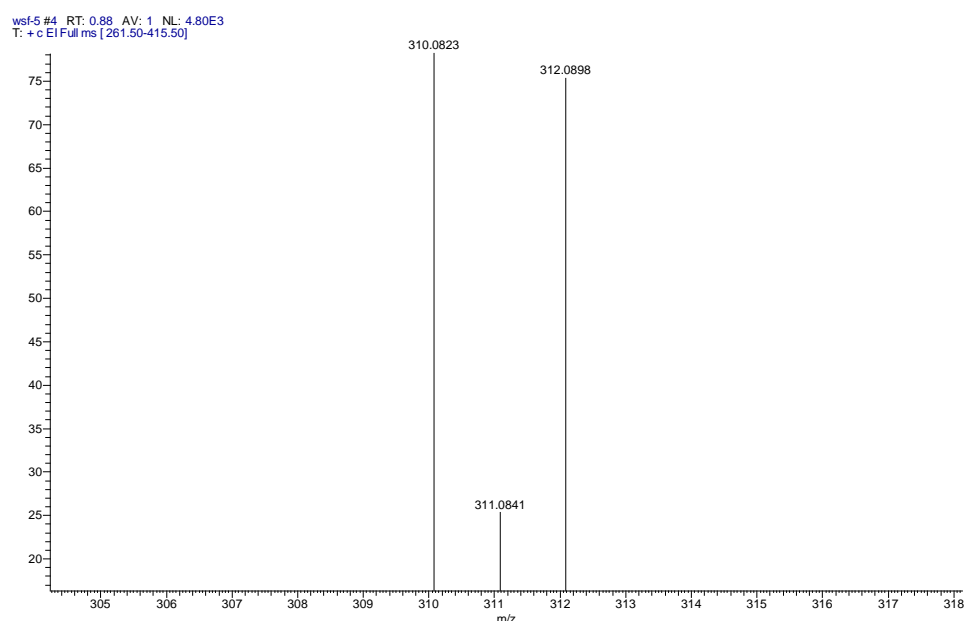
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (10/1) to afford a red solid; HRMS (EI) m/z : $[M]^+$ calcd. for $C_{20}H_{15}NO_4$: 333.1001; found: 333.1002.



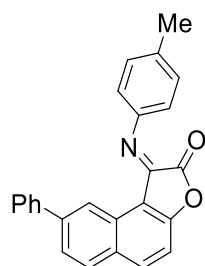
(Z)-2-(4-((2-oxonaphtho[2,1-b]furan-1(2H)-ylidene)amino)phenyl)acetonitrile (4zj)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (10/1) to afford a red solid; HRMS (EI) m/z : $[M]^+$ calcd. for $C_{20}H_{12}N_2O_2$: 312.0899; found: 312.0898.

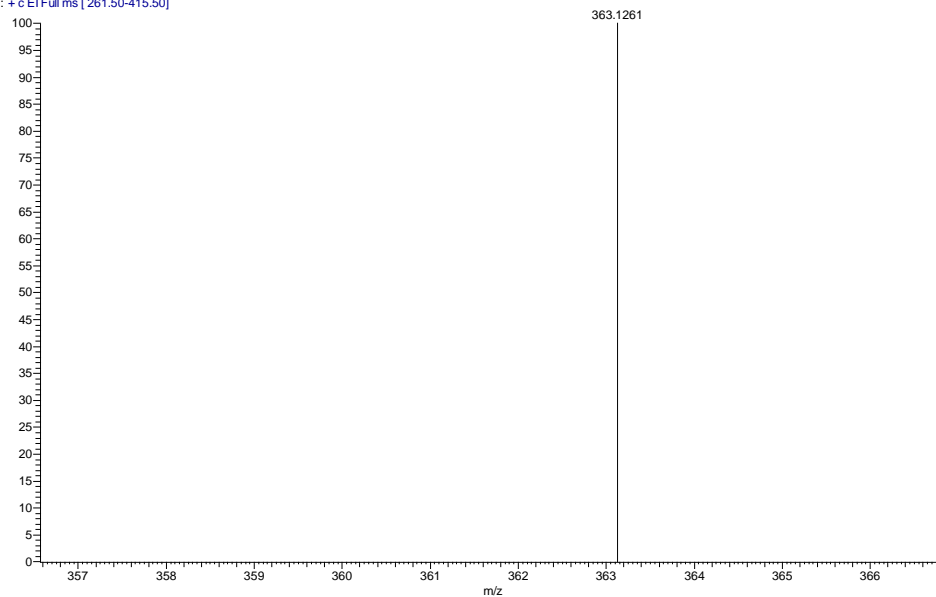


(Z)-8-phenyl-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (4zk)

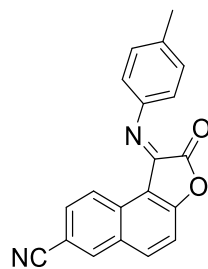


The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a grey solid; HRMS (EI) m/z : $[M]^+$ calcd. for $C_{25}H_{17}NO_2$: 363.1259; found: 363.1261.

wsf-3 #1 RT: 0.22 AV: 1 NL: 4.50E1
T: + c EI Full ms [261.50-415.50]

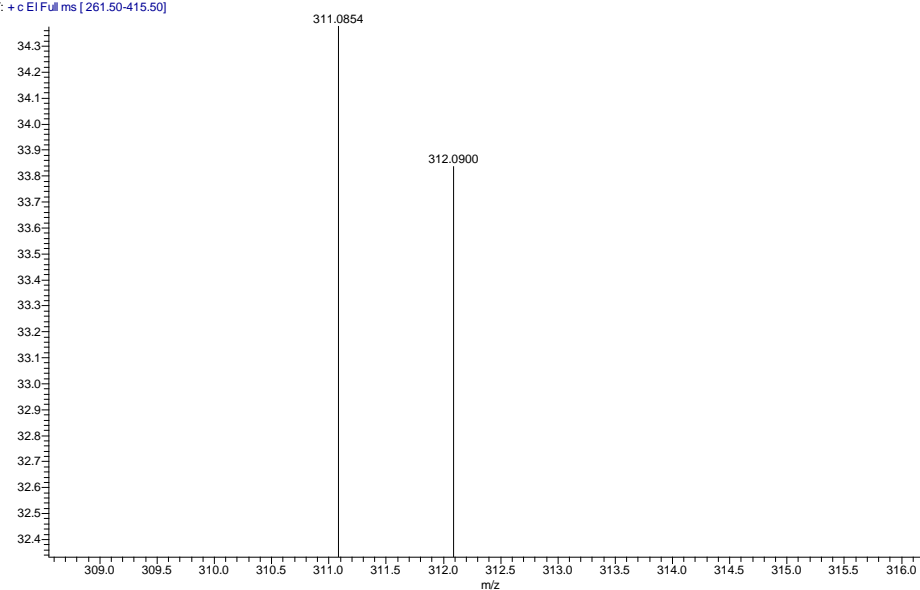


(Z)-2-oxo-1-(p-tolylimino)-1,2-dihydronaphtho[2,1-b]furan-7-carbonitrile (4zl)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (10/1) to afford a red solid; HRMS (EI) m/z : $[M]^+$ calcd. for $C_{20}H_{12}N_2O_2$: 312.0899; found: 312.0900.

wsf-1 #17 RT: 3.46 AV: 1 NL: 2.72E3
T: + c EI Full ms [261.50-415.50]



4.3.2 The UV Absorption Detection of the Above Products

Table S4. The λ_{\max} and ϵ values of compounds

Compound	4zf	4zg	4zh	4zi	4zj	4zk	4zl
λ_{\max}/nm	413	385	361	409	349	346	357
$\epsilon (\times 10^5)$	1.485	0.892	0.721	2.234	0.935	1.158	1.212

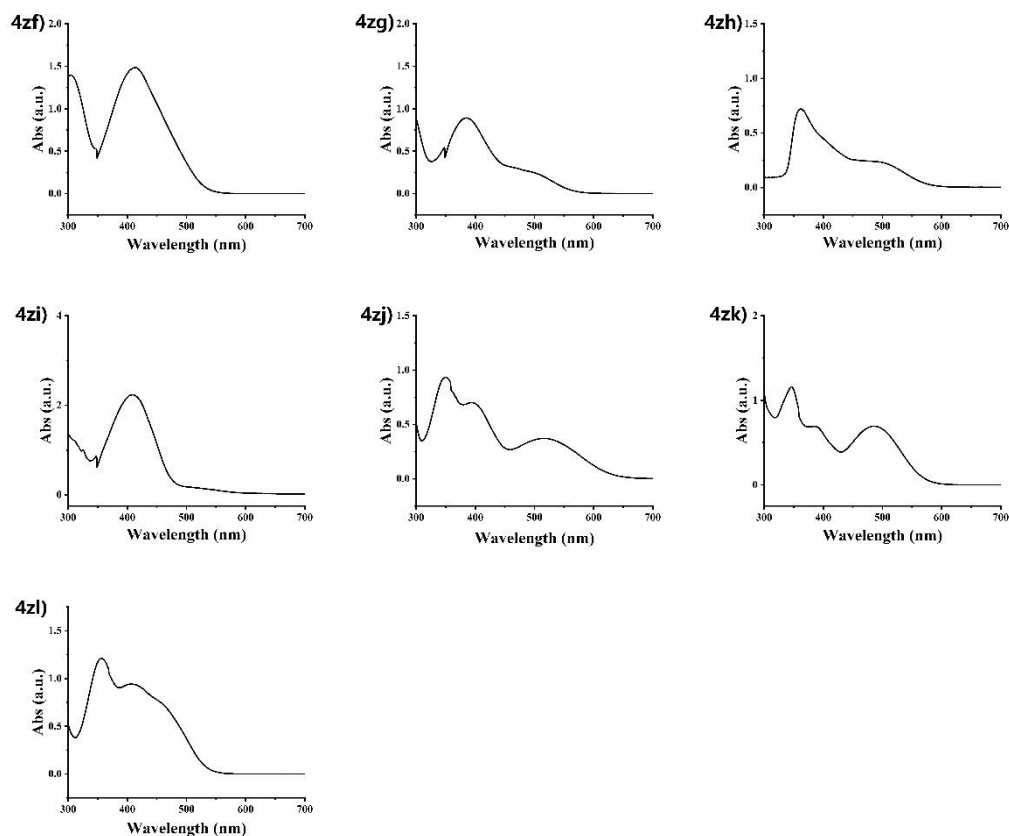


Figure S4. The UV absorption spectra of **4zf–4zl** in DCM solution (1×10^{-5} mol/L) at 25 °C.

4.3.3 The Solution Photographs of Products 4zf–4zl

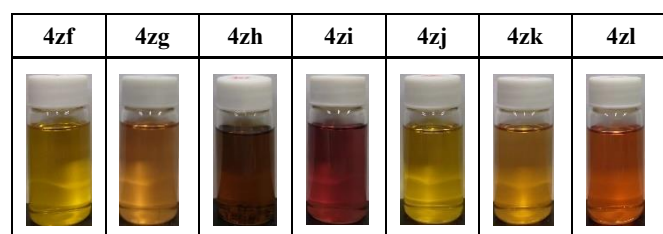


Figure S5. The Solution Photographs of **4zf–4zl** in DCM solution (1×10^{-5} mol/L).

5. Investigations of the Reaction Mechanism

5.1 Detection of hydrogen peroxide

In an oven dried 25 mL Schlenk tube was charged with anilines 2-naphthol (**1a**, 0.3 mmol), **2a** (0.2 mmol), trimethylacetic acid (0.4 mmol), and 5Å molecular sieve (200 mg), after charging oxygen for three times, the ester glyoxalate solution (**3a**, 0.2 mmol), and toluene (4 mL) were added. The reaction mixture was reacted at 100 °C for 14 h. After completion, a small amount of activated MnO₂ powder was added to the reaction solution, bubbles are formed as shown in Figure S6, which supported the inference that oxygen is reduced to hydrogen peroxide.

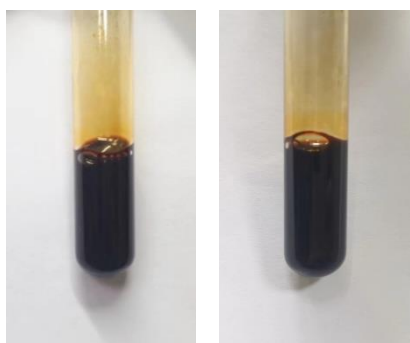


Figure S6. The formation of bubbles when the activated MnO₂ powder was added to the reaction solution.

5.2 Computational details

In our calculations, Gaussian 16 program¹ was used to carry out total density functional theory (DFT) calculations. Geometry optimizations were performed by B3LYP functional^{2,3} with dispersion correction of D3(BJ)⁴, 6-31+G(d) basis set for the all elements. The vibrational frequencies calculations were conducted at the same level of theory to be sure whether every optimized stationary point is an energy minimum or a transition state and evaluate the zero-point vibrational energy and thermal corrections at 298 K. The single point energies and solvent effects based on the gas-phase optimized structures were calculated by the UM06 functional^{5,6} with dispersion correction of D3,⁷ and def2-TZVP basis set⁸ was used for all elements. The solvation energies were evaluated by self-consistent reaction field by SMD implicit solvent model.⁹ In order to adjust the Gibbs free energies from 1 atm to 1 mol/L, a correction of $RT\ln(C_{sol}/C_{gas})$ (1.89 kcal/mol) is added to energies of all species. C_{sol} represents the standard molar concentration in solution (1 mol/L), C_{gas} represents the standard molar concentration in gas phase (0.0446 mol/L), and R represents the gas constant.

Table S5. Zero-point correction (ZPE), thermal correction to enthalpy (TCH), thermal correction to Gibbs free energy (TCG), energies (E), Gibbs free energies(G), and enthalpies (H) (in Hartree) of the structures calculated at the M06-D3/def2-TZVP//B3LYP-D3(BJ)/6-31+G(d) level of theory

	ZPE	TCH	TCG	E	G	H	Imaginary Frequency
O2 (T)	0.003741	0.007048	-0.016239	-150.316860	-150.333099	-150.309812	-

O2 (S)	0.003713	0.007021	-0.015229	-150.257894	-150.273123	-150.250873	-
H2O2	0.026260	0.030427	0.004604	-151.544370	-151.539766	-151.513943	-
HOO•	0.014018	0.017819	-0.008174	-150.901670	-150.909844	-150.883851	-
1a	0.151350	0.159461	0.118628	-460.954329	-460.835701	-460.794868	-
A	0.220382	0.233880	0.180031	-632.006069	-631.826038	-631.772189	-
B	0.138783	0.147529	0.105495	-460.315846	-460.210351	-460.168317	-
C	0.362184	0.385482	0.308500	-1092.337475	-1092.028975	-1091.951993	-
D	0.352207	0.375009	0.298140	-1091.757596	-1091.459456	-1091.382587	-
F	0.207896	0.223232	0.162076	-631.346814	-631.184738	-631.123582	-
TS (T)	0.152610	0.164121	0.114072	-611.232996	-611.118924	-611.068875	-460.79
TS (S)	0.156253	0.167965	0.119289	-611.217161	-611.097872	-611.049196	-194.12
TS1	0.362184	0.385482	0.308500	-1092.317560	-1092.00906	-1091.932078	-513.61
TS2	0.375463	0.402334	0.315439	-1243.223422	-1242.907983	-1242.821088	-1741.41
TS3	0.222031	0.240164	0.172780	-782.226588	-782.053808	-781.986424	-993.59

5.3 Cartesian coordinates of the optimized structure

O2 (T)

O	-3.01716100	2.59493700	0.00000000
O	-1.80208000	2.59493700	0.00000000

O2 (S)

O	-3.01718400	2.59493700	0.00000000
O	-1.80205600	2.59493700	0.00000000

H2O2

O	0.00000000	0.72832800	-0.05465700
O	0.00000000	-0.72832800	-0.05465700
H	0.82272600	0.90151400	0.43725900
H	-0.82272600	-0.90151400	0.43725900

HOO•

O	0.05578100	-0.61182700	0.00000000
O	0.05578100	0.72105800	0.00000000
H	-0.89249700	-0.87385000	0.00000000

1a

O	3.37247800	-0.75422000	0.00000500
C	2.10250300	-0.23415400	-0.00000300
C	1.04033500	-1.11216600	-0.00000500
C	-0.29160000	-0.62665200	-0.00000800
C	-1.41309300	-1.50118200	-0.00000800
C	-2.69796000	-1.00352100	0.00000400
C	-2.92853700	0.39531600	0.00001400

C	-1.86309000	1.26912700	0.00000400
C	-0.52680500	0.78859800	-0.00000800
C	0.59417700	1.65965600	-0.00000800
C	1.87931300	1.16678100	-0.00000600
H	4.02225600	-0.03463800	0.00010100
H	1.23180400	-2.18128700	0.00000600
H	-1.23925100	-2.57456500	-0.00001600
H	-3.54283300	-1.68711500	0.00000400
H	-3.94642900	0.77509600	0.00002600
H	-2.03215100	2.34368400	0.00000400
H	0.42658700	2.73398800	-0.00000600
H	2.72873500	1.84777800	-0.00001400
A			
C	-3.06777900	-1.33120700	-0.00005600
C	-4.05829900	-0.33641700	-0.00000300
C	-3.64619000	1.00409600	0.00003300
C	-2.29502900	1.33711100	0.00002200
C	-1.30717000	0.33835600	-0.00001800
C	-1.71384200	-1.00809700	-0.00006200
C	-5.52431600	-0.69268400	0.00002600
N	0.02178200	0.80152800	-0.00002500
C	1.02350400	0.01285100	0.00005600
C	2.40753000	0.58082200	0.00007000
O	2.71078100	1.75381700	-0.00007000
O	3.30285000	-0.43714400	0.00004000
C	5.52164800	-1.32873000	0.00003700
C	4.69862400	-0.05560400	-0.00005300
H	-3.36494900	-2.37732900	-0.00009800
H	-4.39343900	1.79446700	0.00006600
H	-1.97005800	2.37291500	0.00004500
H	-0.98237000	-1.81021600	-0.00011500
H	-5.67308400	-1.77704700	-0.00022100
H	-6.03133300	-0.28349200	0.88278900
H	-6.03147800	-0.28306200	-0.88245200
H	0.96821300	-1.08124200	0.00014400
H	6.58855600	-1.07767400	-0.00003300
H	5.30983100	-1.93271900	0.88874000
H	5.30975600	-1.93289500	-0.88852900
H	4.88834500	0.56053400	-0.88479900
H	4.88841400	0.56070800	0.88455600
B			
O	3.37292100	-0.75356300	-0.00000500

C	2.20920100	-0.28295700	-0.00000500
C	1.05200500	-1.14844400	-0.00000300
C	-0.25808100	-0.63991800	-0.00000100
C	-1.39816800	-1.49370400	0.00000100
C	-2.67523600	-0.96766000	0.00000500
C	-2.86248200	0.43145400	0.00000800
C	-1.76760200	1.28899900	0.00000500
C	-0.45660400	0.78453700	0.00000100
C	0.69530800	1.65205100	-0.00000200
C	1.96145400	1.15984300	-0.00000500
H	1.23313700	-2.21985900	-0.00000300
H	-1.24309900	-2.56948900	-0.00000200
H	-3.53773200	-1.62765500	0.00000700
H	-3.86891800	0.84023400	0.00001300
H	-1.91912800	2.36561100	0.00000700
H	0.52156000	2.72602000	-0.00000200
H	2.83203200	1.80844000	-0.00000700

C			
C	-1.13863800	-0.58437800	-0.27278100
C	-2.51063300	-1.26280900	-0.14180700
N	-0.18030100	-1.26441700	0.54361200
O	-2.74428300	-2.27063400	0.48028200
C	1.09667600	-1.37853900	0.14926100
C	-1.24016500	0.93408600	0.18614700
C	0.00745500	1.67484200	-0.21935700
C	1.02025300	1.92178200	0.73080500
C	0.78051300	1.60613900	2.13368500
C	-0.40729000	1.15900800	2.59594800
C	-1.53620200	0.93458500	1.68681100
C	0.20301400	2.06873300	-1.54426900
C	1.40044400	2.66640000	-1.94416900
C	2.42235800	2.87228400	-1.01237100
C	2.22660100	2.50922200	0.31818500
O	-2.67331500	0.71507400	2.08961900
C	1.65733000	-0.98951900	-1.10955200
C	3.01377000	-1.12454700	-1.35231400
C	3.89073500	-1.65246800	-0.38658500
C	3.34735400	-2.05959000	0.85204700
C	1.99843300	-1.93707200	1.11456700
C	5.36344100	-1.79996200	-0.66332000
O	-3.42290100	-0.59363900	-0.87950100
C	-5.65351800	-0.13198900	-1.60122500
C	-4.78725400	-1.07045100	-0.78474100

H	-0.91231700	-0.56501100	-1.34913500
H	-2.11349500	1.34157100	-0.32920700
H	1.60030600	1.78399000	2.82740400
H	-0.58699700	0.97888600	3.65124400
H	-0.58671200	1.89561800	-2.27259200
H	1.53647600	2.96733700	-2.97931800
H	3.36026300	3.32570600	-1.32029700
H	3.00878000	2.68440100	1.05302600
H	1.02702500	-0.56925200	-1.88269800
H	3.41413100	-0.80975900	-2.31332200
H	4.00754400	-2.47678600	1.60959500
H	1.57272800	-2.24252400	2.06520800
H	5.64100000	-1.34017300	-1.61701400
H	5.96564400	-1.33419700	0.12695300
H	5.65367100	-2.85844400	-0.70450400
H	-6.69806200	-0.46115200	-1.55584400
H	-5.59743300	0.88926300	-1.20976400
H	-5.34161600	-0.12092100	-2.65141400
H	-4.82335000	-2.09869500	-1.15849600
H	-5.06953500	-1.07987600	0.27191700
D			
C	0.18964400	0.43118800	0.03616300
C	-0.68055900	1.65031100	0.31108700
N	-0.23887500	-0.76504700	0.01366000
O	-1.35817800	1.77851000	1.30514100
C	-1.61153900	-1.07706100	0.06323600
C	1.68831800	0.72886300	-0.12899300
C	2.45629600	-0.49667600	-0.55464500
C	3.25278500	-1.18978600	0.37719200
C	3.40791300	-0.66156000	1.72483700
C	2.90492800	0.53067500	2.11942000
C	2.14899000	1.37298800	1.19381600
C	2.34860500	-0.98191600	-1.85679700
C	3.01619900	-2.14654700	-2.24160600
C	3.79888800	-2.84419000	-1.31617200
C	3.91605100	-2.36483200	-0.01422500
O	1.83166300	2.52904700	1.45324400
C	-2.03619700	-2.08291500	0.93886200
C	-3.38263500	-2.43961300	0.99342200
C	-4.33074400	-1.84339600	0.15119100
C	-3.88374400	-0.86885300	-0.75277700
C	-2.54646900	-0.48352300	-0.79953100
C	-5.78623900	-2.24248000	0.19868800

O	-0.57097800	2.55231100	-0.67713000
C	-0.92990600	4.71108500	-1.64303400
C	-1.24093600	3.82278900	-0.45586300
H	1.76237100	1.51404400	-0.89066300
H	3.99522100	-1.25456000	2.42394900
H	3.08020500	0.93209500	3.11277900
H	1.72563800	-0.45194200	-2.57315100
H	2.92163000	-2.51175900	-3.26041700
H	4.31538600	-3.75313900	-1.61096500
H	4.52871800	-2.89482300	0.71126400
H	-1.30565300	-2.56185000	1.58375900
H	-3.70082700	-3.20534700	1.69708800
H	-4.59463800	-0.40324000	-1.43244200
H	-2.21742800	0.26159500	-1.51891200
H	-5.96177200	-3.01692800	0.95239600
H	-6.42626100	-1.38570700	0.44476400
H	-6.12630300	-2.63387300	-0.76847200
H	-1.41866400	5.68328300	-1.51201700
H	0.14862600	4.87716100	-1.73273400
H	-1.29389500	4.26579500	-2.57531000
H	-2.31288800	3.63171400	-0.34624000
H	-0.86526900	4.23655700	0.48403300
H	-4.89479200	0.76900800	0.57593200
H	-4.95621600	-0.12599400	-0.95203600
F			
C	3.14273200	-0.83365900	-0.90240500
C	4.03256900	-0.27929600	0.03463700
C	3.51095700	0.56674400	1.01995700
C	2.14643100	0.85573200	1.07565300
C	1.27746700	0.29552700	0.13704200
C	1.78191500	-0.55552100	-0.85919800
C	5.50732700	-0.59284600	-0.03149500
N	-0.09248900	0.61780400	0.23387500
C	-1.05115300	0.25048400	-0.44566500
C	-2.47719800	0.58588100	-0.46743600
O	-2.92680000	1.55577800	-1.05055300
O	-3.21002900	-0.36492800	0.14287000
C	-5.27499000	-1.36331300	0.81515500
C	-4.65004000	-0.18404100	0.09715100
H	3.52675700	-1.49341500	-1.67752100
H	4.17927600	1.00796200	1.75509800
H	1.74420600	1.51356100	1.83983900
H	1.09933600	-0.98609700	-1.58585000

H	5.68824700	-1.67161700	0.05311000
H	5.93994200	-0.26603600	-0.98537800
H	6.05748800	-0.09601900	0.77372800
H	-6.36640900	-1.26325600	0.80377300
H	-5.00900000	-2.30598200	0.32566400
H	-4.94288500	-1.40769200	1.85760100
H	-4.89479200	0.76900800	0.57593200
H	-4.95621600	-0.12599400	-0.95203600

TS (T)

O	-2.67951200	0.78684600	-0.78371100
C	-1.42765900	0.79017900	-0.50411700
C	-0.61754800	-0.37225900	-0.69376600
C	0.75068100	-0.37828000	-0.35999600
C	1.56024600	-1.53167900	-0.53499700
C	2.90089600	-1.50863300	-0.20603800
C	3.48198500	-0.33255800	0.30865400
C	2.71430400	0.81000400	0.48784600
C	1.34773300	0.81622000	0.16273200
C	0.52855300	1.98524900	0.33133100
C	-0.79540900	1.98203600	0.01914100
H	-3.29620200	-0.26804400	-0.15148000
H	-1.07809900	-1.25324400	-1.12567600
H	1.10265400	-2.43508200	-0.92836400
H	3.51023700	-2.39694500	-0.34142700
H	4.53681100	-0.32072400	0.56626400
H	3.16661100	1.71552400	0.88357500
H	0.99742800	2.88361500	0.72545100
H	-1.42241000	2.85715000	0.15240300
O	-3.60096100	-1.07302900	0.51227100
O	-2.49199200	-1.50680800	1.04075400

TS (S)

O	2.77824300	0.16961200	1.12801200
C	1.50816600	0.40023500	0.70739100
C	0.58852000	-0.63503700	0.80748100
C	-0.76171300	-0.43535700	0.42041200
C	-1.73059700	-1.46399500	0.52297500
C	-3.03338000	-1.24447600	0.12808800
C	-3.42019300	0.01285100	-0.38625400
C	-2.49678000	1.03244000	-0.49877600
C	-1.15340700	0.83751200	-0.10274100
C	-0.17448700	1.87024800	-0.20709600
C	1.12101800	1.66505600	0.18355600

H	3.39829800	0.42509600	0.41492000
H	0.91562900	-1.58979200	1.20381800
H	-1.42790300	-2.43055400	0.91635200
H	-3.76741500	-2.04057900	0.21057600
H	-4.44884900	0.17464900	-0.69464500
H	-2.79121600	2.00049000	-0.89579400
H	-0.47841200	2.83543800	-0.60349300
H	1.86346400	2.45444400	0.11375900
O	3.22113500	-1.00408000	-1.05903300
O	2.00731400	-0.92378800	-1.33344200

TS1

C	-1.15060500	-0.89149500	-0.48288000
C	-2.59595100	-1.26984900	-0.28073400
N	-0.27151700	-1.47747800	0.34268700
O	-3.04055100	-1.86472400	0.66994700
C	1.06764600	-1.45620400	0.06598100
C	-1.27759400	1.10708600	-0.00187000
C	0.05812200	1.60232500	-0.27286100
C	1.01577600	1.60111200	0.77758000
C	0.59450100	1.22602800	2.11478400
C	-0.69405800	0.96130000	2.42512300
C	-1.74577300	1.02718200	1.40423900
C	0.44723600	2.03506800	-1.55357700
C	1.75318900	2.43787300	-1.79949600
C	2.70409300	2.40263200	-0.77020900
C	2.33403400	1.99248000	0.50548900
O	-2.94482100	0.98980700	1.68789700
C	1.67362700	-1.07259700	-1.15966200
C	3.05051900	-1.03273500	-1.28175900
C	3.89450500	-1.37244300	-0.20918600
C	3.30109700	-1.79437700	0.99250500
C	1.92474000	-1.85088400	1.12633600
C	5.38984300	-1.26258600	-0.34577300
O	-3.32003700	-0.82507200	-1.33722000
C	-5.36380800	0.30842600	-0.66838400
C	-4.75827500	-0.96596100	-1.23323400
H	-0.90786600	-0.66813800	-1.52586000
H	-2.05598600	1.34144400	-0.72340500
H	1.36094200	1.19659200	2.88617900
H	-1.01035500	0.73160500	3.43698000
H	-0.29093200	2.04766100	-2.35168500
H	2.03906400	2.77447400	-2.79173900
H	3.72848400	2.70493200	-0.96669000

H	3.06653400	1.97223100	1.30784300
H	1.06114200	-0.80041100	-2.01047000
H	3.49267500	-0.72440900	-2.22586200
H	3.93619400	-2.07993600	1.82738100
H	1.45446000	-2.16111200	2.05336600
H	5.73074400	-1.61500900	-1.32558300
H	5.71616100	-0.21782400	-0.24925100
H	5.90808700	-1.84109200	0.42502500
H	-6.44980800	0.19524500	-0.57212500
H	-4.93641100	0.52745200	0.31342400
H	-5.16456800	1.15833000	-1.33042000
H	-5.09490700	-1.16520600	-2.25389700
H	-4.97694100	-1.82887100	-0.60148300

TS2

C	0.99336400	-0.35452300	-0.27598100
C	2.31789200	-0.86376200	-0.81167500
N	-0.04315700	-0.48922100	-1.18592900
O	2.49242500	-1.37292800	-1.89097100
C	-1.28316500	-0.91575500	-0.95487400
C	1.09015600	1.11458300	0.27428600
C	-0.21215100	1.49526600	0.93396500
C	-1.14640500	2.28465300	0.23239900
C	-0.77785900	2.86526100	-1.05080000
C	0.45443900	2.75056100	-1.58945000
C	1.51050800	1.99077400	-0.91548500
C	-0.54273600	1.00467100	2.19684400
C	-1.79639400	1.26502500	2.75327200
C	-2.73532200	2.01952000	2.04776400
C	-2.40662700	2.53151800	0.79634700
O	2.67061000	1.98647500	-1.30754700
C	-1.73346100	-1.55148200	0.24835100
C	-3.06362500	-1.89103400	0.39043800
C	-4.00852800	-1.64413300	-0.62524000
C	-3.56315400	-1.04953300	-1.82448500
C	-2.24221400	-0.69767900	-1.99597700
C	-5.46053100	-1.98284700	-0.42675800
O	3.29875100	-0.68341000	0.11282300
C	5.59003200	-0.54051800	0.75748500
C	4.64699100	-0.85975400	-0.38439600
H	0.89672400	-1.05353200	0.84696500
H	1.91191500	1.10721900	0.99309000
H	-1.53958400	3.44970100	-1.56351100
H	0.72565300	3.23534600	-2.52167000

H	0.17455900	0.39169700	2.73407000
H	-2.04030400	0.87035100	3.73522800
H	-3.71459900	2.21403700	2.47501700
H	-3.12377000	3.13441800	0.24500300
H	-1.02296100	-1.76943800	1.03650600
H	-3.39284800	-2.36654800	1.31094100
H	-4.27821600	-0.86620800	-2.62243700
H	-1.88927100	-0.23051800	-2.90907100
H	-6.00740400	-1.12688200	-0.00709200
H	-5.94528800	-2.24369000	-1.37318600
H	-5.58601100	-2.81953700	0.26774700
H	6.62752500	-0.64397000	0.42267300
H	5.43886500	0.48555000	1.10616300
H	5.43640700	-1.22101700	1.60268000
H	4.75986800	-1.88579700	-0.74658500
H	4.78039400	-0.17953800	-1.22960300
O	1.03700900	-1.60989900	1.94305400
O	1.94034200	-2.65853200	1.66787100
H	2.79007700	-2.17561800	1.64054500

TS3

C	-3.26158600	0.93667500	0.17770500
C	-4.15314300	-0.14790000	0.20400300
C	-3.63422300	-1.44004900	0.05624900
C	-2.26987100	-1.64805200	-0.11339200
C	-1.39690800	-0.55349200	-0.14648300
C	-1.89722100	0.75641100	-0.00138300
C	-5.63394800	0.08260800	0.36549800
N	-0.03928500	-0.82061800	-0.30123800
C	0.97145600	-0.17023900	-0.54499500
C	2.42223400	-0.50657700	-0.59553800
O	3.08586500	-0.34490700	-1.59539200
O	2.85289600	-0.92758500	0.59738800
C	4.99112100	0.20423200	0.96918400
C	4.29183800	-1.12096600	0.72117300
H	-3.64683700	1.94617400	0.29623400
H	-4.30643300	-2.29319600	0.08074400
H	-1.86273200	-2.64855000	-0.21307200
H	-1.22016400	1.61025400	-0.01620800
H	-6.07582600	0.45964000	-0.56585600
H	-6.15722600	-0.84109600	0.62966700
H	-5.83819500	0.82657900	1.14314900
H	0.84493700	1.32050500	-1.01440600
H	6.06095300	0.03088600	1.13028000

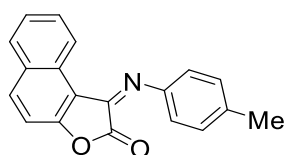
H	4.87506800	0.86882800	0.10911200
H	4.58086500	0.69807800	1.85500500
H	4.38905300	-1.80178900	1.56859900
H	4.65625800	-1.60943400	-0.18510300
O	1.30720800	2.10112500	-0.44299100
O	0.42862800	2.77406000	0.28329400

6. Reference

- (1) Gaussian 16, Revision B.01, Frisch, M. J.; Trucks, G. W.; Schlegel, H. B.; Scuseria, G. E.; Robb, M. A.; Cheeseman, J. R.; Scalmani, G.; Barone, V.; Petersson, G. A.; Nakatsuji, H.; Li, X.; Caricato, M.; Marenich, A. V.; Bloino, J.; Janesko, B. G.; Gomperts, R.; Mennucci, B.; Hratchian, H. P.; Ortiz, J. V.; Izmaylov, A. F.; Sonnenberg, J. L.; Williams-Young, D.; Ding, F.; Lipparini, F.; Egidi, F.; Goings, J.; Peng, B.; Petrone, A.; Henderson, T.; Ranasinghe, D.; Zakrzewski, V. G.; Gao, J.; Rega, N.; Zheng, G.; Liang, W.; Hada, M.; Ehara, M.; Toyota, K.; Fukuda, R.; Hasegawa, J.; Ishida, M.; Nakajima, T.; Honda, Y.; Kitao, O.; Nakai, H.; Vreven, T.; Throssell, K.; Montgomery, J. A.; Peralta, Jr., J. E.; Ogliaro, F.; Bearpark, M. J.; Heyd, J. J.; Brothers, E. N.; Kudin, K. N.; Staroverov, V. N.; Keith, T. A.; Kobayashi, R.; Normand, J.; Raghavachari, K.; Rendell, A. P.; Burant, J. C.; Iyengar, S. S.; Tomasi, J.; Cossi, M.; Millam, J. M.; Klene, M.; Adamo, C.; Cammi, R.; Ochterski, J. W.; Martin, R. L.; Morokuma, K.; Farkas, O.; Foresman, J. B.; Fox, D. J. Gaussian, Inc., Wallingford CT, **2016**.
- (2) Becke, A. D. Density-functional thermochemistry. III. The role of exact exchange. *J. Chem. Phys.* **2016**, *98*, 5648.
- (3) Lee, C.; Yang, W.; Parr, R. G. Development of the Colle-Salvetti correlation-energy formula into a functional of the electron density. *Phys. Rev. B: Condens. Matter Mater. Phys.* **1988**, *37*, 785.
- (4) Grimme, S.; Ehrlich, S.; and Goerigk, L. Effect of the damping function in dispersion corrected density functional theory. *J. Comput. Chem.* **2011**, *32*, 1456.
- (5) Zhao, Y.; Truhlar, D. G. The M06 suite of density functionals for main group thermochemistry, thermochemical kinetics, noncovalent interactions, excited states, and transition elements: two new functionals and systematic testing of four M06-class functionals and 12 other functionals. *Theor. Chem. Acc.* **2008**, *120*, 215.
- (6) Zhao, Y.; Truhlar, D. G. Density functionals with broad applicability in chemistry. *Acc. Chem. Res.* **2008**, *41*, 157.
- (7) Grimme, S.; Antony, J.; Ehrlich, S.; Krieg, H. A consistent and accurate ab initio parametrization of density functional dispersion correction (DFT-D) for the 94 elements H-Pu. *J. Chem. Phys.* **2010**, *132*, 154104.
- (8) Weigend, F.; Ahlrichs, R. Balanced basis sets of split valence, triple zeta valence and quadruple zeta valence quality for H to Rn: Design and assessment of accuracy. *Phys. Chem. Chem. Phys.* **2005**, *7*, 3297.
- (9) Cossi, M.; Rega, N.; Scalmani, G.; Barone, V. Energies, structures, and electronic properties of molecules in solution with the C-PCM solvation model. *J. Comput. Chem.* **2003**, *24*, 669.

7. Characterization Data for the Products

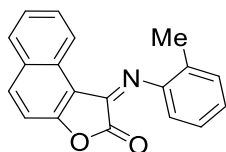
(Z)-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (4a)



The title compound was prepared according to the general procedure and purified by column chromatography on

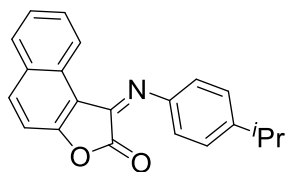
silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 92% yield (52.8 mg). mp 170–172 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.96 (d, *J* = 8.4 Hz, 1H), 8.06 (d, *J* = 8.8 Hz, 1H), 7.88 (d, *J* = 8.0 Hz, 1H), 7.68 (t, *J* = 7.6 Hz, 1H), 7.54–7.51 (m, 1H), 7.29 (d, *J* = 8.8 Hz, 1H), 7.26–7.22 (m, 2H), 7.09–7.07 (m, 2H), 2.40 (s, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 158.4, 155.2, 147.2, 146.0, 136.8, 136.2, 130.9, 130.0, 129.4, 129.1, 128.6, 126.1, 124.2, 119.9, 115.1, 111.8, 21.2. HRMS (EI) *m/z*: [M]⁺ calcd. for C₁₉H₁₃NO₂: 287.0946; found: 287.0943.

(Z)-1-(*o*-tolylimino)naphtho[2,1-*b*]furan-2(1*H*)-one (4b)



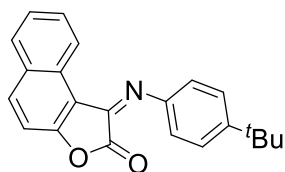
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 89% yield (51.1 mg). mp 171–173 °C; ¹H NMR (400 MHz, CDCl₃): δ 9.00 (d, *J* = 8.0 Hz, 1H), 8.09 (d, *J* = 8.8 Hz, 1H), 7.90 (d, *J* = 8.4 Hz, 1H), 7.70 (t, *J* = 7.6 Hz, 1H), 7.54 (t, *J* = 7.6 Hz, 1H), 7.33–7.30 (m, 2H), 7.25–7.14 (m, 2H), 6.90 (d, *J* = 7.6 Hz, 1H), 2.29 (s, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 158.8, 155.2, 147.8, 147.6, 137.1, 131.0, 130.6, 130.2, 129.2, 128.7, 128.2, 126.2, 126.1, 126.0, 124.1, 117.3, 114.8, 111.9, 18.2. HRMS (EI) *m/z*: [M]⁺ calcd. for C₁₉H₁₃NO₂: 287.0946; found: 287.0949.

(Z)-1-((4-isopropylphenyl)imino)naphtho[2,1-*b*]furan-2(1*H*)-one (4c)



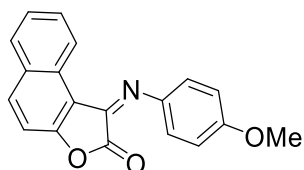
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 93% yield (58.6 mg). mp 175–177 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.96 (d, *J* = 8.4 Hz, 1H), 8.06 (d, *J* = 8.8 Hz, 1H), 7.88 (d, *J* = 8.2 Hz, 1H), 7.68 (t, *J* = 7.7 Hz, 1H), 7.52 (t, *J* = 7.6 Hz, 1H), 7.36–7.23 (m, 3H), 7.14 (d, *J* = 8.2 Hz, 2H), 3.01–2.93 (m, 1H), 1.30 (d, *J* = 6.9 Hz, 6H); ¹³C NMR (101 MHz, CDCl₃): δ 158.4, 155.2, 147.3, 146.9, 146.0, 136.8, 130.9, 130.0, 129.1, 128.6, 126.7, 126.0, 124.1, 120.2, 115.1, 111.7, 33.8, 24.0. HRMS (EI) *m/z*: [M]⁺ calcd. for C₂₁H₁₇NO₂: 315.1259; found: 315.1255.

(Z)-1-((4-*tert*-butyl)phenyl)imino)naphtho[2,1-*b*]furan-2(1*H*)-one (4d)



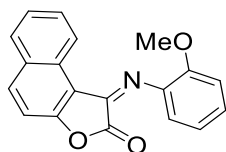
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 91% yield (59.9 mg). mp 181–183 °C ; ¹H NMR (400 MHz, CDCl₃): δ 8.94 (d, *J* = 8.4 Hz, 1H), 8.03 (d, *J* = 8.8 Hz, 1H), 7.86 (d, *J* = 8.0 Hz, 1H), 7.66 (t, *J* = 7.6 Hz, 1H), 7.50 (t, *J* = 7.6 Hz, 1H), 7.44 (d, *J* = 7.6 Hz, 2H), 7.28–7.24 (m, 1H), 7.14 (d, *J* = 7.6 Hz, 2H), 1.36 (s, 9H); ¹³C NMR (101 MHz, CDCl₃): δ 158.4, 155.3, 149.7, 146.9, 145.7, 136.8, 130.9, 130.1, 129.1, 128.6, 126.1, 125.7, 124.2, 120.0, 115.2, 111.8, 34.7, 31.4. HRMS (EI) *m/z*: [M]⁺ calcd. for C₂₂H₁₉NO₂: 329.1416; found: 329.1418.

(Z)-1-((4-methoxyphenyl)imino)naphtho[2,1-*b*]furan-2(1*H*)-one (4e)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a dark red solid in 95% yield (57.6 mg). mp 195–197 °C ; ¹H NMR (400 MHz, CDCl₃): δ 8.99 (d, *J* = 8.4 Hz, 1H), 8.03 (d, *J* = 8.8 Hz, 1H), 7.88 (d, *J* = 8.4 Hz, 1H), 7.68 (t, *J* = 7.6 Hz, 1H), 7.52 (t, *J* = 7.6 Hz, 1H), 7.34–7.26 (m, 3H), 6.97 (d, *J* = 8.8 Hz, 2H), 3.09 (s, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 159.1, 158.0, 155.7, 145.8, 141.2, 136.3, 131.0, 129.9, 129.1, 128.6, 126.0, 124.2, 123.5, 115.6, 113.9, 111.7, 55.5. HRMS (EI) *m/z*: [M]⁺ calcd. for C₁₉H₁₃NO₃: 303.0895; found: 303.0896.

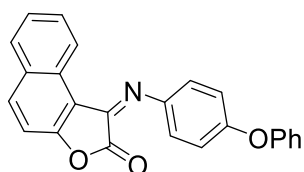
(Z)-1-((2-methoxyphenyl)imino)naphtho[2,1-*b*]furan-2(1*H*)-one (4f)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 89% yield (54.0 mg). mp 201–

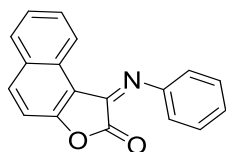
202 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.98 (d, *J* = 8.4 Hz, 1H), 8.06 (d, *J* = 8.8 Hz, 1H), 7.88 (d, *J* = 8.0 Hz, 1H), 7.70–7.66 (m, 1H), 7.52 (t, *J* = 7.6 Hz, 1H), 7.29 (d, *J* = 8.8 Hz, 1H), 7.21–7.17 (m, 1H), 7.08–6.97 (m, 3H), 3.79 (s, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 158.7, 155.1, 149.7, 148.2, 138.4, 137.0, 130.9, 130.1, 129.1, 128.8, 126.6, 126.1, 124.4, 120.9, 120.8, 114.8, 111.8, 111.4, 55.6. HRMS (EI) *m/z*: [M]⁺ calcd. for C₁₉H₁₃NO₃: 303.0895; found: 303.0899.

(Z)-1-((4-phenoxyphenyl)imino)naphtho[2,1-*b*]furan-2(1*H*)-one (4g)



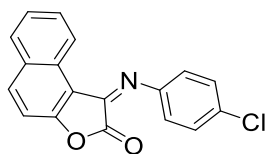
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a dark red solid in 89% yield (65.0 mg). mp 181–183 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.95 (d, *J* = 8.4 Hz, 1H), 8.04 (d, *J* = 8.8 Hz, 1H), 7.87 (d, *J* = 8.4 Hz, 1H), 7.69–7.66 (m, 1H), 7.52 (t, *J* = 7.6 Hz, 1H), 7.36 (t, *J* = 7.6 Hz, 2H), 7.28 (d, *J* = 8.8 Hz, 1H), 7.23 (d, *J* = 8.8 Hz, 2H), 7.15–7.04 (m, 5H); ¹³C NMR (101 MHz, CDCl₃): δ 158.4, 157.0, 156.2, 155.4, 146.9, 143.5, 136.9, 130.9, 130.1, 129.9, 129.2, 128.6, 126.1, 124.1, 123.5, 122.5, 119.2, 118.8, 115.2, 111.8. HRMS (EI) *m/z*: [M]⁺ calcd. for C₂₄H₁₅NO₃: 365.1052; found: 365.1056.

(Z)-1-(phenylimino)naphtho[2,1-*b*]furan-2(1*H*)-one (4h)



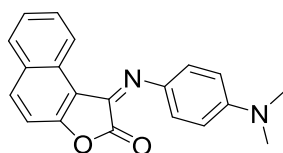
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford an orange solid in 88% yield (48.1 mg). mp 184–186 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.94 (d, *J* = 8.4 Hz, 1H), 8.09 (d, *J* = 8.8 Hz, 1H), 7.90 (d, *J* = 8.0 Hz, 1H), 7.70 (t, *J* = 7.2 Hz, 1H), 7.54 (t, *J* = 8.0 Hz, 1H), 7.43 (t, *J* = 7.2 Hz, 2H), 7.26 (d, *J* = 3.2 Hz, 1H), 7.25–7.23 (m, 1H), 7.10 (d, *J* = 7.6 Hz, 2H); ¹³C NMR (101 MHz, CDCl₃): δ 158.8, 155.1, 148.8, 147.7, 137.2, 130.9, 130.2, 129.2, 128.9, 128.6, 126.2, 125.9, 124.2, 119.2, 114.8, 111.8. HRMS (EI) *m/z*: [M]⁺ calcd. for C₁₈H₁₁NO₂: 273.0790; found: 273.0791.

(Z)-1-((4-chlorophenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4i)



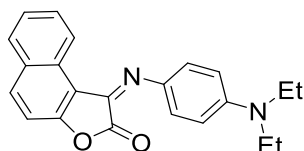
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 96% yield (59.0 mg). mp 213–214 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.89 (d, *J* = 8.4 Hz, 1H), 8.10 (d, *J* = 8.8 Hz, 1H), 7.90 (d, *J* = 8.2 Hz, 1H), 7.70 (t, *J* = 7.7 Hz, 1H), 7.54 (t, *J* = 7.6 Hz, 1H), 7.38 (d, *J* = 8.5 Hz, 2H), 7.31 (d, *J* = 8.9 Hz, 1H), 7.06 (d, *J* = 8.5 Hz, 2H); ¹³C NMR (101 MHz, CDCl₃): δ 159.0, 155.2, 148.1, 147.0, 137.6, 131.4, 130.9, 130.3, 129.2, 129.0, 128.6, 126.3, 124.1, 120.8, 114.7, 111.8. HRMS (EI) *m/z*: [M]⁺ calcd. for C₁₈H₁₀ClNO₂: 307.0400; found: 307.0404.

(Z)-1-((4-(dimethylamino)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4j)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a dark purple solid in 93% yield (58.8 mg). mp 169–171 °C; ¹H NMR (400 MHz, CDCl₃): δ 9.12 (d, *J* = 8.4 Hz, 1H), 7.91 (d, *J* = 8.8 Hz, 1H), 7.84 (d, *J* = 8.0 Hz, 1H), 7.70 (d, *J* = 8.8 Hz, 2H), 7.66–7.62 (m, 1H), 7.49–7.45 (m, 1H), 7.25 (s, 1H), 6.73 (d, *J* = 8.8 Hz, 2H), 3.08 (s, 6H); ¹³C NMR (101 MHz, CDCl₃): δ 156.9, 156.1, 150.9, 140.6, 136.9, 134.0, 131.0, 129.2, 128.9, 128.6, 127.5, 125.5, 124.2, 116.9, 111.6, 111.3, 40.3. HRMS (EI) *m/z*: [M]⁺ calcd. for C₂₀H₁₆N₂O₂: 316.1212; found: 316.1212.

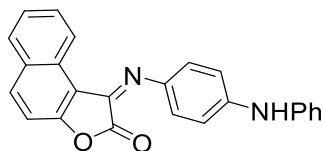
(Z)-1-((4-(diethylamino)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4k)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a dark purple solid in 89% yield (61.3 mg). mp 170–171 °C; ¹H NMR (400 MHz, CDCl₃): δ 9.14 (d, *J* = 8.4 Hz, 1H), 7.87 (d, *J* = 8.8 Hz, 1H), 7.82 (d, *J* = 8.0 Hz, 1H), 7.77 (d, *J* = 8.8 Hz, 2H), 7.62 (t, *J* = 7.6 Hz, 1H), 7.47–7.43 (m, 1H), 7.23 (d, *J* = 8.4 Hz, 1H), 6.69 (d, *J* = 8.8 Hz, 2H), 3.44 (q, *J* = 6.8 Hz, 4H), 1.25–1.21 (m, 6H); ¹³C NMR (101 MHz, CDCl₃): δ 157.3, 155.6, 148.9, 139.3,

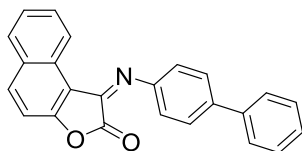
136.2, 133.5, 131.0, 129.0, 128.9, 128.8, 125.4, 124.3, 117.3, 111.6, 110.7, 44.8, 12.8. HRMS (EI) m/z : $[M]^+$ calcd. for $C_{22}H_{20}N_2O_2$: 344.1525; found: 344.1523.

(Z)-1-((4-(phenylamino)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4l)



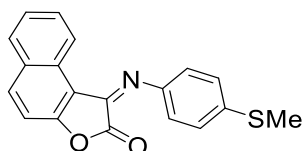
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a dark purple solid in 90% yield (65.5 mg). mp 148–150 °C; 1H NMR (400 MHz, $CDCl_3$): δ 9.03 (d, J = 8.4 Hz, 1H), 7.99 (d, J = 8.8 Hz, 1H), 7.86 (d, J = 8.0 Hz, 1H), 7.66 (t, J = 7.6 Hz, 1H), 7.52–7.48 (m, 1H), 7.44 (d, J = 8.4 Hz, 2H), 7.33 (d, J = 7.6 Hz, 1H), 7.29 (d, J = 5.6 Hz, 1H), 7.16 (d, J = 7.6 Hz, 2H), 7.09 (d, J = 8.0 Hz, 2H), 7.02–6.99 (m, 1H), 6.00 (s, 1H); ^{13}C NMR (101 MHz, $CDCl_3$): δ 157.3, 156.2, 144.0, 143.6, 141.9, 140.6, 135.6, 131.0, 129.7, 129.5, 129.1, 128.6, 125.9, 125.1, 124.2, 122.1, 119.2, 116.3, 116.1, 111.7. HRMS (EI) m/z : $[M]^+$ calcd. for $C_{24}H_{16}N_2O_2$: 364.1212; found: 364.1221.

(Z)-1-([1,1'-biphenyl]-4-ylimino)naphtho[2,1-b]furan-2(1H)-one (4m)



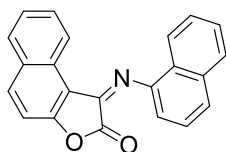
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 90% yield (62.8 mg). mp 173–175 °C; 1H NMR (400 MHz, $CDCl_3$): δ 8.96 (d, J = 8.4 Hz, 1H), 8.08 (d, J = 8.8 Hz, 1H), 7.89 (d, J = 8.2 Hz, 1H), 7.75–7.61 (m, 5H), 7.54 (t, J = 7.6 Hz, 1H), 7.45 (t, J = 7.5 Hz, 2H), 7.39–7.28 (m, 2H), 7.24 (d, J = 7.5 Hz, 2H); ^{13}C NMR (101 MHz, $CDCl_3$): δ 158.8, 155.3, 147.7, 147.5, 140.6, 139.1, 137.2, 131.0, 130.2, 129.2, 128.8, 128.6, 127.5, 127.3, 127.0, 126.2, 124.2, 120.4, 115.0, 111.8. HRMS (EI) m/z : $[M]^+$ calcd. for $C_{24}H_{15}NO_2$: 349.1103; found: 349.1092.

(Z)-1-((4-(methylthio)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4n)



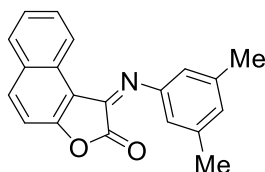
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a dark red solid in 96% yield (61.3 mg). mp 185–187 °C ; ¹H NMR (400 MHz, CDCl₃): δ 8.94 (d, *J* = 8.4 Hz, 1H), 8.07 (d, *J* = 8.8 Hz, 1H), 7.89 (d, *J* = 8.0 Hz, 1H), 7.69 (t, *J* = 7.6 Hz, 1H), 7.53 (t, *J* = 7.6 Hz, 1H), 7.32–7.29 (m, 3H), 7.17 (d, *J* = 8.2 Hz, 2H), 2.54 (s, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 158.5, 155.4, 147.0, 145.5, 134.0, 130.9, 130.1, 129.2, 128.6, 126.8, 126.2, 124.2, 121.2, 115.2, 111.8, 16.1. HRMS (EI) *m/z*: [M]⁺ calcd. for C₁₉H₁₃NO₂S: 319.0667; found: 319.0665.

(Z)-1-(naphthalen-1-ylimino)naphtho[2,1-b]furan-2(1H)-one (4o)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 88% yield (56.9 mg). mp 182–183 °C ; ¹H NMR (400 MHz, CDCl₃): δ 9.10 (d, *J* = 8.4 Hz, 1H), 8.10 (d, *J* = 8.8 Hz, 1H), 8.05 (d, *J* = 8.4 Hz, 1H), 7.93–7.88 (m, 2H), 7.77–7.71 (m, 2H), 7.58–7.46 (m, 4H), 7.32 (d, *J* = 8.8 Hz, 1H), 7.12 (d, *J* = 7.2 Hz, 1H); ¹³C NMR (101 MHz, CDCl₃): δ 158.8, 155.3, 148.3, 145.0, 137.4, 134.2, 131.0, 130.4, 129.3, 128.7, 128.2, 126.6, 126.5, 126.5, 126.3, 126.1, 125.5, 124.2, 123.4, 115.1, 113.0, 111.9. HRMS (EI) *m/z*: [M]⁺ calcd. for C₂₂H₁₃NO₂: 323.0946; found: 323.0939.

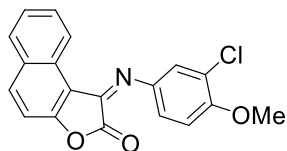
(Z)-1-((3,5-dimethylphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4p)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 96% yield (57.8 mg). mp 165–167 °C ; ¹H NMR (400 MHz, CDCl₃): δ 8.93 (d, *J* = 8.4 Hz, 1H), 8.08 (d, *J* = 8.8 Hz, 1H), 7.89 (d, *J* = 8.0 Hz, 1H), 7.68 (t, *J* = 7.6 Hz, 1H), 7.53 (t, *J* = 7.2 Hz, 1H), 7.30 (d, *J* = 8.8 Hz, 1H), 6.89 (s, 1H); 6.73 (s, 2H); 2.36 (s, 6H);

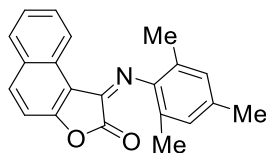
^{13}C NMR (101 MHz, CDCl_3): δ 158.6, 155.0, 148.9, 147.4, 138.5, 137.0, 130.9, 130.1, 129.1, 128.6, 127.7, 126.1, 124.2, 116.7, 114.9, 111.8, 21.4. HRMS (EI) m/z : $[\text{M}]^+$ calcd. for $\text{C}_{20}\text{H}_{15}\text{NO}_2$: 301.1103; found: 301.1095.

(Z)-1-(3-chloro-4-methoxyphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4q)



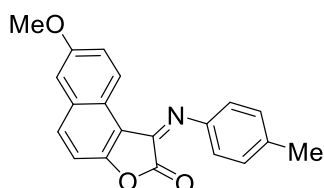
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 85% yield (57.3 mg). mp 177–178 °C ; ^1H NMR (400 MHz, CDCl_3): δ 8.93 (d, J = 6.0 Hz, 1H), 8.07 (d, J = 8.0 Hz, 1H), 7.89 (d, J = 6.4 Hz, 1H), 7.69 (s, 1H), 7.54 (s, 1H), 7.39 (s, 1H), 7.31 (d, J = 8.0 Hz, 1H), 7.19 (d, J = 6.8 Hz, 1H), 6.98 (d, J = 7.6 Hz, 1H), 3.96 (s, 3H); ^{13}C NMR (101 MHz, CDCl_3): δ 158.5, 155.5, 153.9, 147.2, 141.5, 137.1, 130.9, 130.2, 129.2, 128.6, 126.2, 124.1, 123.8, 122.6, 120.3, 115.2, 111.8, 111.7, 56.41. HRMS (EI) m/z : $[\text{M}]^+$ calcd. for $\text{C}_{19}\text{H}_{12}\text{ClNO}_3$: 337.0506; found: 337.0505.

(Z)-1-(mesitylimino)naphtho[2,1-b]furan-2(1H)-one (4r)



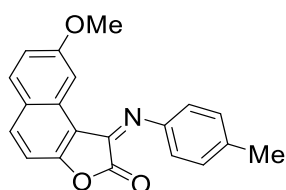
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a dark red solid in 43% yield (27.1 mg). mp 161–162 °C ; ^1H NMR (400 MHz, CDCl_3): δ 9.02 (d, J = 8.0 Hz, 1H), 8.09 (d, J = 8.8 Hz, 1H), 7.91 (d, J = 8.4 Hz, 1H), 7.72–7.69 (m, 1H), 7.57–7.53 (m, 1H), 7.32 (d, J = 8.8 Hz, 1H), 6.94 (s, 2H), 2.32 (s, 3H), 2.08 (s, 6H). ^{13}C NMR (101 MHz, CDCl_3): δ 159.0, 154.8, 149.4, 145.4, 137.1, 133.8, 131.0, 130.2, 129.2, 128.8, 126.2, 124.3, 124.3, 114.3, 112.0, 20.9, 18.2. HRMS (EI) m/z : $[\text{M}]^+$ calcd. for $\text{C}_{21}\text{H}_{17}\text{NO}_2$: 315.1259; found: 315.1259.

(Z)-7-methoxy-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (4s)



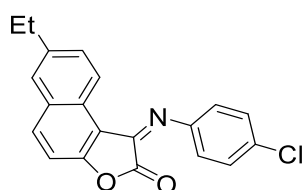
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 92% yield (58.3 mg). mp 176–178 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.84 (d, *J* = 9.2 Hz, 1H), 7.92 (d, *J* = 8.8 Hz, 1H), 7.34–7.32 (m, 1H), 7.25–7.21 (m, 3H), 7.17 (s, 1H), 7.07 (d, *J* = 8.0 Hz, 2H), 3.93 (s, 3H), 2.39 (s, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 157.6, 156.9, 147.3, 146.0, 136.2, 135.3, 132.3, 129.4, 125.5, 123.8, 122.3, 112.0, 115.3, 112.1, 107.5, 99.4, 55.4, 21.2. HRMS (EI) *m/z*: [M]⁺ calcd. for C₂₀H₁₅NO₃: 317.1052; found: 317.1054.

(*Z*)-8-methoxy-1-(*p*-tolylimino)naphtho[2,1-*b*]furan-2(1*H*)-one (4t)



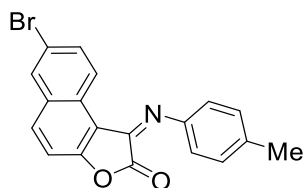
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 92% yield (58.3 mg). mp 183–185 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.34 (s, 1H), 7.96 (d, *J* = 8.8 Hz, 1H), 7.76 (d, *J* = 9.2 Hz, 1H), 7.23 (d, *J* = 7.6 Hz, 2H), 7.16–7.06 (m, 4H), 3.94 (s, 3H), 2.40 (s, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 161.6, 158.8, 155.5, 147.3, 146.2, 136.4, 136.0, 130.7, 130.5, 129.4, 126.4, 112.0, 118.7, 114.2, 108.8, 102.8, 55.6, 21.2. HRMS (EI) *m/z*: [M]⁺ calcd. for C₂₀H₁₅NO₃: 317.1052; found: 317.1058.

(*Z*)-1-((4-chlorophenyl)imino)-7-ethylnaphtho[2,1-*b*]furan-2(1*H*)-one (4u)



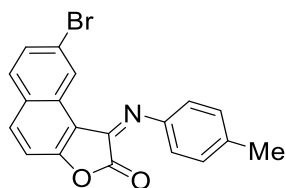
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 86% yield (57.6 mg). mp 177–179 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.79 (d, *J* = 8.5 Hz, 1H), 8.03 (d, *J* = 8.8 Hz, 1H), 7.68 (s, 1H), 7.56 (d, *J* = 8.6 Hz, 1H), 7.38 (d, *J* = 8.5 Hz, 2H), 7.28 (s, 1H), 7.05 (d, *J* = 8.5 Hz, 2H), 2.83 (q, *J* = 7.5 Hz, 2H), 1.34 (t, *J* = 7.6 Hz, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 158.5, 155.3, 148.2, 147.1, 142.3, 137.1, 131.5, 131.3, 128.9, 127.0, 126.8, 123.9, 120.8, 114.6, 111.6, 28.9, 15.4. HRMS (EI) *m/z*: [M]⁺ calcd. for C₂₀H₁₄ClNO₂: 335.0713; found: 335.0711.

(Z)-7-bromo-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (4v)



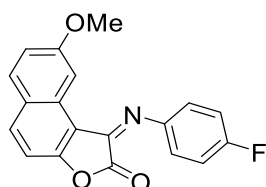
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 71% yield (51.8 mg). mp 216–218 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.84 (d, *J* = 8.8 Hz, 1H), 8.04 (s, 1H), 7.96 (d, *J* = 8.8 Hz, 1H), 7.73 (d, *J* = 8.8 Hz, 1H), 7.32 (d, *J* = 8.8 Hz, 1H), 7.24 (d, *J* = 8.0 Hz, 2H), 7.10 (d, *J* = 8.0 Hz, 2H), 2.40 (s, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 158.3, 154.9, 146.7, 145.6, 136.8, 135.6, 133.2, 132.0, 131.0, 129.4, 127.0, 125.9, 120.2, 112.0, 115.4, 113.0, 21.2. HRMS (EI) *m/z*: [M]⁺ calcd. for C₁₉H₁₂BrNO₂: 365.0051; found: 365.0044.

(Z)-8-bromo-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (4w)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 70% yield (51.1 mg). mp 225–227 °C; ¹H NMR (400 MHz, CDCl₃): δ 9.11 (s, 1H), 7.99 (d, *J* = 8.8 Hz, 1H), 7.71 (d, *J* = 8.8 Hz, 1H), 7.59 (d, *J* = 8.4 Hz, 1H), 7.29 (d, *J* = 8.8 Hz, 1H), 7.24 (d, *J* = 8.8 Hz, 2H), 7.11 (d, *J* = 8.8 Hz, 2H), 2.40 (s, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 158.7, 154.9, 146.4, 145.7, 136.8, 136.5, 130.5, 129.7, 129.4, 129.3, 126.5, 125.3, 120.2, 114.4, 112.2, 21.2. HRMS (EI) *m/z*: [M]⁺ calcd. for C₁₉H₁₂BrNO₂: 365.0051; found: 365.0059.

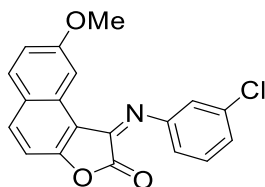
(Z)-1-((4-fluorophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4x)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford an orange solid in 87% yield (55.9 mg). mp 230–232 °C; ¹H NMR (400 MHz, DMSO-*d*₆): δ 8.24–8.20 (m, 2H), 8.00 (d, *J* = 8.8 Hz, 1H), 7.35–7.23 (m, 6H),

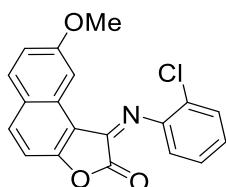
3.89 (s, 3H); ^{13}C NMR (101 MHz, DMSO- d_6): δ 161.5, 160.6 (d, $J = 242.7$ Hz), 159.3, 155.8, 149.4, 145.4 (d, $J = 2.8$ Hz), 137.5, 131.8, 130.3, 126.3, 122.6 (d, $J = 8.4$ Hz), 118.3, 115.7 (d, $J = 22.6$ Hz), 113.9, 109.6, 102.9, 55.9. HRMS (EI) m/z : $[\text{M}]^+$ calcd. for $\text{C}_{19}\text{H}_{12}\text{FNO}_3$: 321.0801; found: 321.0793.

(Z)-1-((3-chlorophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4y)



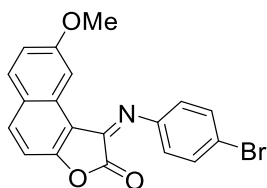
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 72% yield (48.5 mg). mp 182–184 °C; ^1H NMR (400 MHz, CDCl_3): δ 8.22 (s, 1H), 7.99 (d, $J = 8.6$ Hz, 1H), 7.76 (d, $J = 9.0$ Hz, 1H), 7.33 (t, $J = 7.9$ Hz, 1H), 7.22 – 7.03 (m, 4H), 6.93 (d, $J = 7.8$ Hz, 1H), 3.94 (s, 3H); ^{13}C NMR (101 MHz, CDCl_3): δ 161.8, 159.5, 155.3, 150.2, 148.6, 137.4, 134.5, 130.8, 130.5, 129.8, 126.3, 125.2, 119.3, 118.9, 117.1, 113.5, 108.8, 102.7, 55.6. HRMS (EI) m/z : $[\text{M}]^+$ calcd. for $\text{C}_{19}\text{H}_{12}\text{ClNO}_3$: 337.0506; found: 337.0506.

(Z)-1-((2-chlorophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4z)



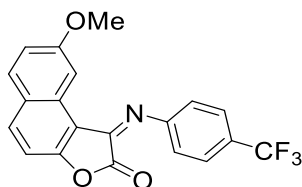
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 75% yield (50.6 mg). mp 181–182 °C; ^1H NMR (400 MHz, CDCl_3): δ 8.37 (s, 1H), 8.00 (d, $J = 8.7$ Hz, 1H), 7.77 (d, $J = 9.0$ Hz, 1H), 7.49 (d, $J = 7.9$ Hz, 1H), 7.30 (t, $J = 7.5$ Hz, 1H), 7.15 (dd, $J = 12.6, 8.7$ Hz, 3H), 7.07 (d, $J = 7.7$ Hz, 1H), 3.96 (s, 3H); ^{13}C NMR (101 MHz, CDCl_3): δ 161.9, 159.8, 155.5, 149.4, 146.2, 137.5, 130.8, 130.7, 129.9, 127.1, 126.4, 126.3, 124.4, 119.6, 119.0, 113.6, 108.8, 102.9, 55.7. HRMS (EI) m/z : $[\text{M}]^+$ calcd. for $\text{C}_{19}\text{H}_{12}\text{ClNO}_3$: 337.0506; found: 337.0505.

(Z)-1-((4-bromophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4za)



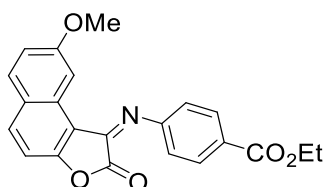
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a red solid in 73% yield (55.6 mg). mp 233–235 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.25 (s, 1H), 8.00 (d, *J* = 8.8 Hz, 1H), 7.78 (d, *J* = 8.8 Hz, 1H), 7.53 (d, *J* = 8.0 Hz, 2H), 7.14 (t, *J* = 8.8 Hz, 2H), 6.98 (d, *J* = 8.4 Hz, 2H), 3.95 (s, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 161.8, 159.4, 155.5, 148.2, 147.8, 137.2, 131.9, 130.8, 130.5, 126.4, 121.2, 119.0, 118.8, 113.8, 108.8, 102.8, 55.6. HRMS (EI) *m/z*: [M]⁺ calcd. for C₁₉H₁₂BrNO₃: 381.0001; found: 381.0002.

(Z)-8-methoxy-1-((4-(trifluoromethyl)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4zb)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford an orange solid in 90% yield (66.8 mg). mp 173–175 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.21 (s, 1H), 8.01 (d, *J* = 8.4 Hz, 1H), 7.78 (d, *J* = 8.8 Hz, 1H), 7.67 (d, *J* = 8.0 Hz, 2H), 7.18–7.10 (m, 4H), 3.94 (s, 3H); ¹³C NMR (101 MHz, CDCl₃): δ 162.0, 159.8, 155.5, 152.2, 148.7, 137.7, 130.9, 130.5, 128.5 (q, *J* = 340.5 Hz), 127.1 (q, *J* = 32.7 Hz), 126.4, 126.2 (q, *J* = 3.9 Hz), 118.9, 118.8, 113.4, 108.8, 102.8, 55.7. HRMS (EI) *m/z*: [M]⁺ calcd. for C₂₀H₁₂F₃NO₃: 371.0769; found: 371.0768.

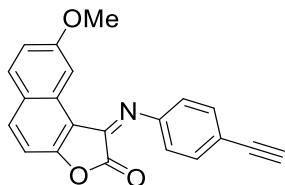
ethyl (Z)-4-((8-methoxy-2-oxonaphtho[2,1-b]furan-1(2H)-ylidene)amino)benzoate (4zc)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a dark red solid in 83% yield (62.3 mg). mp 221–223 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.22 (s, 1H), 8.11 (d, *J* = 8.4 Hz, 2H), 8.01 (d, *J* = 8.8 Hz, 1H), 7.78 (d, *J* = 8.8 Hz, 1H), 7.18–7.12 (m, 2H), 7.06 (d, *J* = 8.4 Hz, 2H), 4.39 (q, *J* = 7.2 Hz, 2H), 3.95 (s, 3H), 1.41 (t, *J* = 7.2

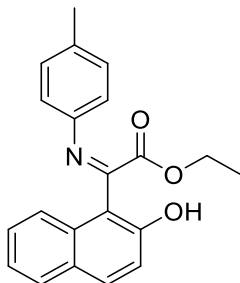
Hz, 3H); ^{13}C NMR (101 MHz, CDCl_3): δ 166.3, 161.9, 159.7, 153.5, 148.4, 137.6, 130.9, 130.6, 130.5, 127.1, 126.4, 118.9, 118.4, 113.4, 108.8, 102.8, 60.9, 55.7, 14.4. HRMS (EI) m/z : $[\text{M}]^+$ calcd. for $\text{C}_{22}\text{H}_{17}\text{NO}_5$: 375.1107; found: 375.1106.

(Z)-1-((4-ethynylphenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4zd)



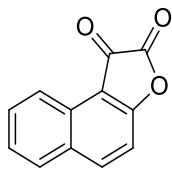
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a pale yellow solid in 82% yield (53.6 mg). mp 181–182 °C; ^1H NMR (400 MHz, CDCl_3): δ 8.23 (s, 1H), 7.98 (d, $J = 8.4$ Hz, 1H), 7.76 (d, $J = 8.8$ Hz, 1H), 7.54 (d, $J = 7.6$ Hz, 2H), 7.16–7.10 (m, 2H), 7.04 (d, $J = 8.0$ Hz, 2H), 3.94 (s, 3H), 3.10 (s, 1H); ^{13}C NMR (101 MHz, CDCl_3): δ 161.8, 159.4, 155.5, 149.3, 148.1, 137.3, 132.8, 130.8, 130.5, 126.3, 119.4, 119.2, 118.8, 113.7, 108.8, 102.8, 83.7, 77.27, 55.67. HRMS (EI) m/z : $[\text{M}]^+$ calcd. for $\text{C}_{21}\text{H}_{13}\text{NO}_3$: 327.0895; found: 327.0891.

ethyl (Z)-2-(2-hydroxynaphthalen-1-yl)-2-(p-tolylimino)acetate (5a)



The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a pale yellow oil in 46% yield (30.6 mg). ^1H NMR (400 MHz, CDCl_3): 7.86 (d, $J = 9.0$ Hz, 1H), 7.78 – 7.75 (m, 2H), 7.44 – 7.39 (m, 1H), 7.36 – 7.31 (m, 1H), 7.21 (d, $J = 9.0$ Hz, 1H), 7.18 (d, $J = 8.1$ Hz, 2H), 6.97 (d, $J = 8.2$ Hz, 2H), 4.16 (d, $J = 7.1$ Hz, 2H), 2.37 (s, 3H), 1.06 (t, $J = 7.1$ Hz, 3H); ^{13}C NMR (101 MHz, CDCl_3): δ 164.2, 164.1, 162.4, 143.6, 135.6, 135.6, 131.8, 129.5, 129.4, 128.2, 127.6, 123.5, 122.1, 121.0, 120.3, 108.97, 62.0, 21.0, 13.5. HRMS (EI) m/z : $[\text{M}]^+$ calcd. for $\text{C}_{21}\text{H}_{19}\text{NO}_3$: 333.1365; found: 333.1363.

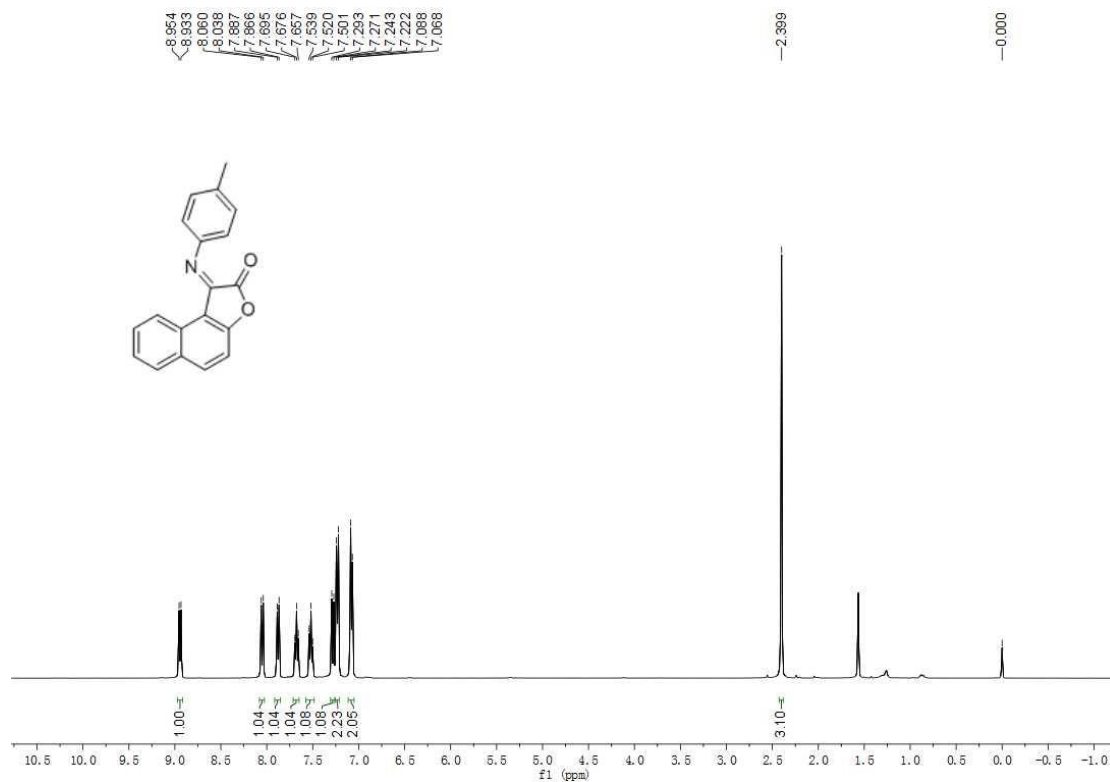
naphtho[2,1-b]furan-1,2-dione (6a)



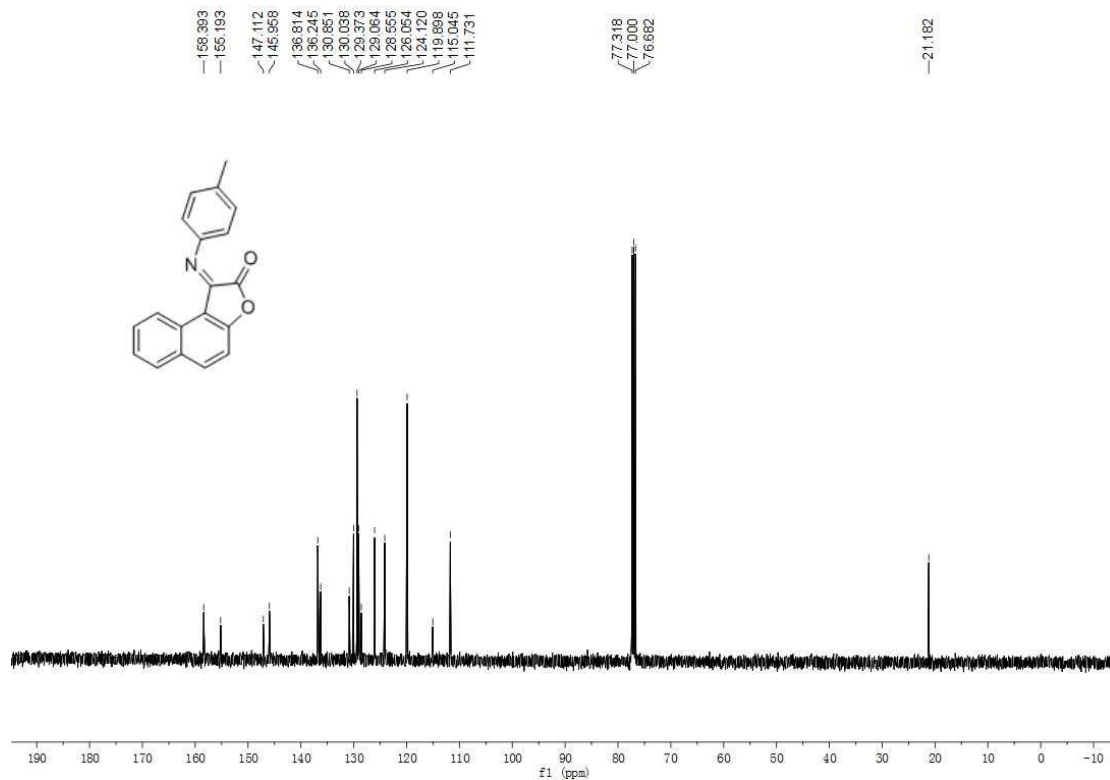
The title compound was prepared according to the general procedure and purified by column chromatography on silica gel and eluted with petroleum ether/ethyl acetate (20/1) to afford a pale yellow oil in 76% yield (30.1 mg). ^1H NMR (400 MHz, CDCl_3): δ 8.64 (d, $J = 8.3$ Hz, 1H), 8.30 (d, $J = 8.9$ Hz, 1H), 7.90 (d, $J = 8.2$ Hz, 1H), 7.77 (t, $J = 7.7$ Hz, 1H), 7.58 (t, $J = 7.6$ Hz, 1H), 7.36 (d, $J = 8.9$ Hz, 1H); ^{13}C NMR (101 MHz, CDCl_3): δ 176.1, 166.8, 156.1, 143.1, 131.8, 130.7, 129.3, 128.4, 127.1, 123.8, 112.7, 112.7. HRMS (EI) m/z : $[\text{M}]^+$ calcd. for $\text{C}_{12}\text{H}_6\text{O}_3$: 198.0317; found: 198.0316.

8. Copies of ^1H and ^{13}C NMR Spectra of the Products

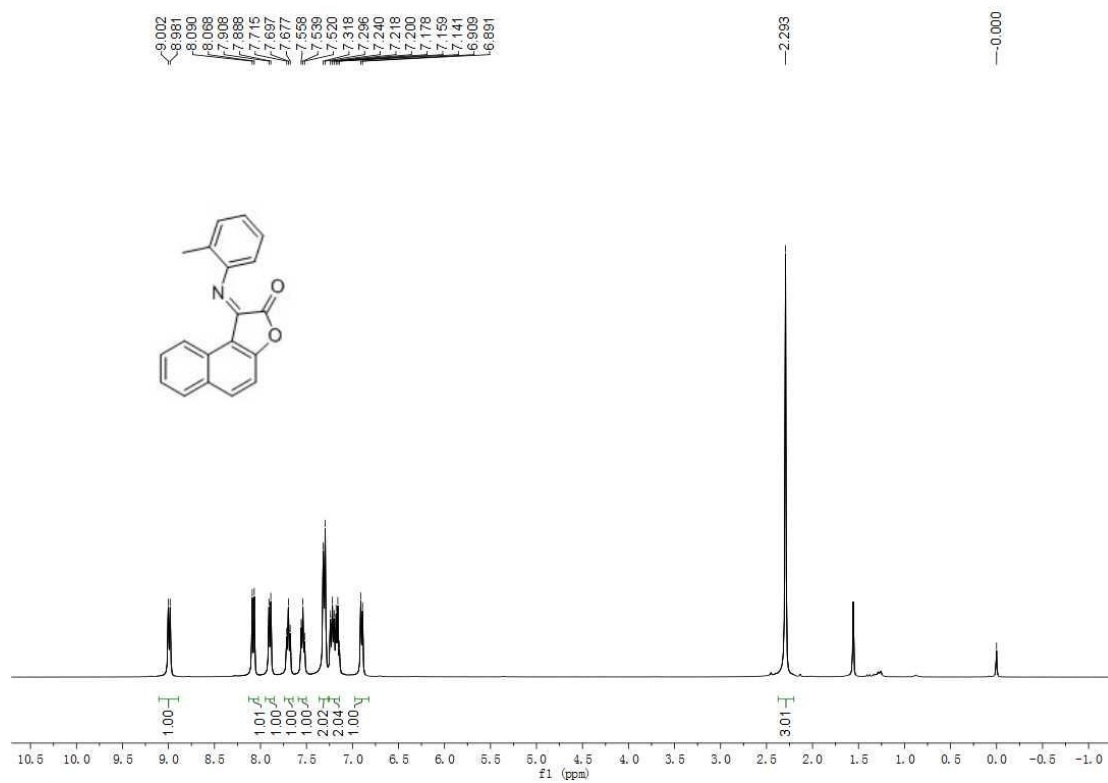
^1H NMR Spectrum of (Z)-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (4a)



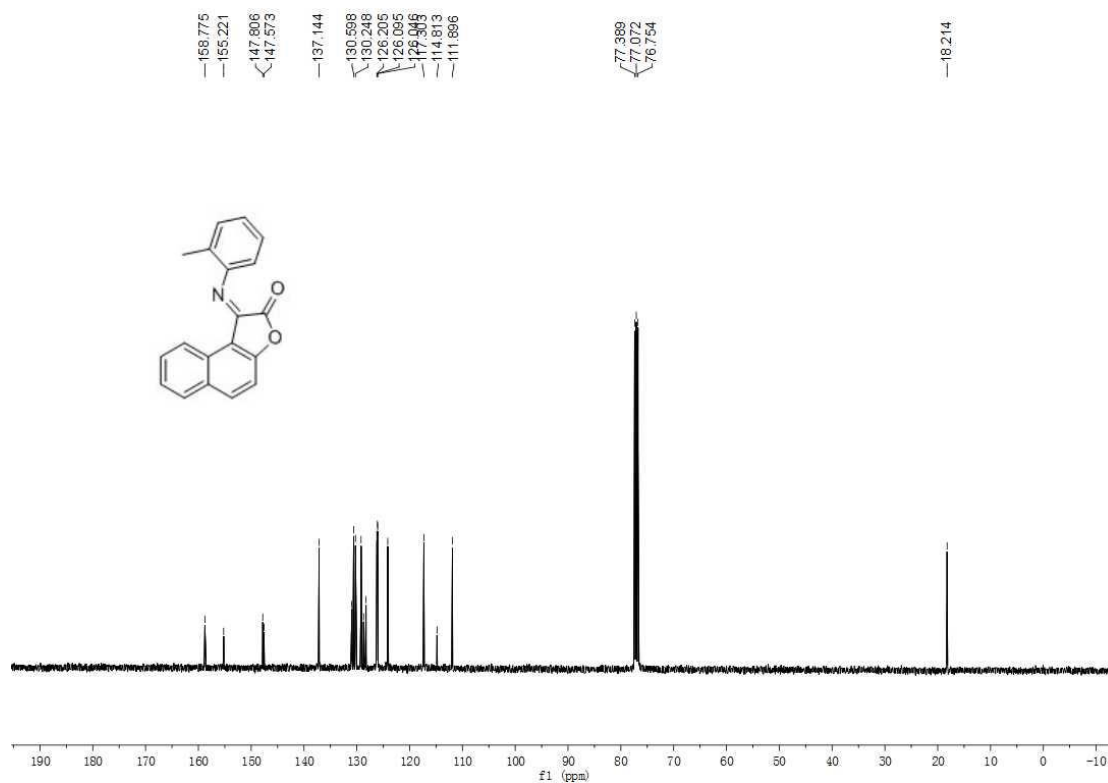
^{13}C NMR Spectrum of (Z)-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (4a)



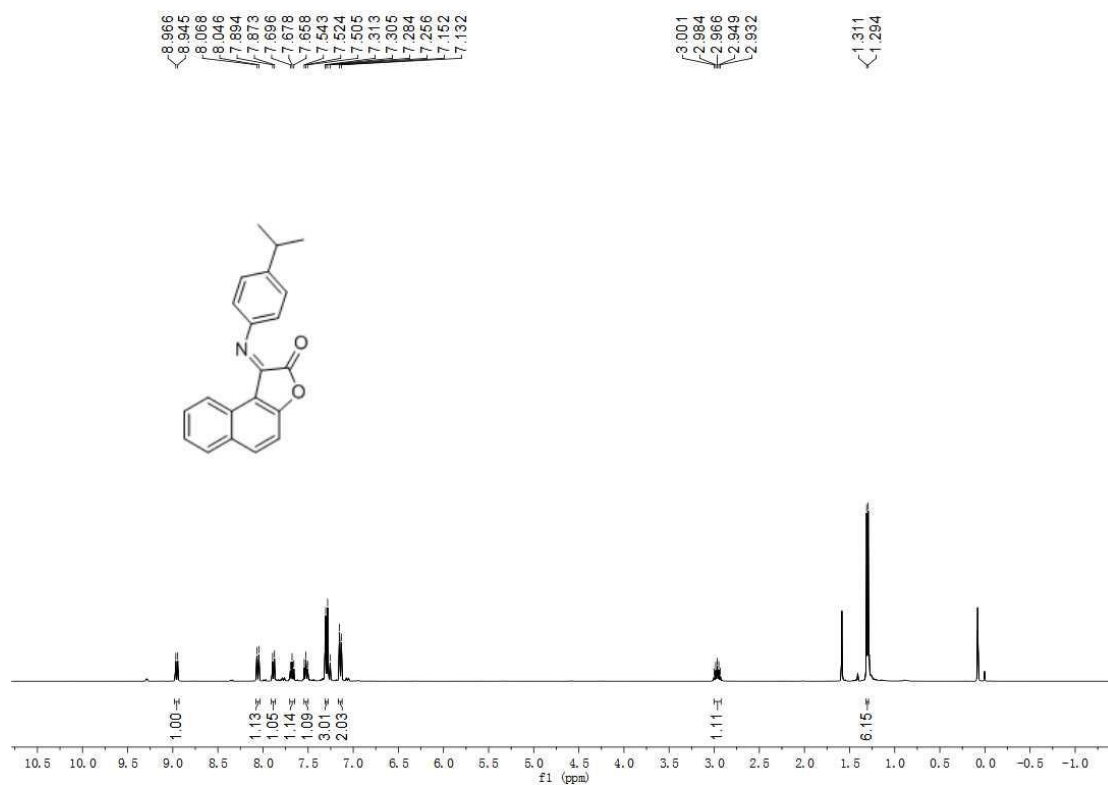
¹H NMR Spectrum of (Z)-1-(*o*-tolylimino)naphtho[2,1-*b*]furan-2(1*H*)-one (4b)



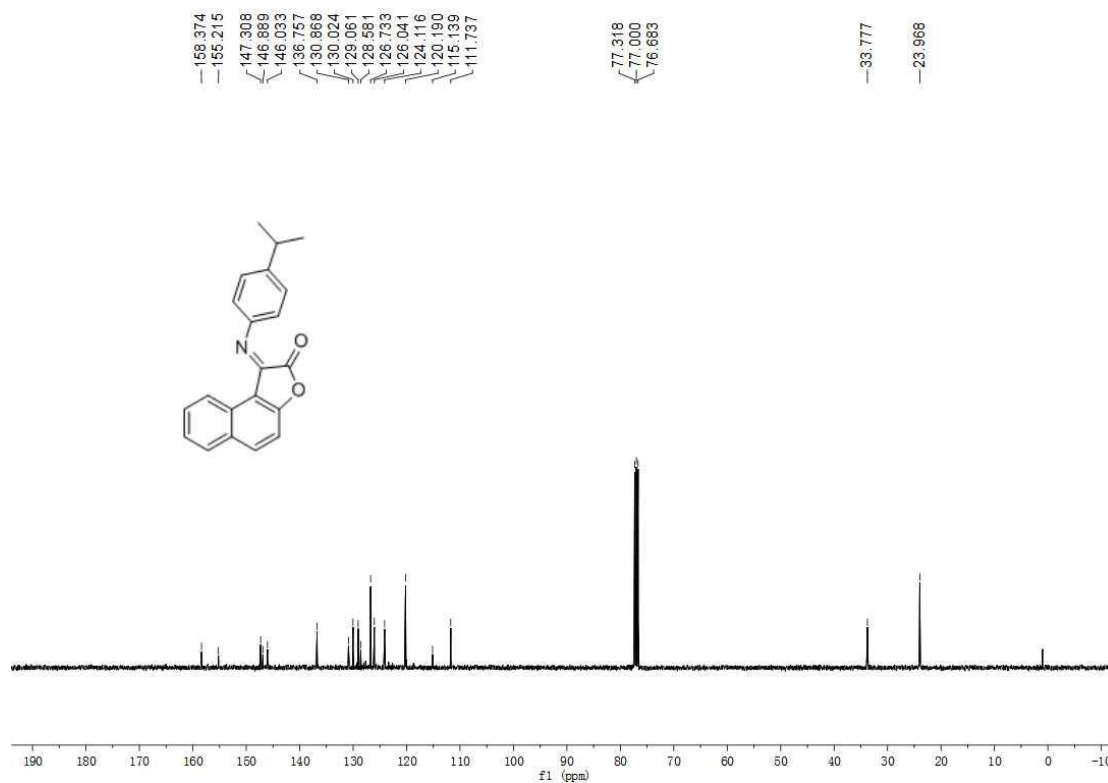
¹³C NMR Spectrum of (Z)-1-(*o*-tolylimino)naphtho[2,1-*b*]furan-2(1*H*)-one (4b)



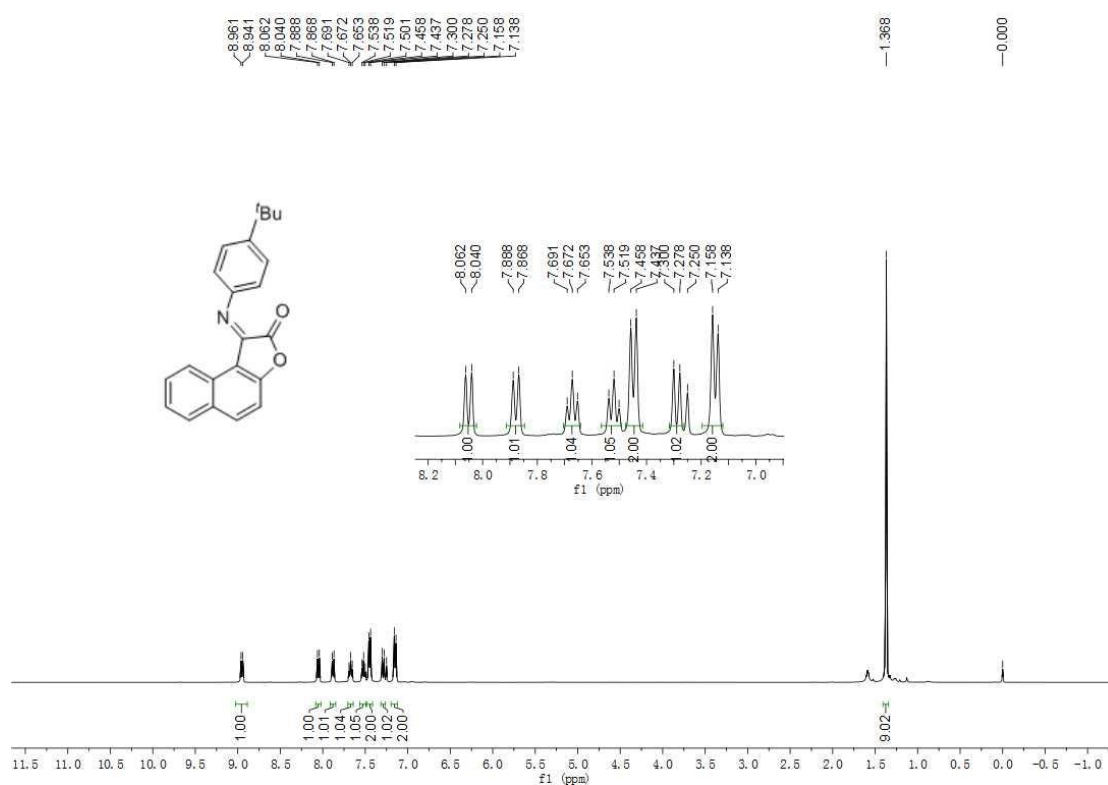
¹H NMR Spectrum of (Z)-1-((4-isopropylphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4c)



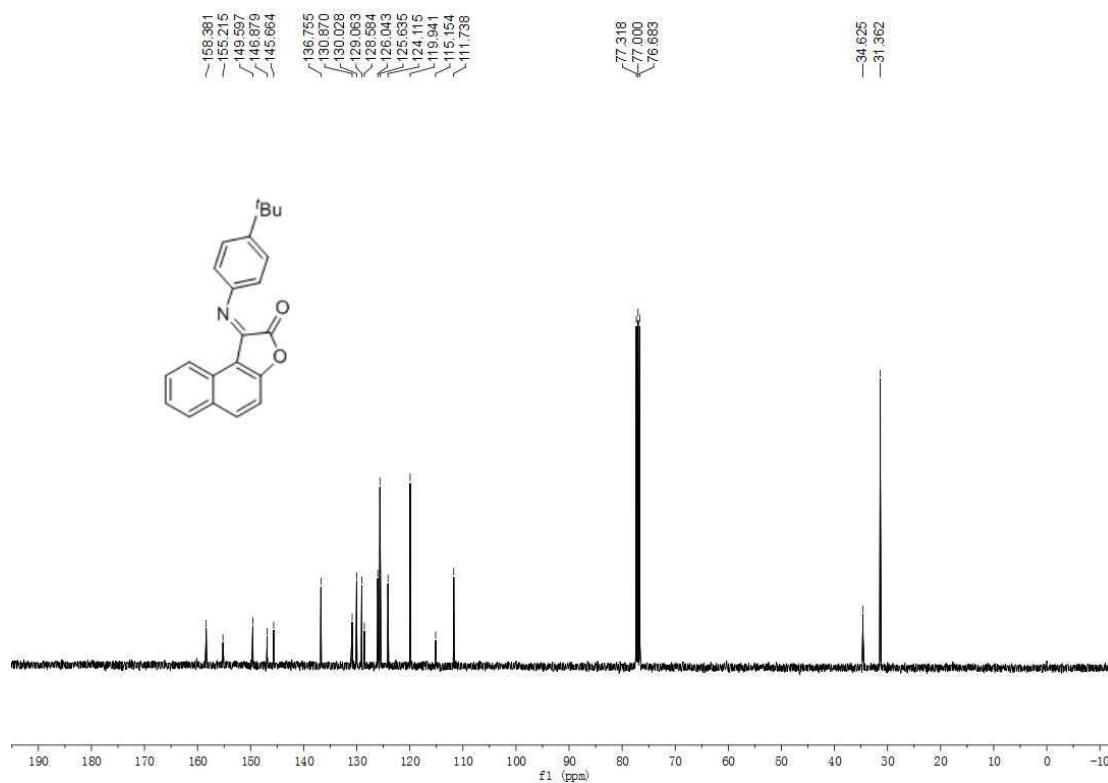
¹³C NMR Spectrum of (Z)-1-((4-isopropylphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4c)



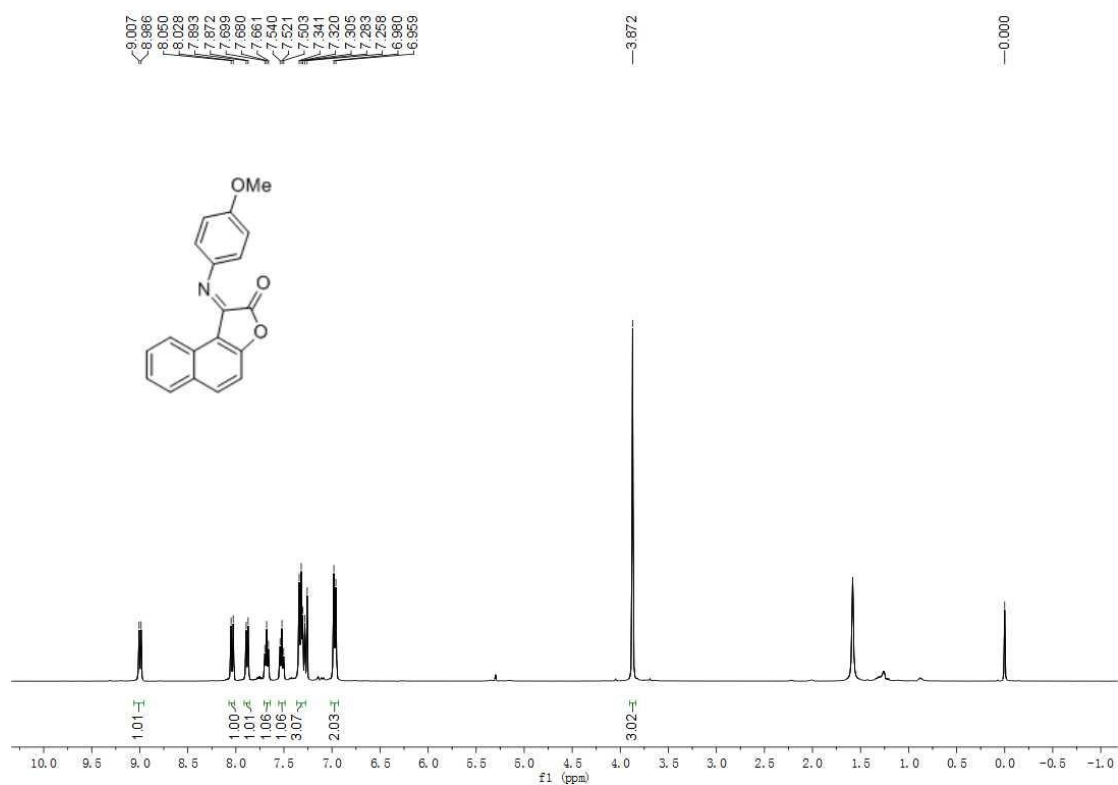
¹H NMR Spectrum of (Z)-1-((4-(*tert*-butyl)phenyl)imino)naphtho[2,1-*b*]furan-2(1*H*)-one (**4d**)



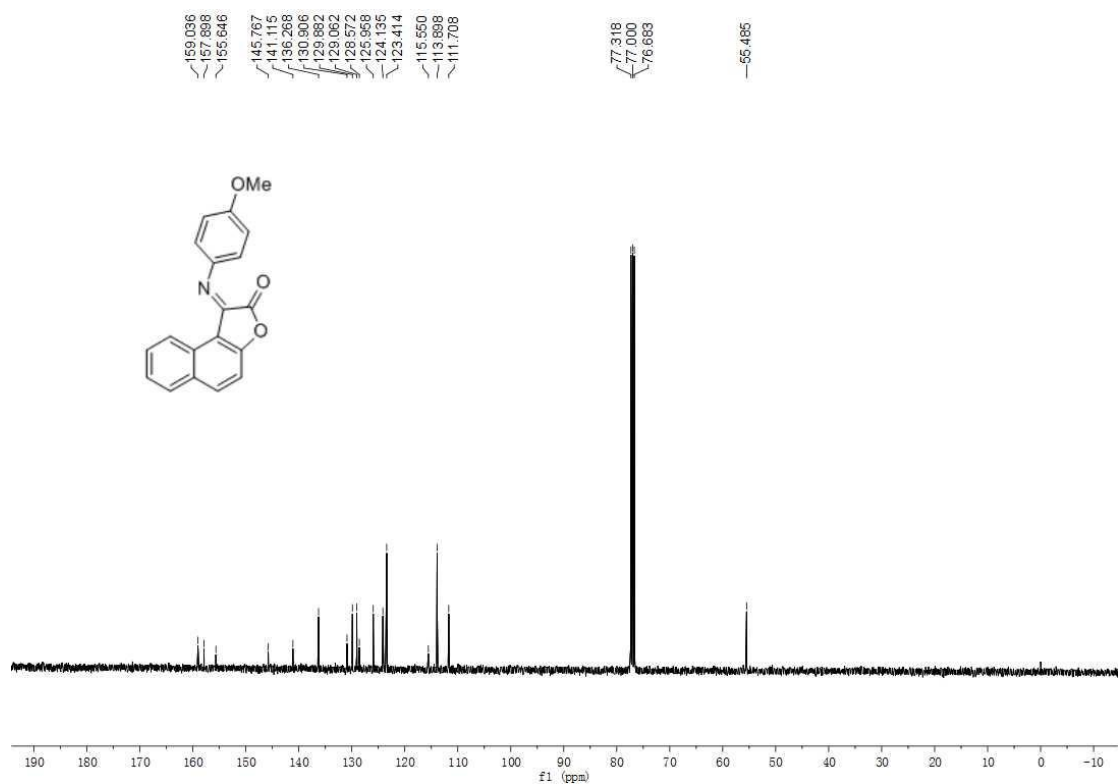
¹³C NMR Spectrum of (Z)-1-((4-(*tert*-butyl)phenyl)imino)naphtho[2,1-*b*]furan-2(1*H*)-one (**4d**)



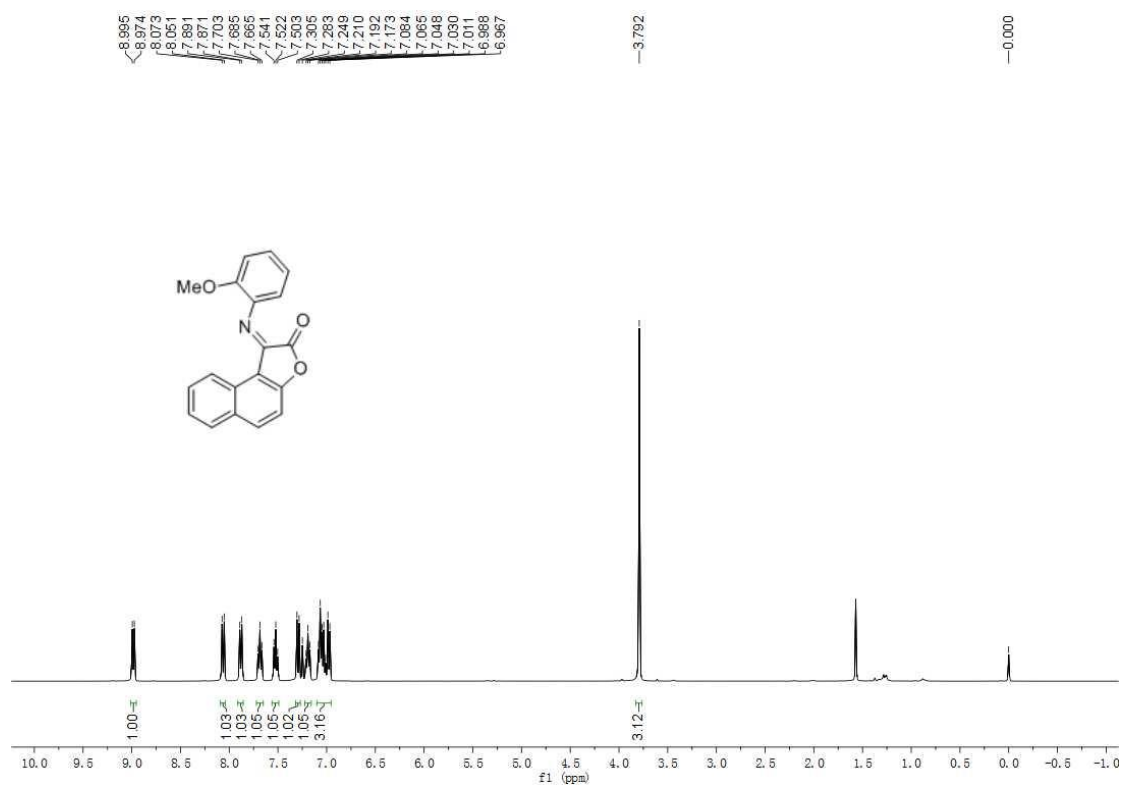
¹H NMR Spectrum of (Z)-1-((4-methoxyphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4e**)



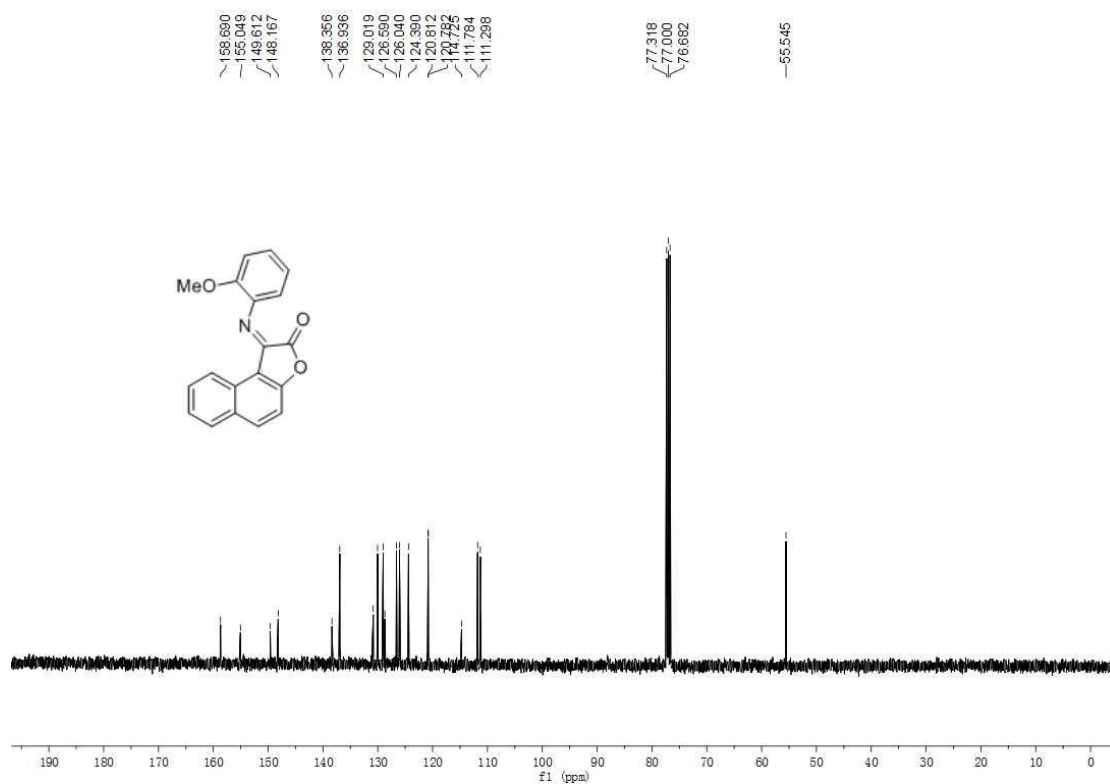
¹³C NMR Spectrum of (Z)-1-((4-methoxyphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4e**)



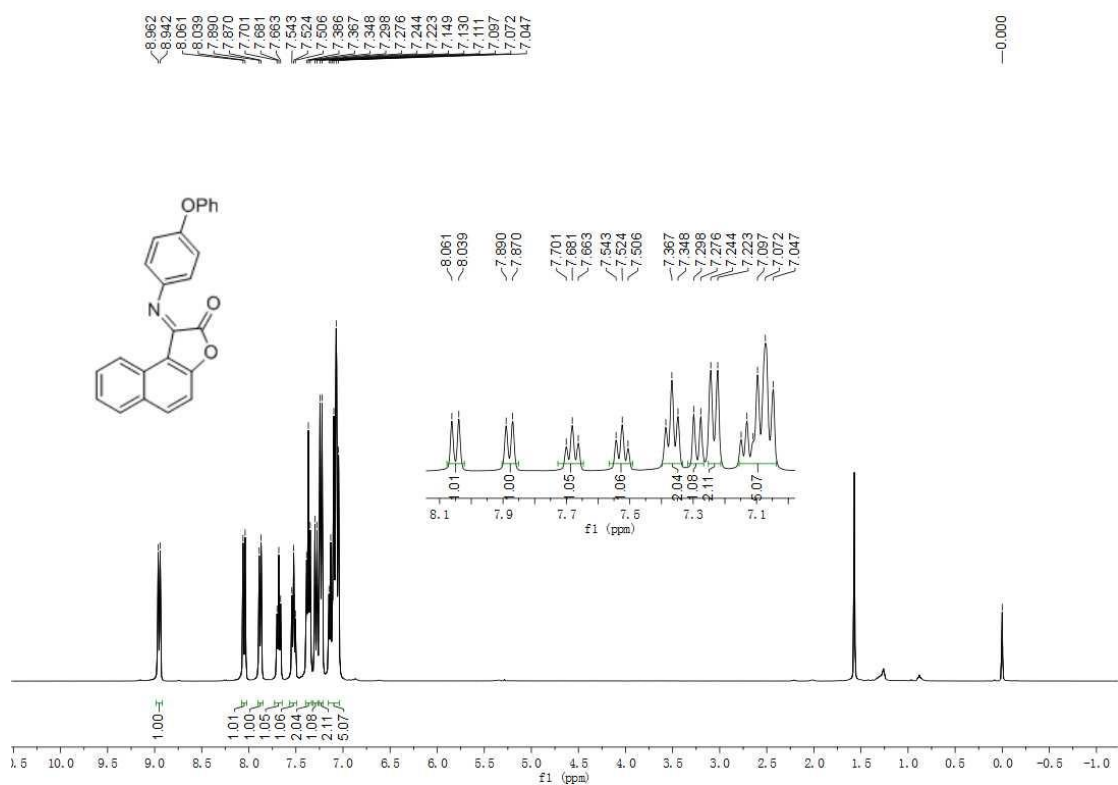
¹H NMR Spectrum of (Z)-1-((2-methoxyphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4f**)



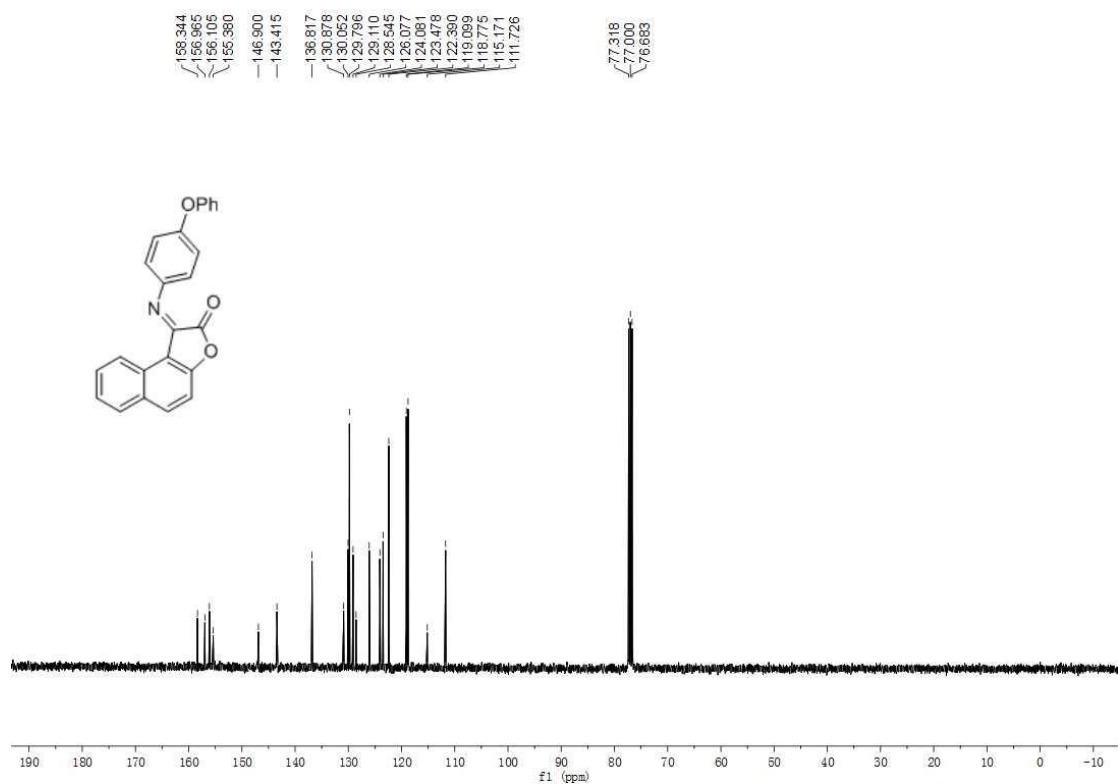
¹³C NMR Spectrum of (Z)-1-((2-methoxyphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4f**)



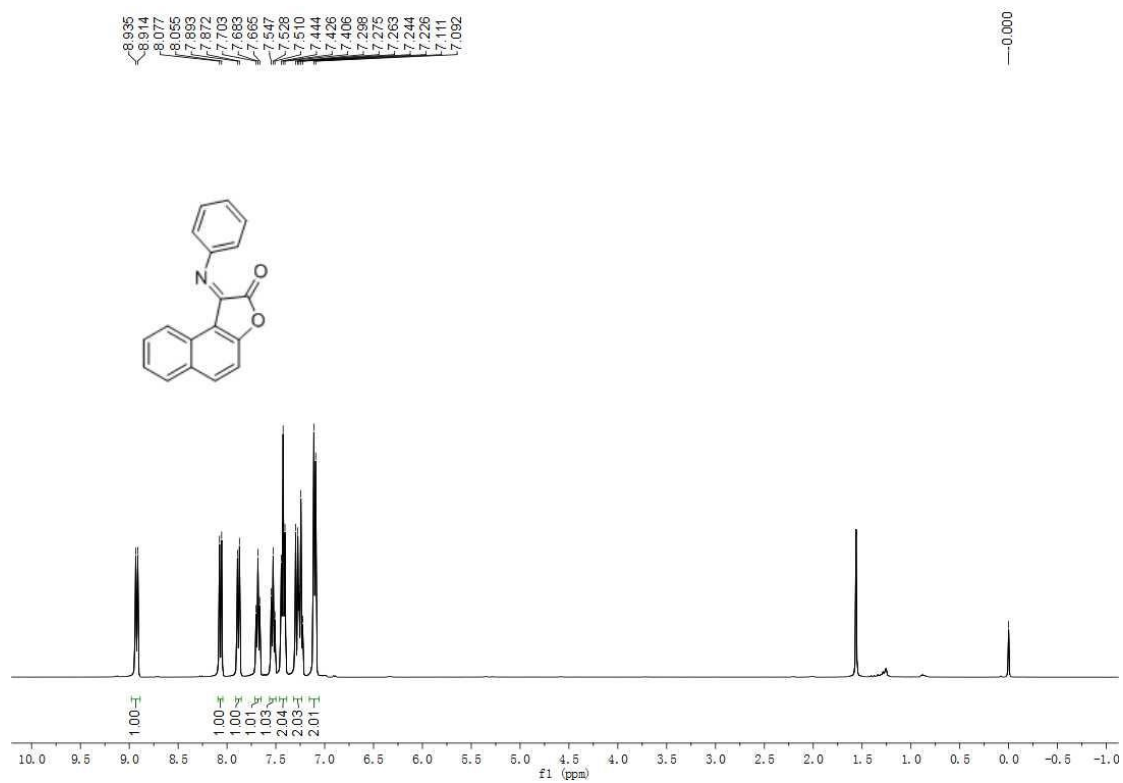
¹H NMR Spectrum of (Z)-1-((4-phenoxyphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4g**)



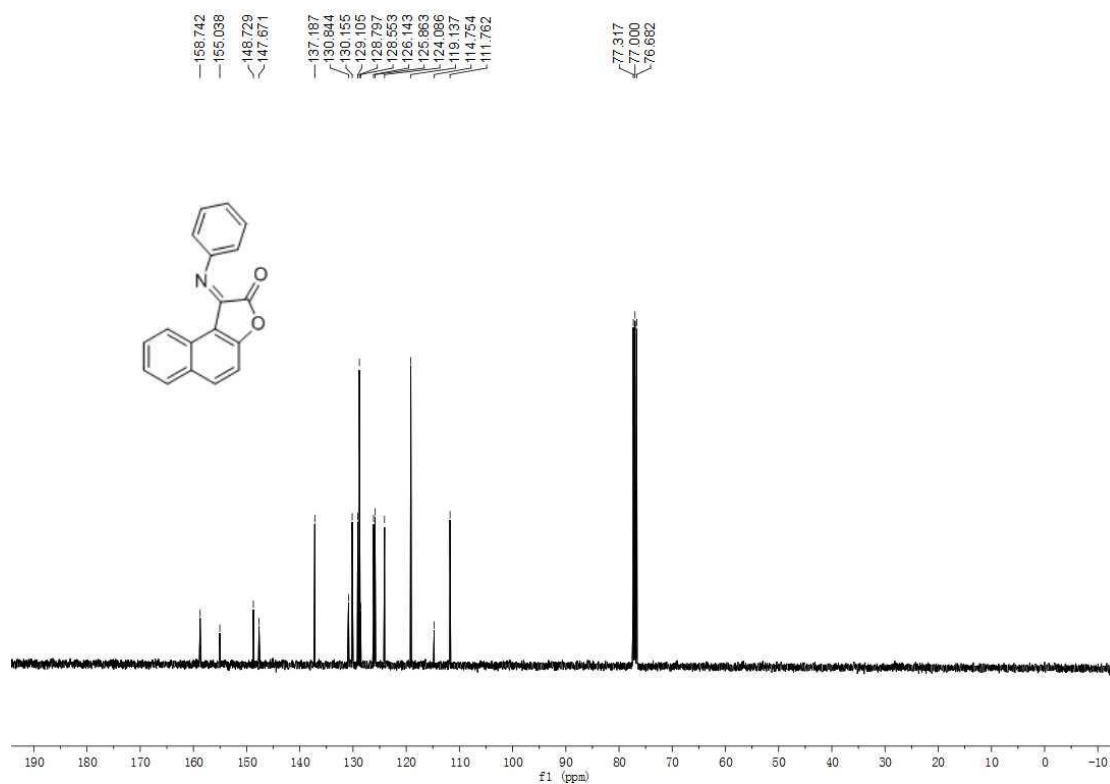
¹³C NMR Spectrum of (Z)-1-((4-phenoxyphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4g**)



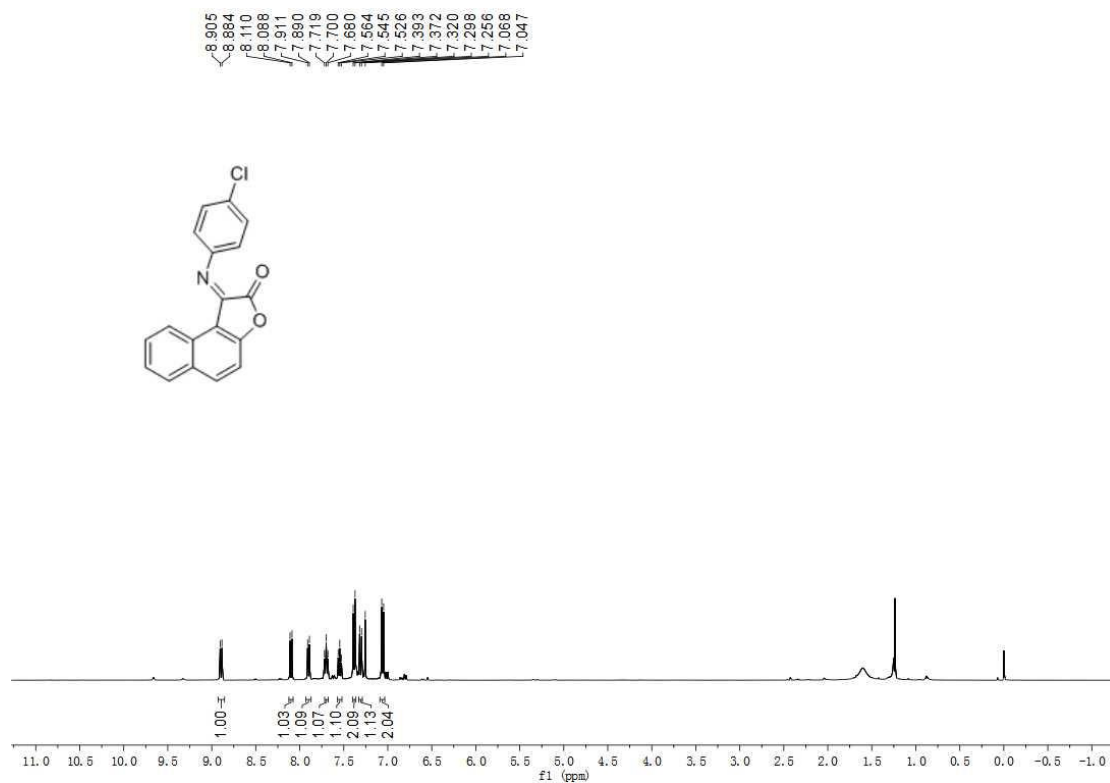
¹H NMR Spectrum of (Z)-1-(phenylimino)naphtho[2,1-b]furan-2(1H)-one (**4h**)



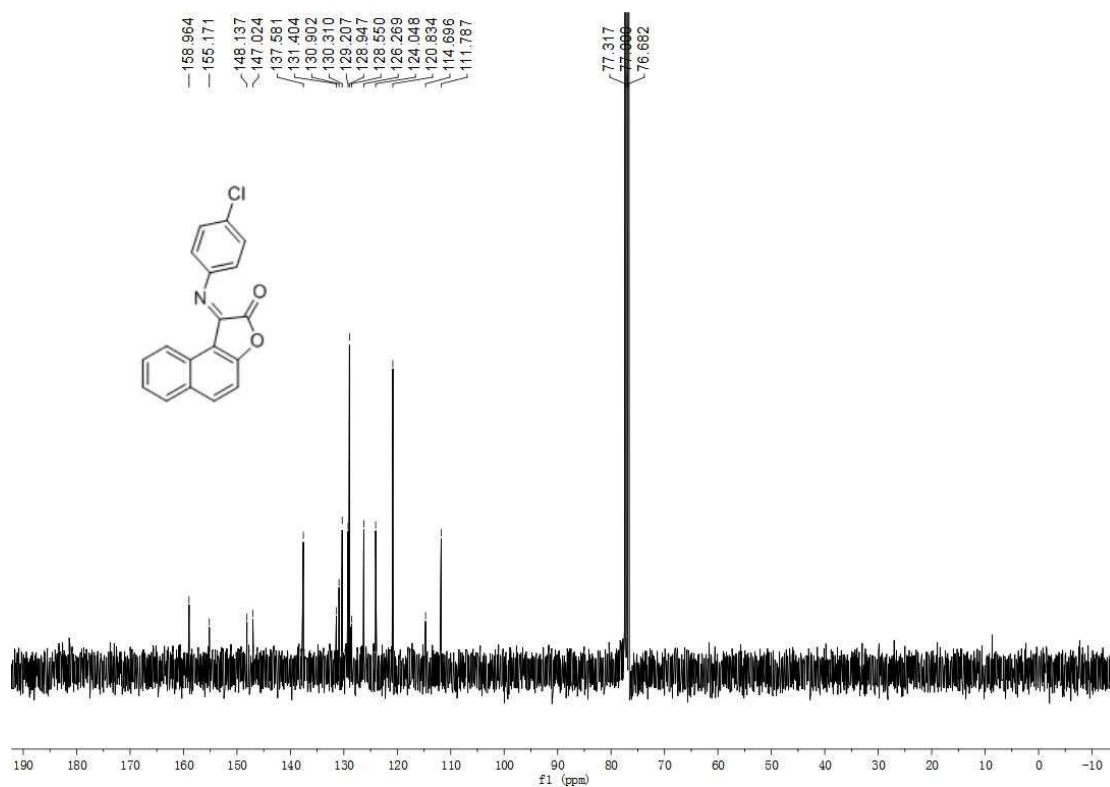
¹³C NMR Spectrum of (Z)-1-(phenylimino)naphtho[2,1-b]furan-2(1H)-one (**4h**)



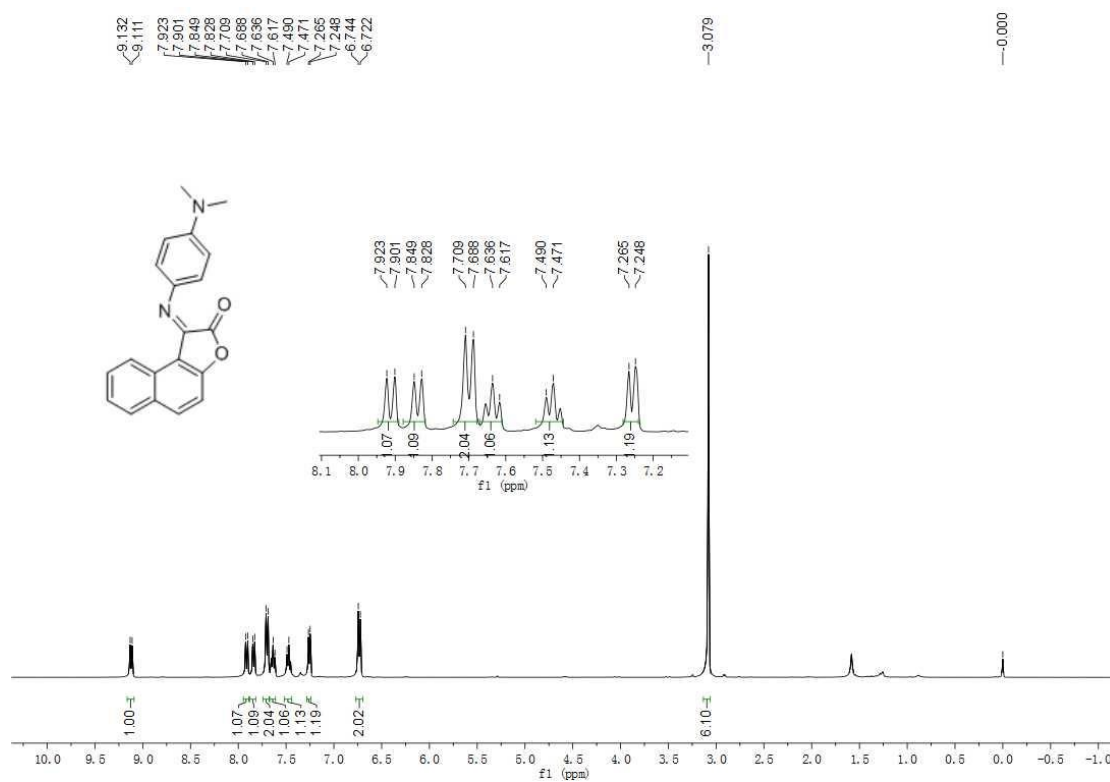
¹H NMR Spectrum of (Z)-1-((4-chlorophenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4i**)



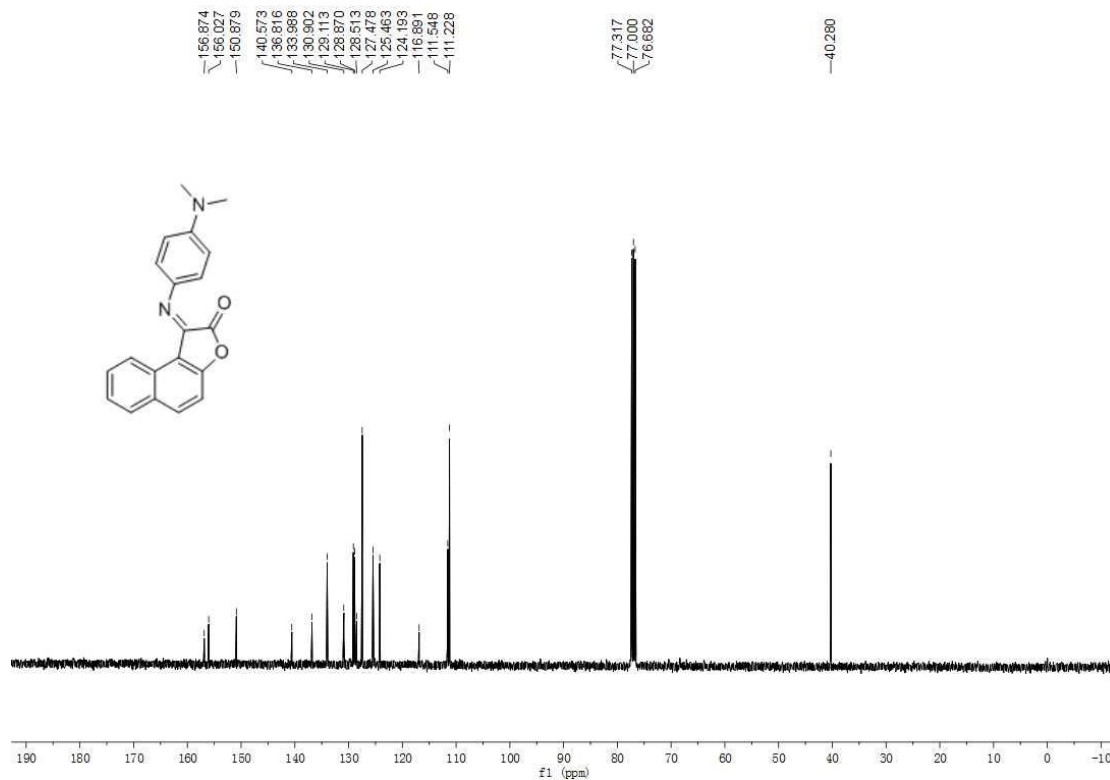
¹³C NMR Spectrum of (Z)-1-((4-chlorophenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4i**)



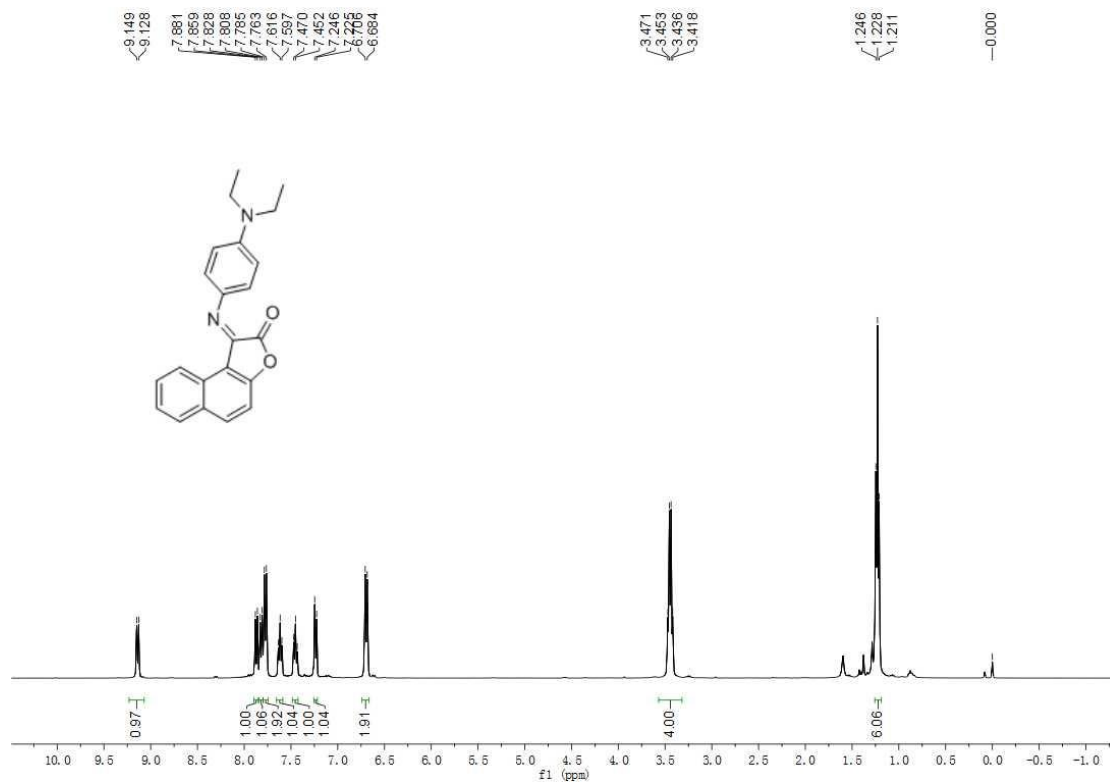
¹H NMR Spectrum of (Z)-1-((4-(dimethylamino)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4j)



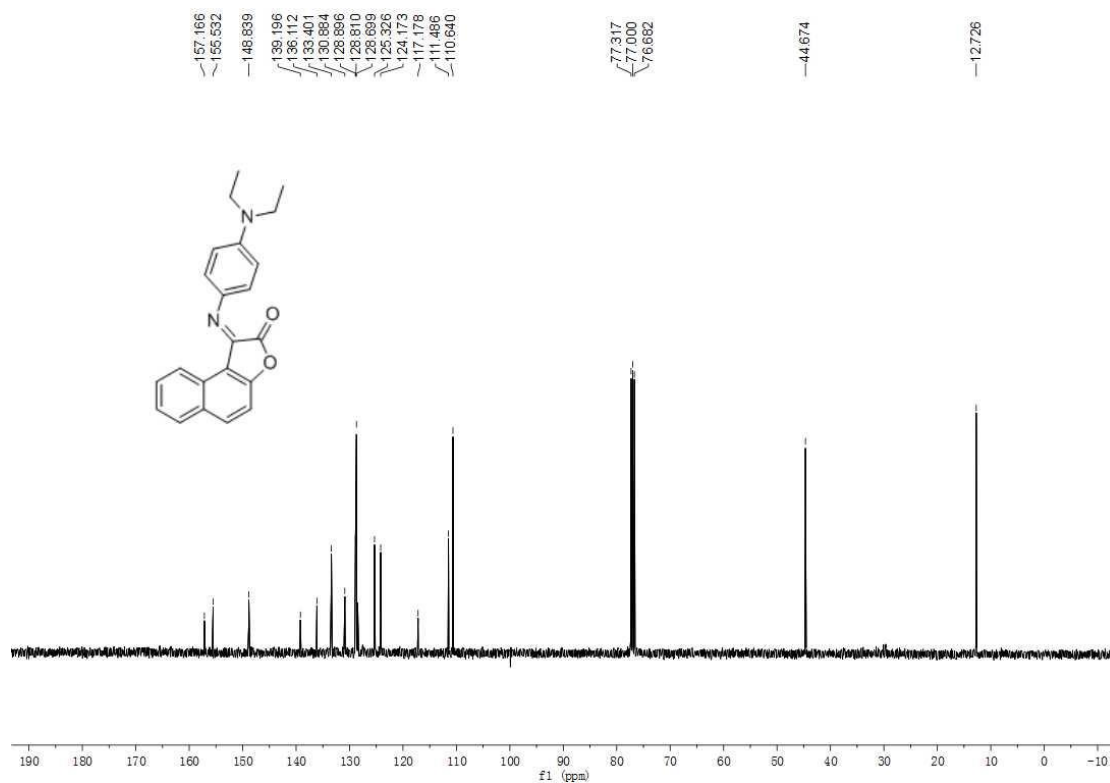
¹³C NMR Spectrum of (Z)-1-((4-(dimethylamino)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4j)



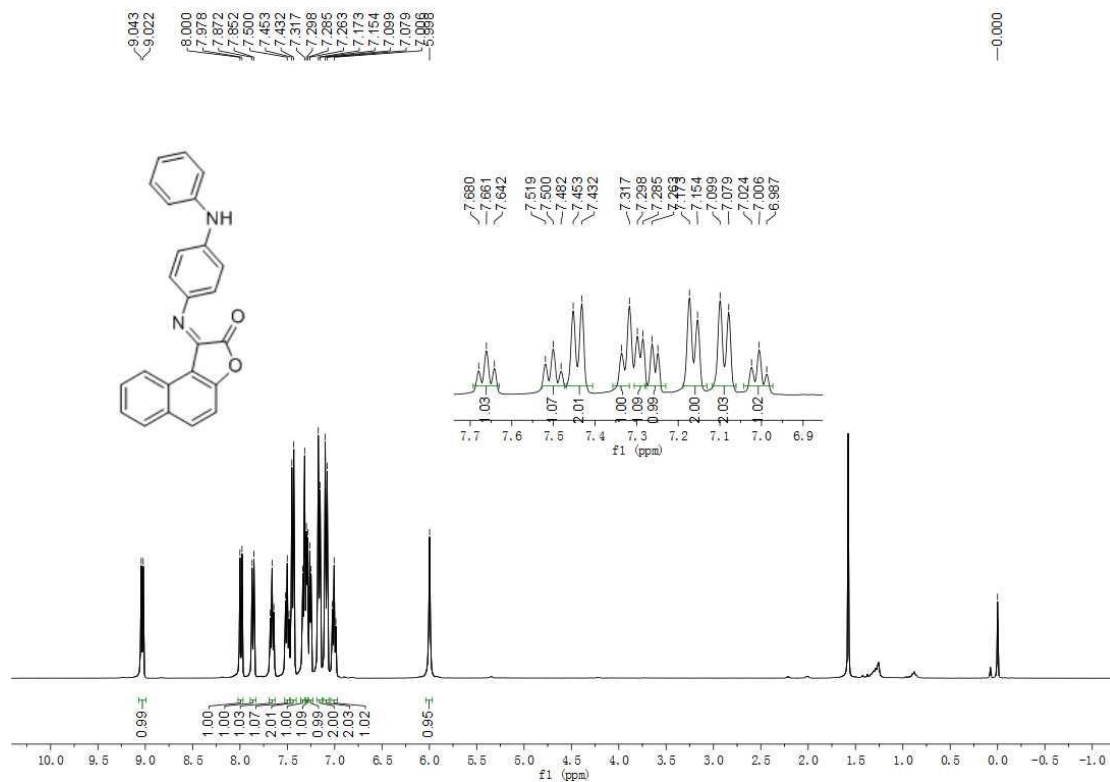
¹H NMR Spectrum of (Z)-1-((4-(diethylamino)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4k**)



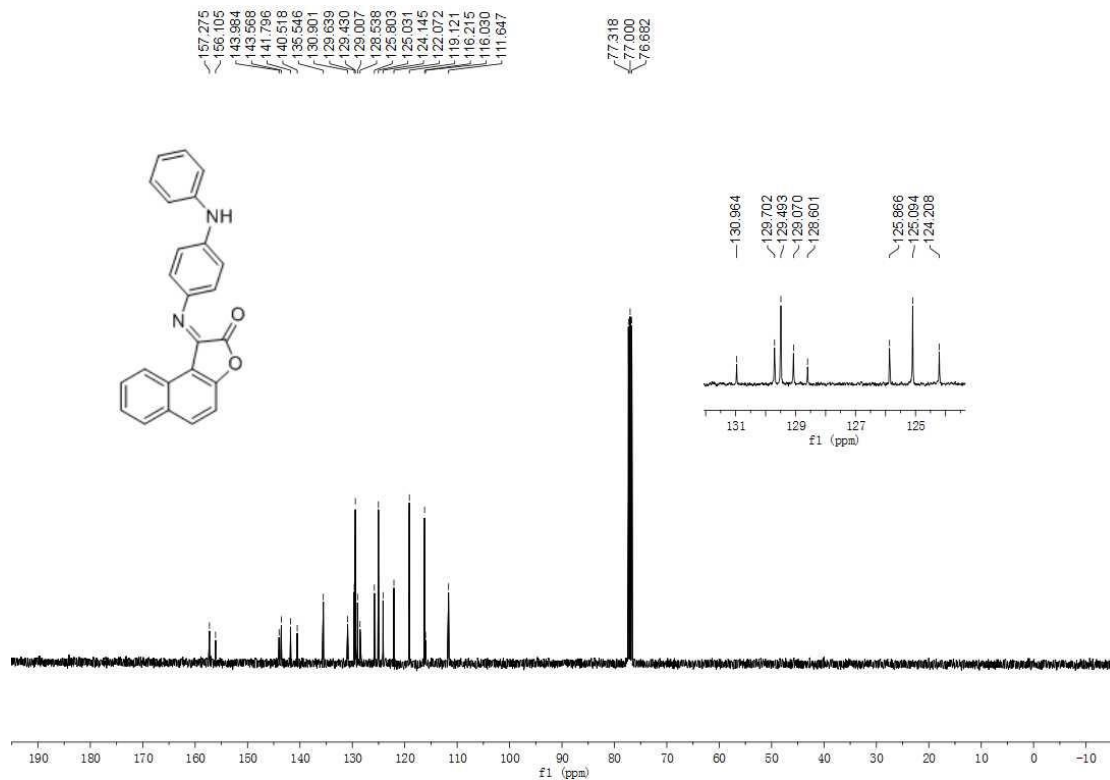
¹³C NMR Spectrum of (Z)-1-((4-(diethylamino)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4k**)



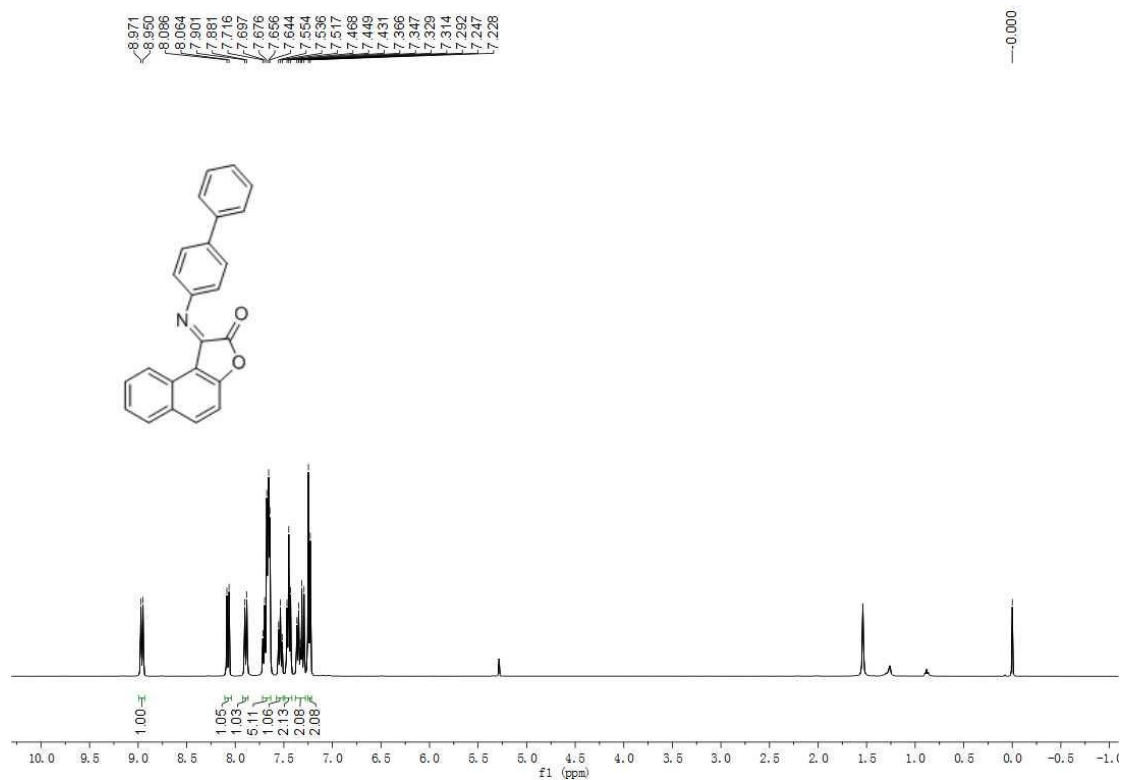
¹H NMR Spectrum of (Z)-1-((4-(phenylamino)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**41**)



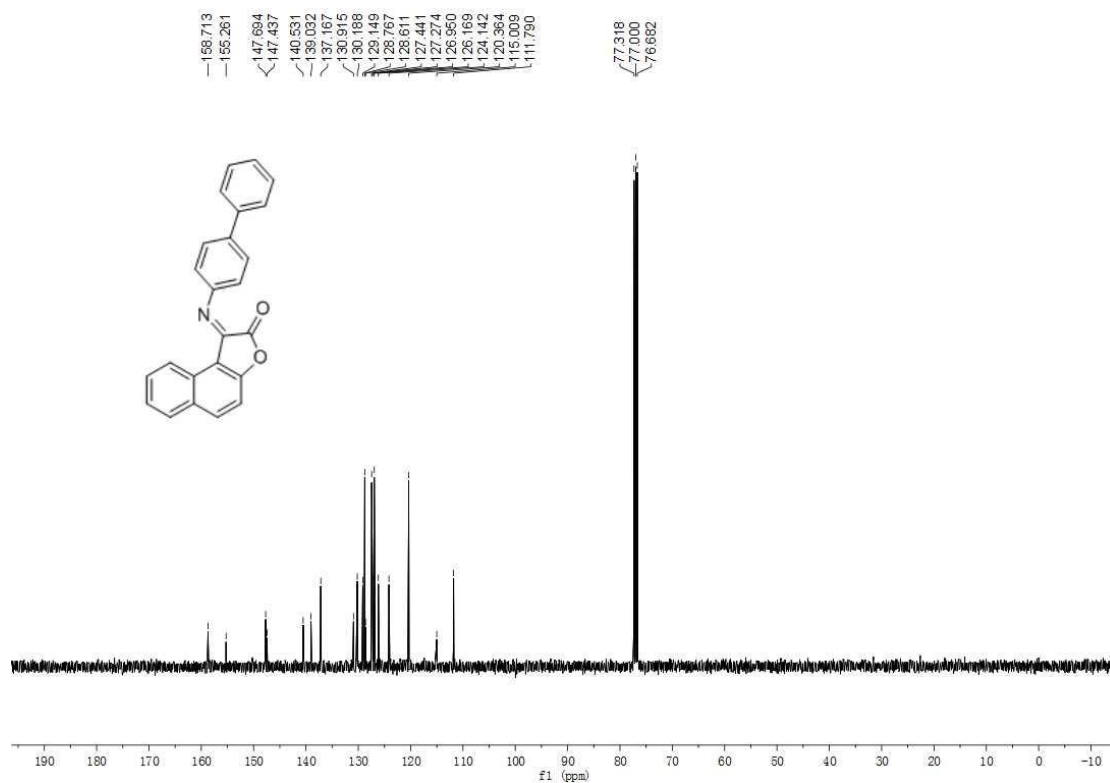
¹³C NMR Spectrum of (Z)-1-((4-(phenylamino)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**41**)



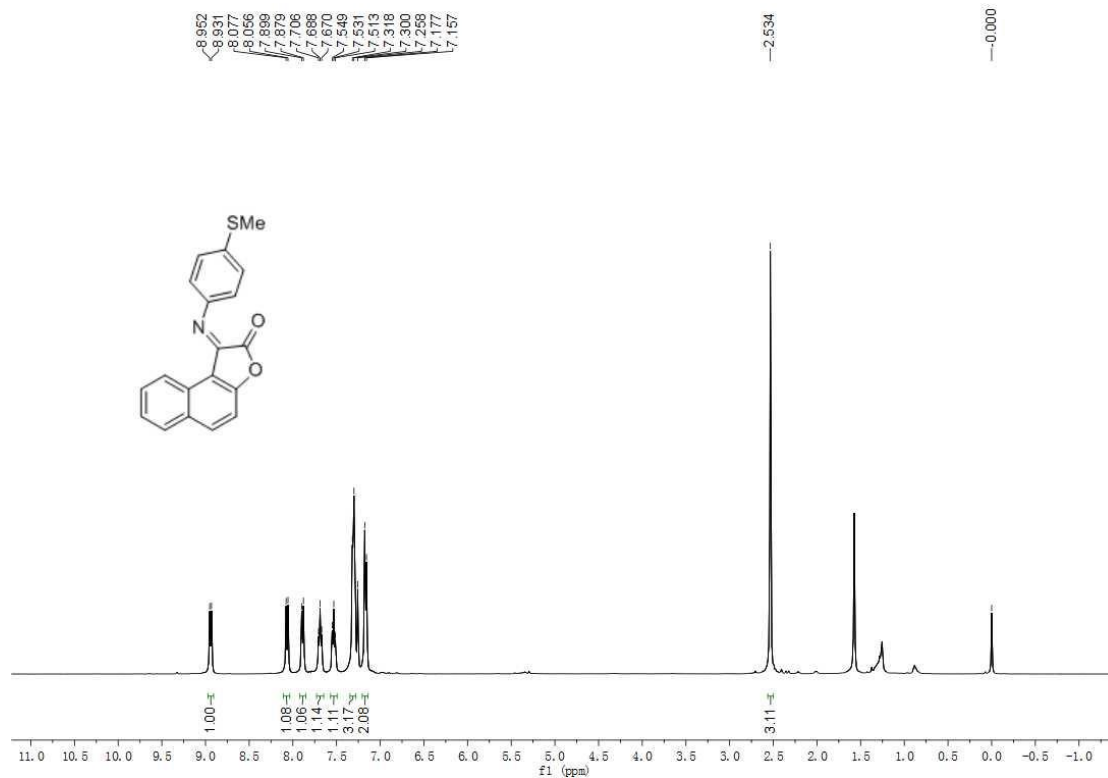
¹H NMR Spectrum of (Z)-1-([1,1'-biphenyl]-4-ylimino)naphtho[2,1-b]furan-2(1H)-one (**4m**)



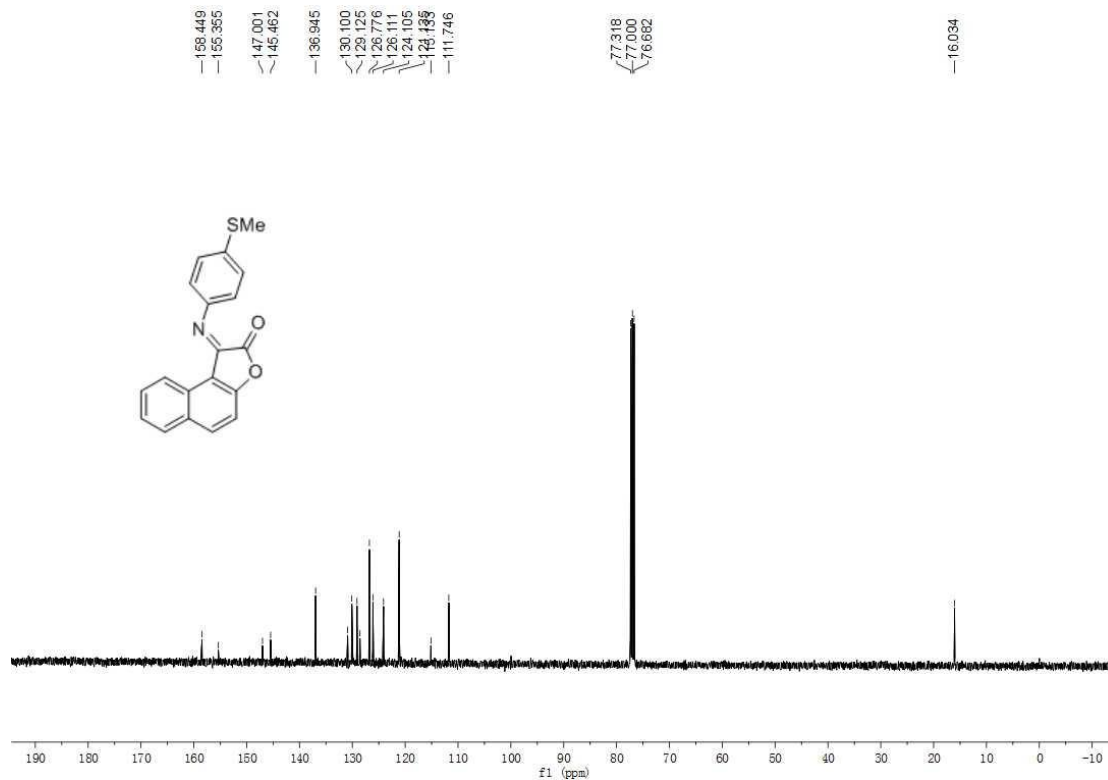
¹³C NMR Spectrum of (Z)-1-([1,1'-biphenyl]-4-ylimino)naphtho[2,1-b]furan-2(1H)-one (**4m**)



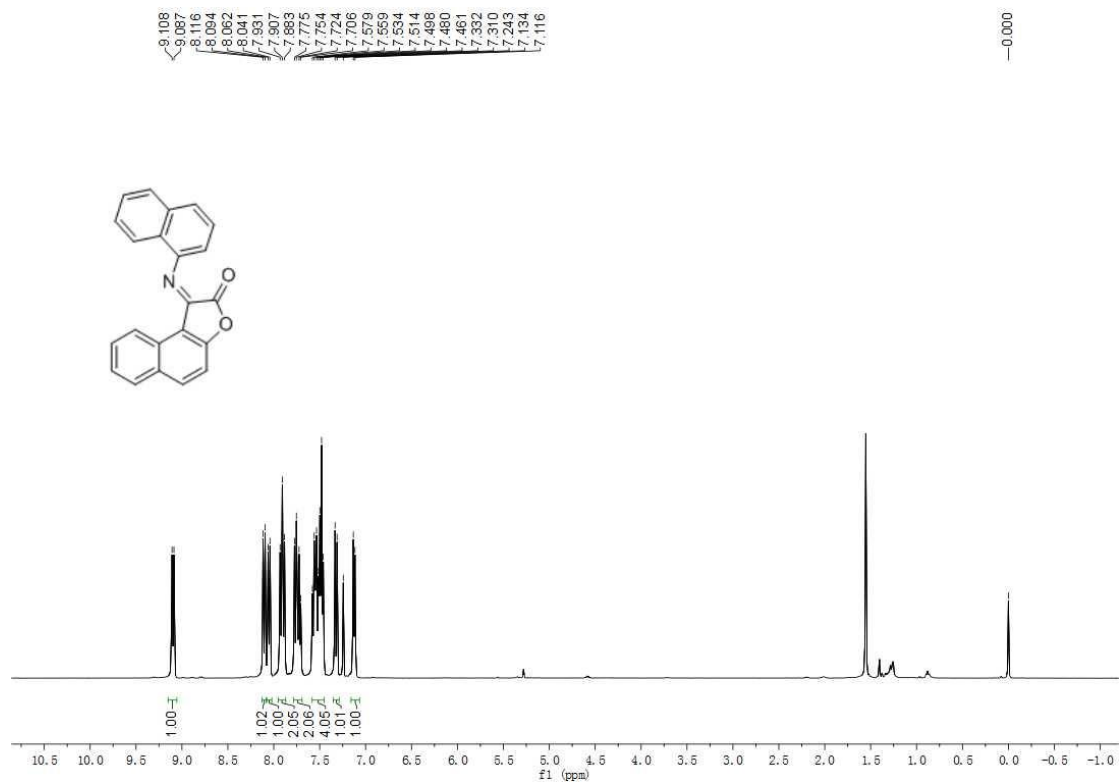
¹H NMR Spectrum of (Z)-1-((4-(methylthio)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4n**)



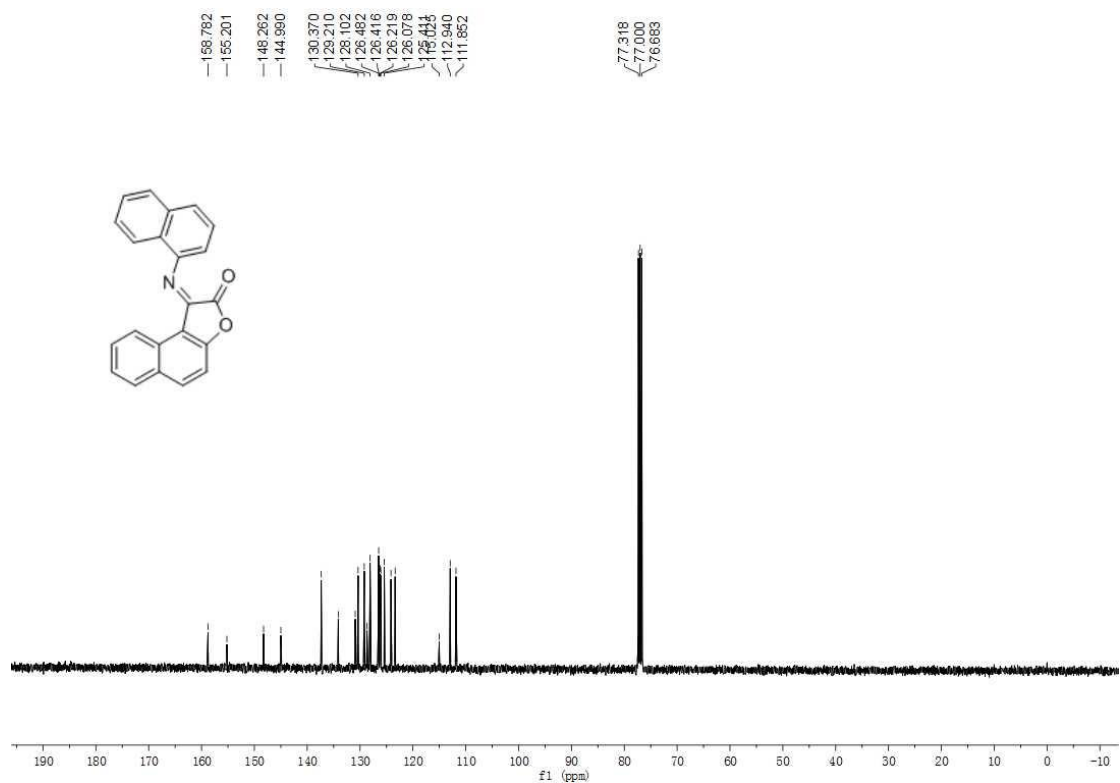
¹³C NMR Spectrum of (Z)-1-((4-(methylthio)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4n**)



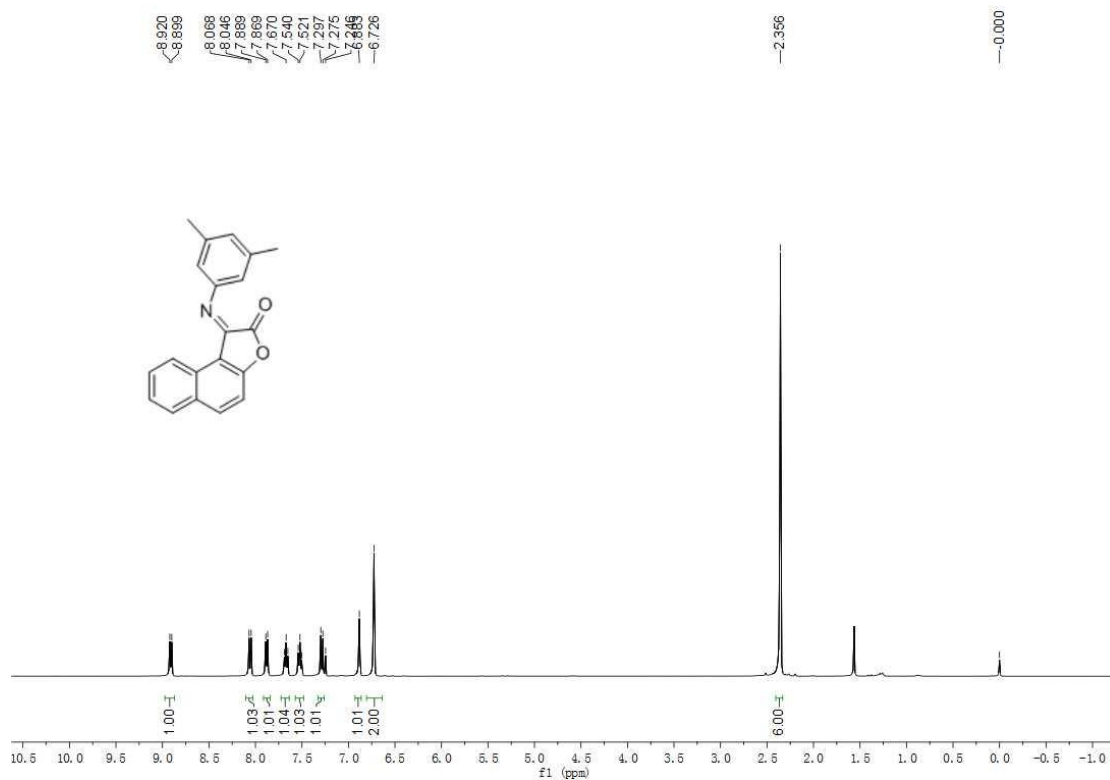
¹H NMR Spectrum of (Z)-1-(naphthalen-1-ylimino)naphtho[2,1-b]furan-2(1H)-one (**4o**)



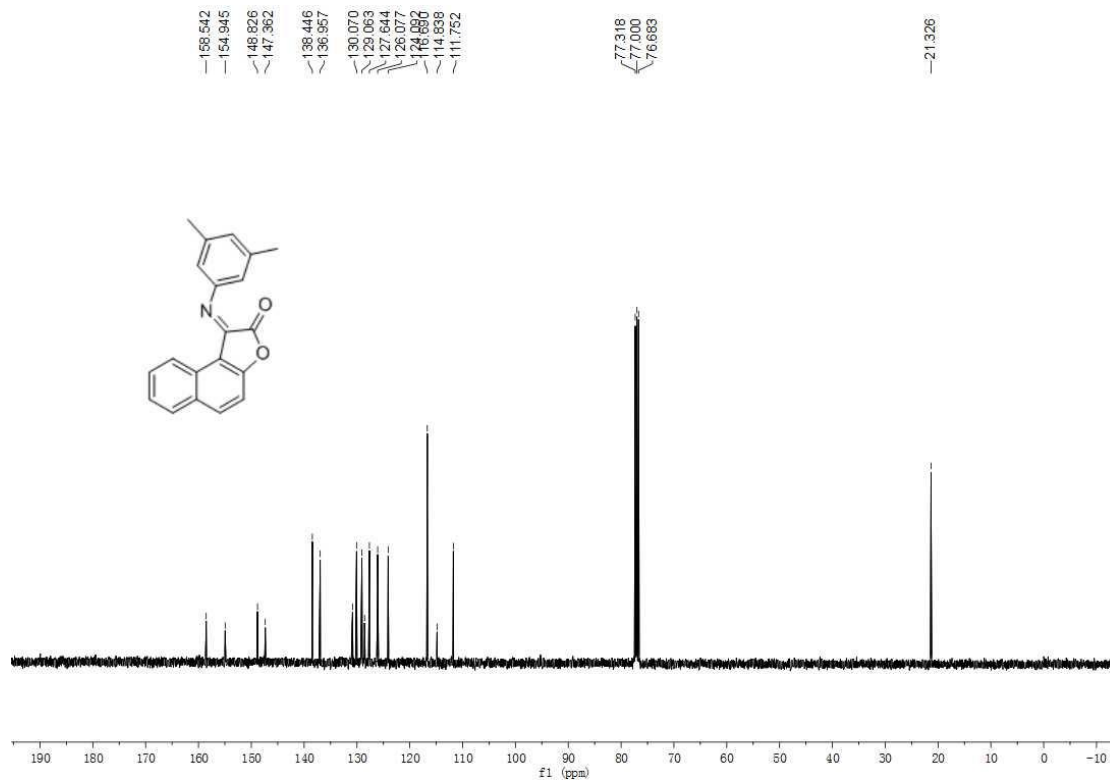
¹³C NMR Spectrum of (Z)-1-(naphthalen-1-ylimino)naphtho[2,1-b]furan-2(1H)-one (**4o**)



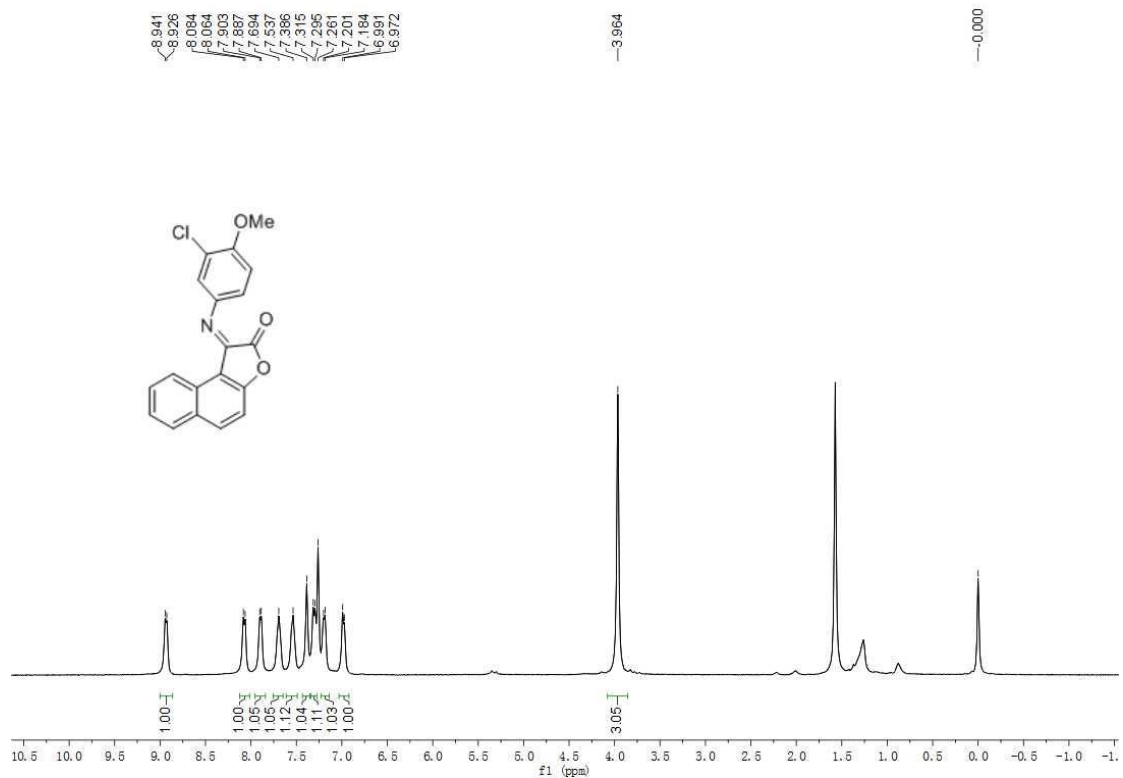
¹H NMR Spectrum of (Z)-1-((3,5-dimethylphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4p**)



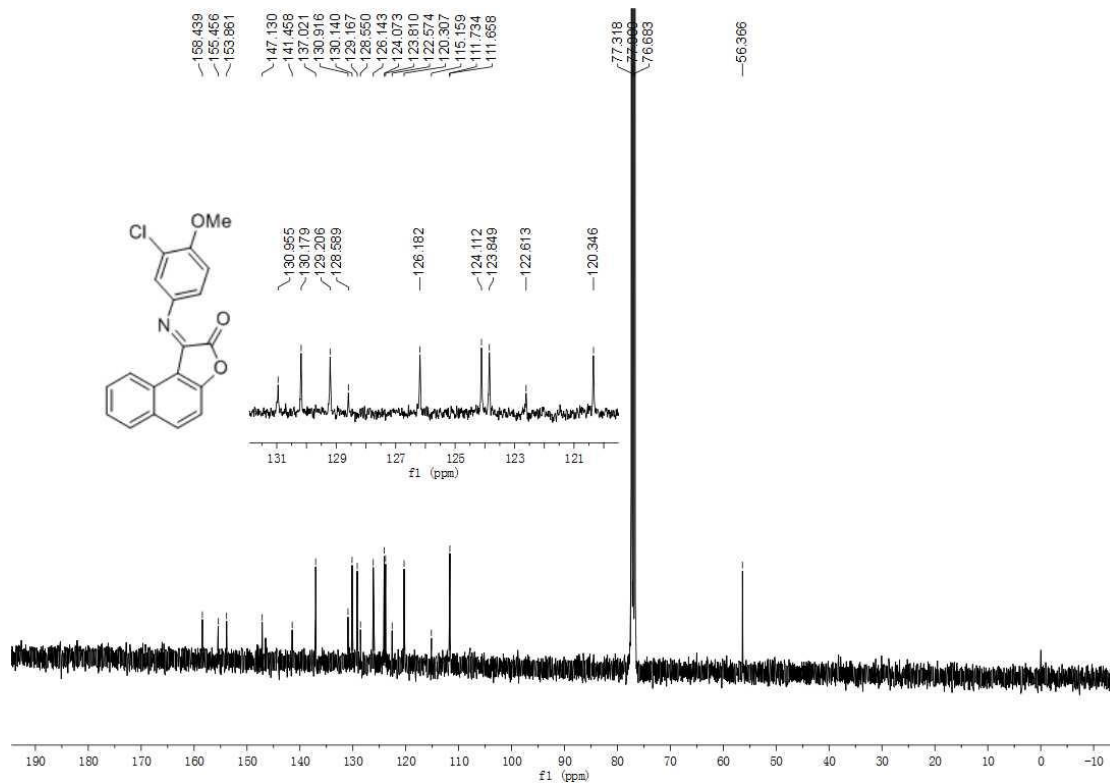
¹³C NMR Spectrum of (Z)-1-((3,5-dimethylphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4p**)



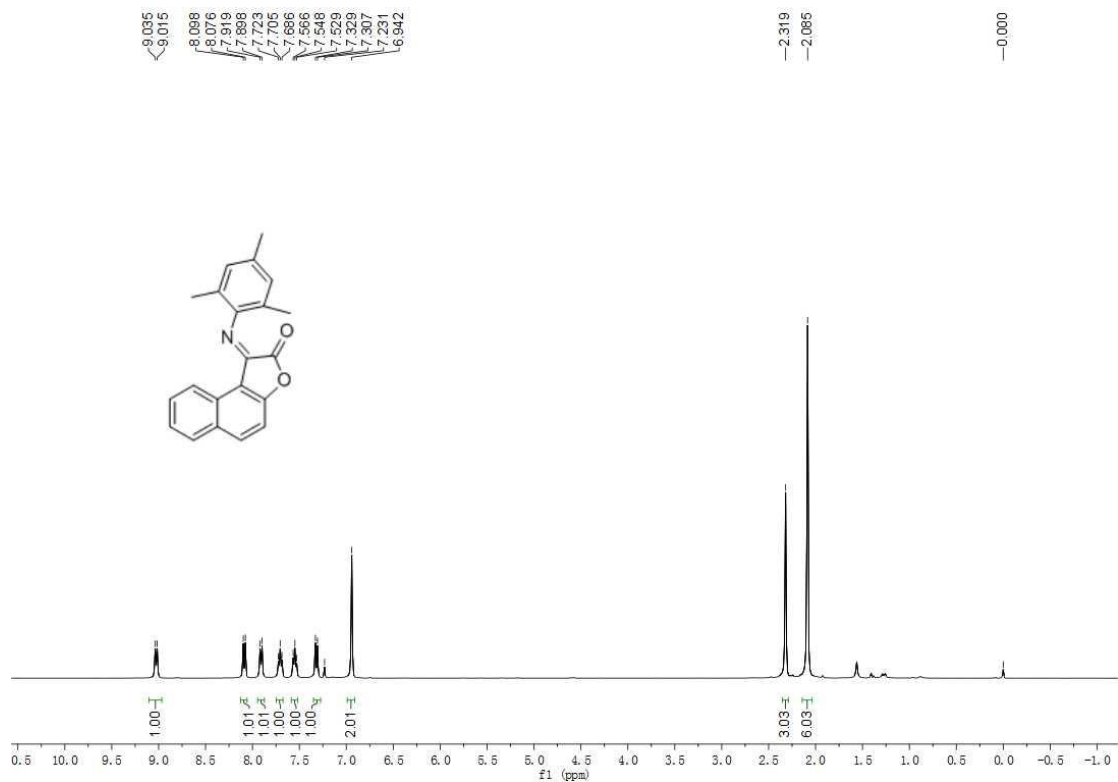
¹H NMR Spectrum of (Z)-1-((3-chloro-4-methoxyphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4q)



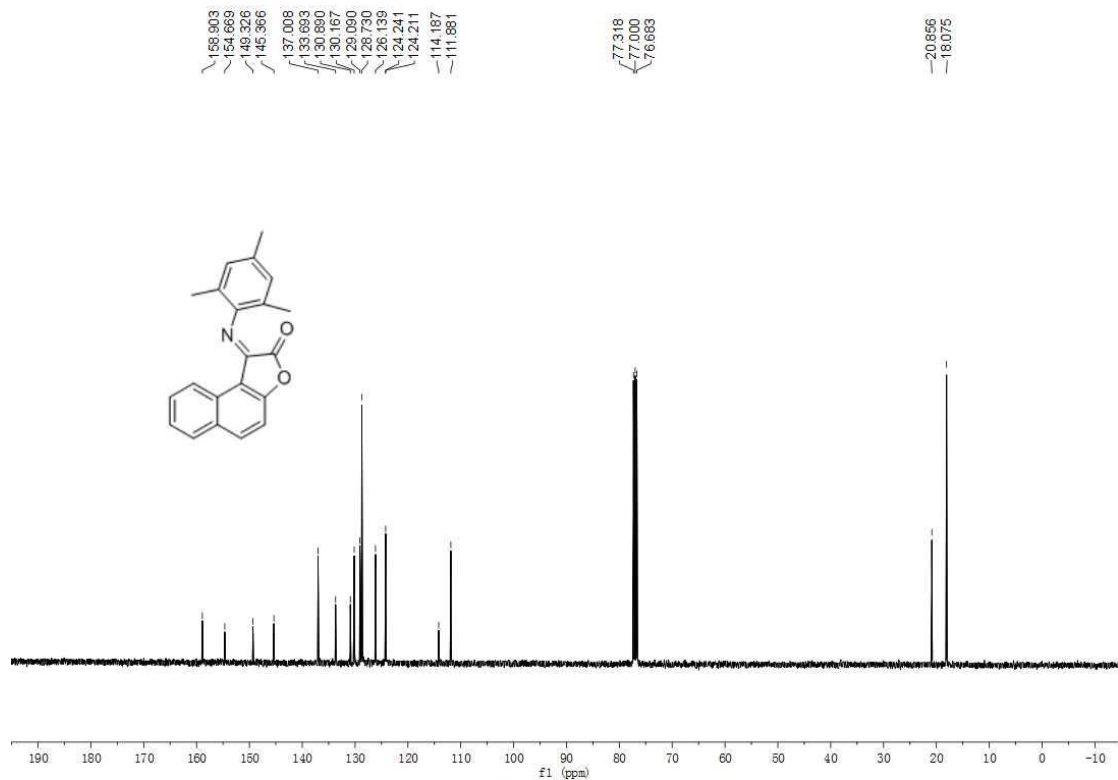
¹³C NMR Spectrum of (Z)-1-((3-chloro-4-methoxyphenyl)imino)naphtho[2,1-b]furan-2(1H)-one (4q)



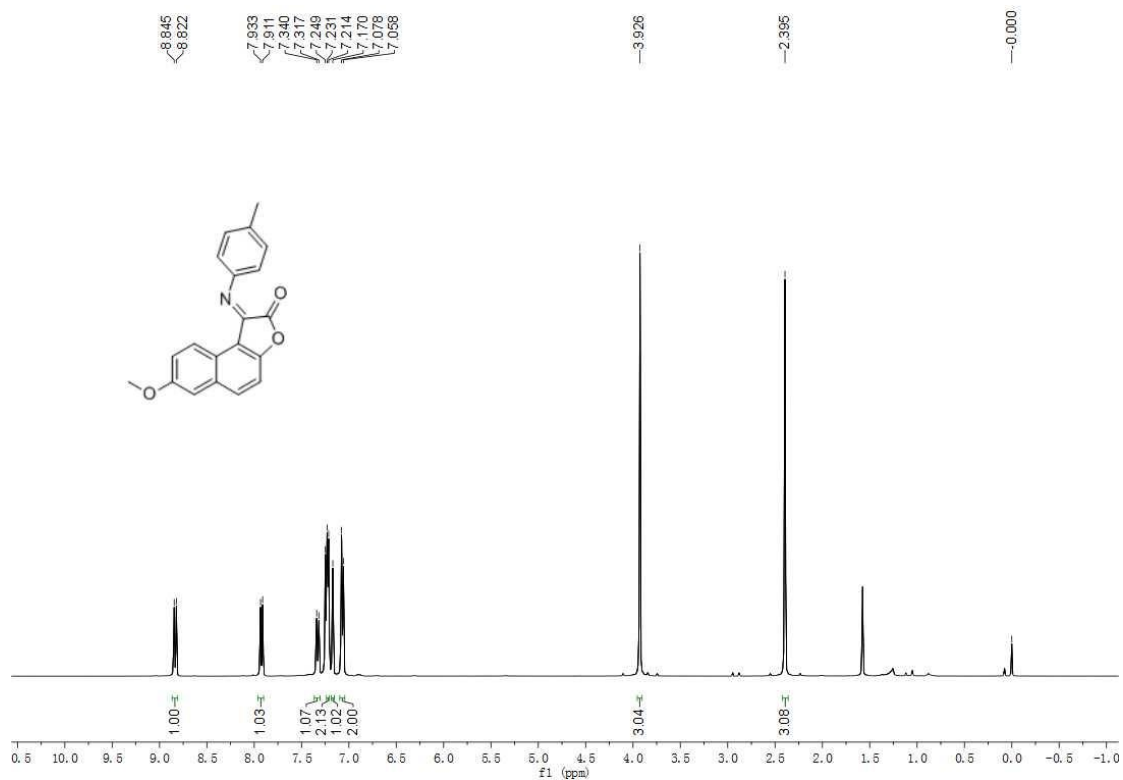
¹H NMR Spectrum of (Z)-1-(mesitylimino)naphtho[2,1-b]furan-2(1H)-one (**4r**)



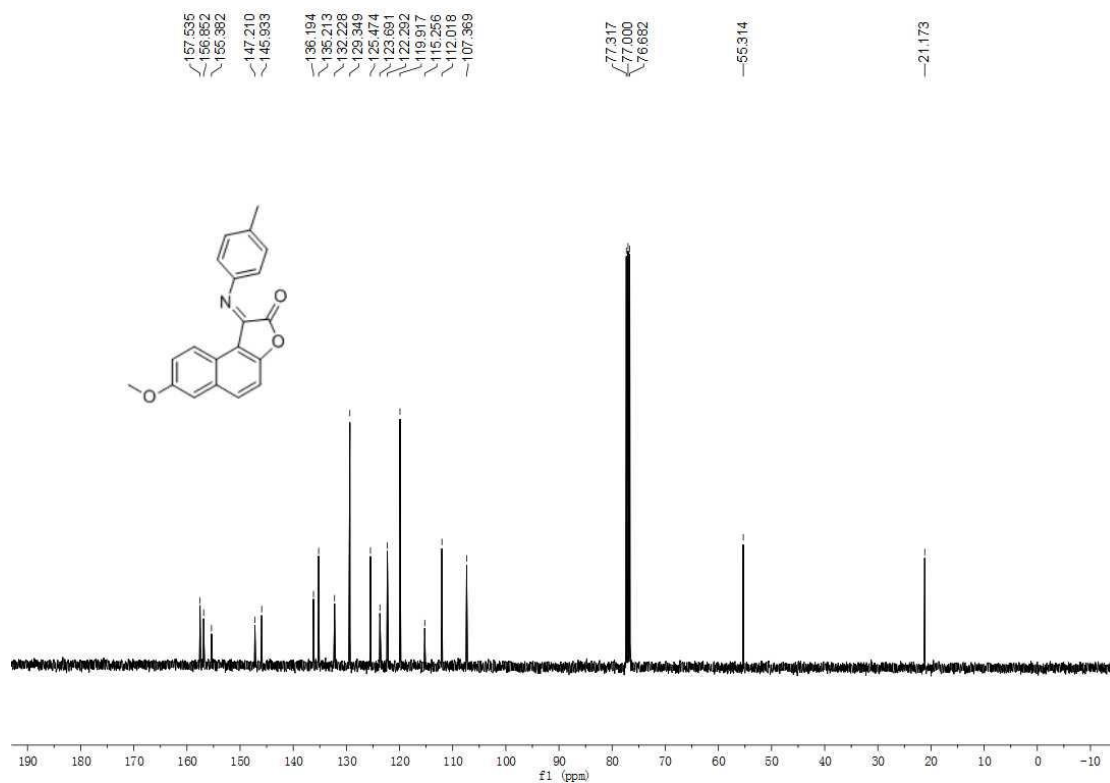
¹³C NMR Spectrum of (Z)-1-(mesitylimino)naphtho[2,1-b]furan-2(1H)-one (**4r**)



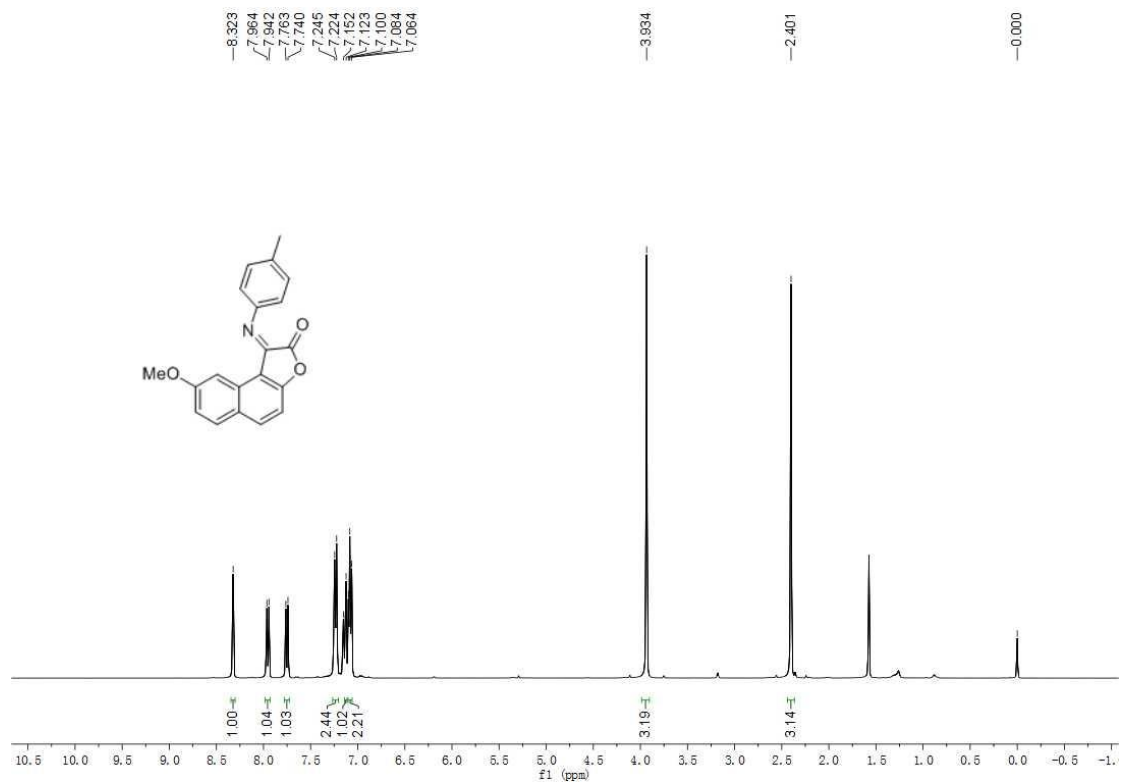
¹H NMR Spectrum of (Z)-7-methoxy-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (**4s**)



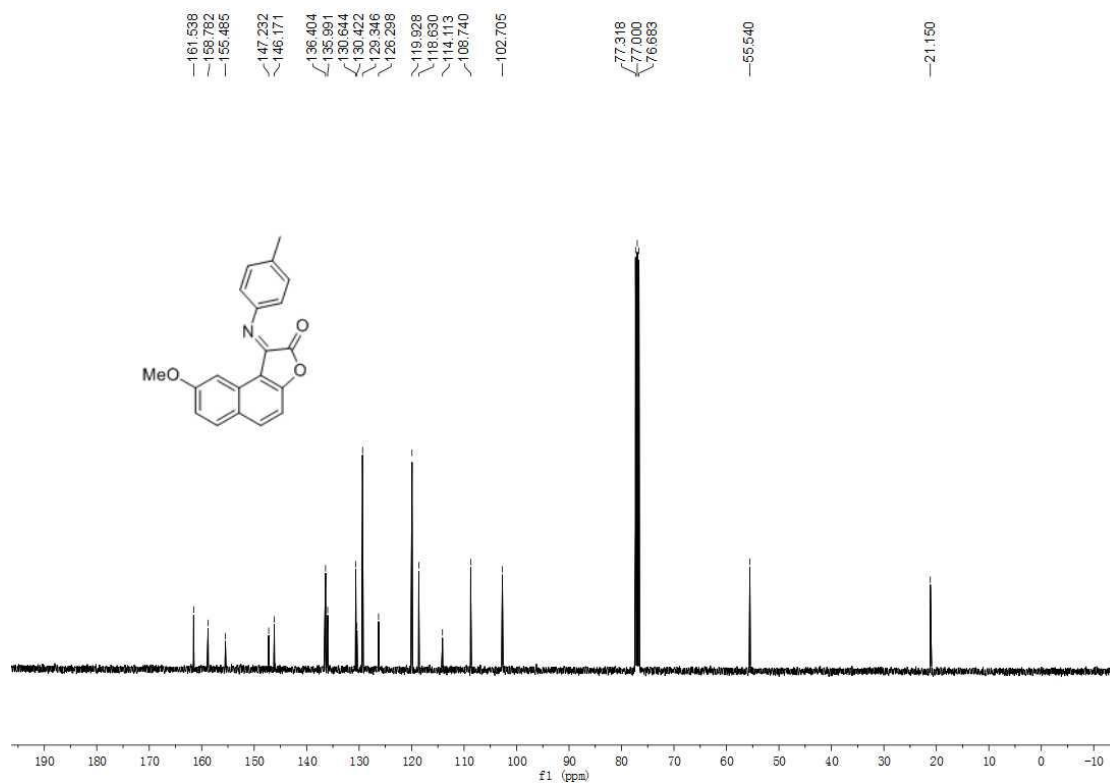
¹³C NMR Spectrum of (Z)-7-methoxy-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (**4s**)



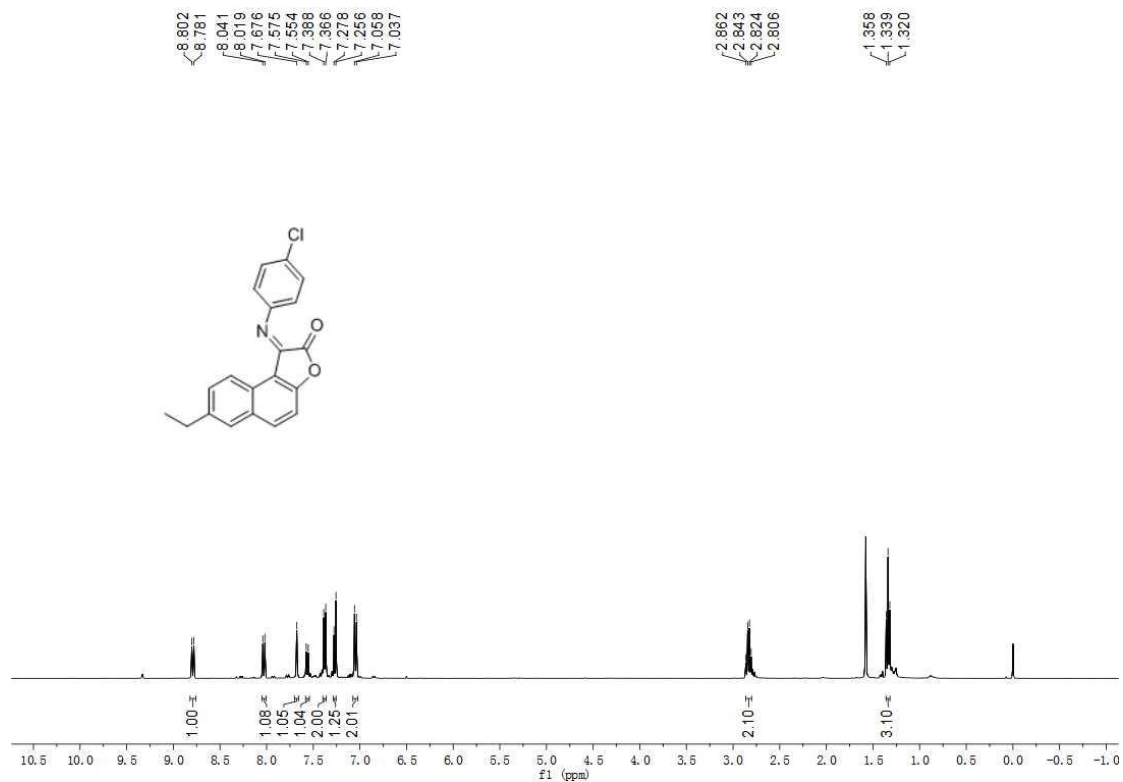
¹H NMR Spectrum of (Z)-8-methoxy-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (**4t**)



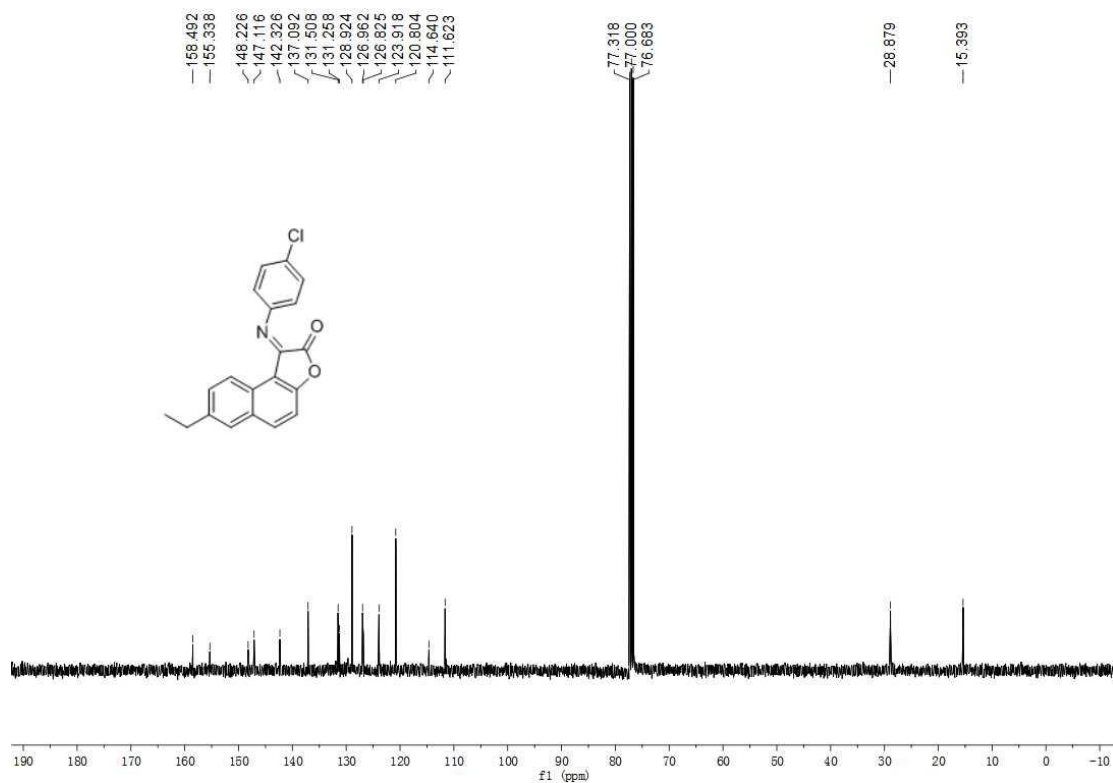
¹³C NMR Spectrum of (Z)-8-methoxy-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (**4t**)



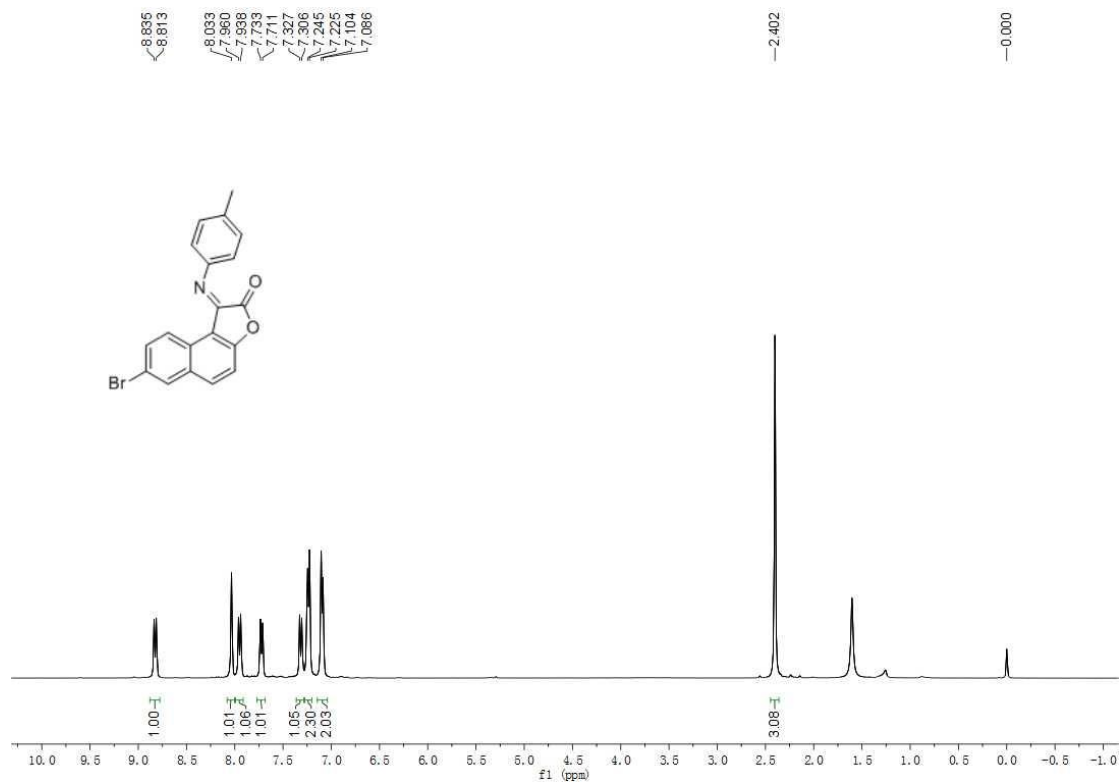
¹H NMR Spectrum of (Z)-1-((4-chlorophenyl)imino)-7-ethylnaphtho[2,1-*b*]furan-2(1*H*)-one (**4u**)



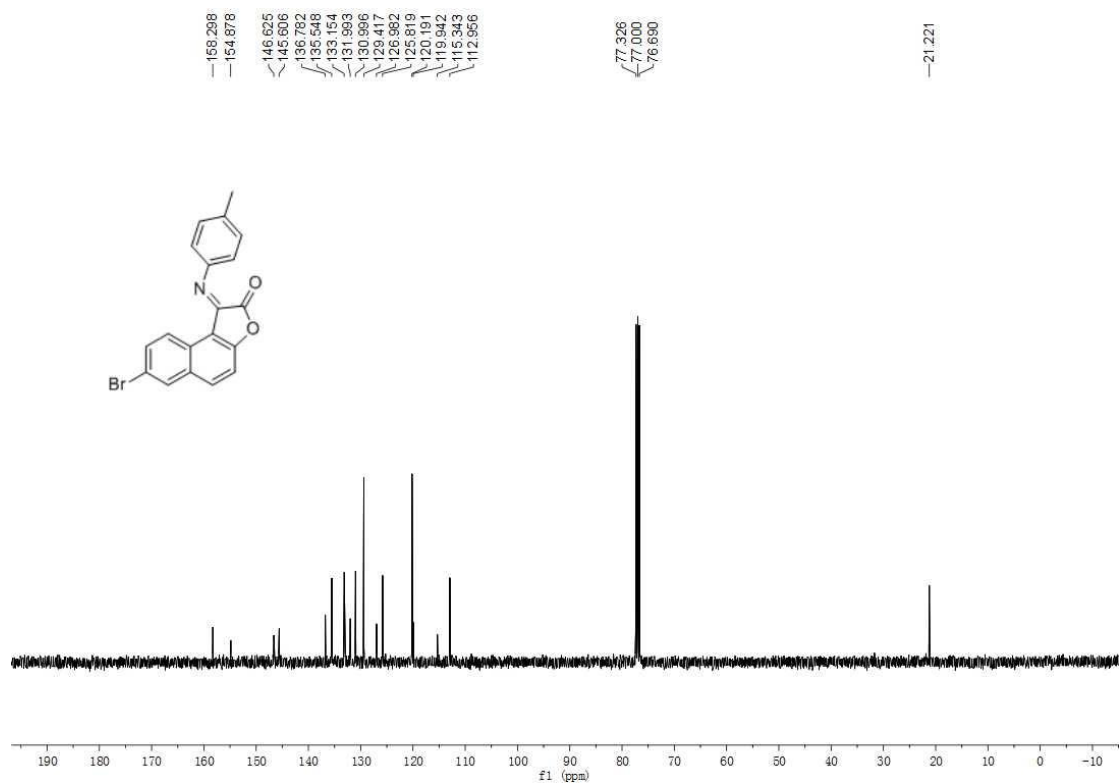
¹³C NMR Spectrum of (Z)-1-((4-chlorophenyl)imino)-7-ethylnaphtho[2,1-*b*]furan-2(1*H*)-one (**4u**)



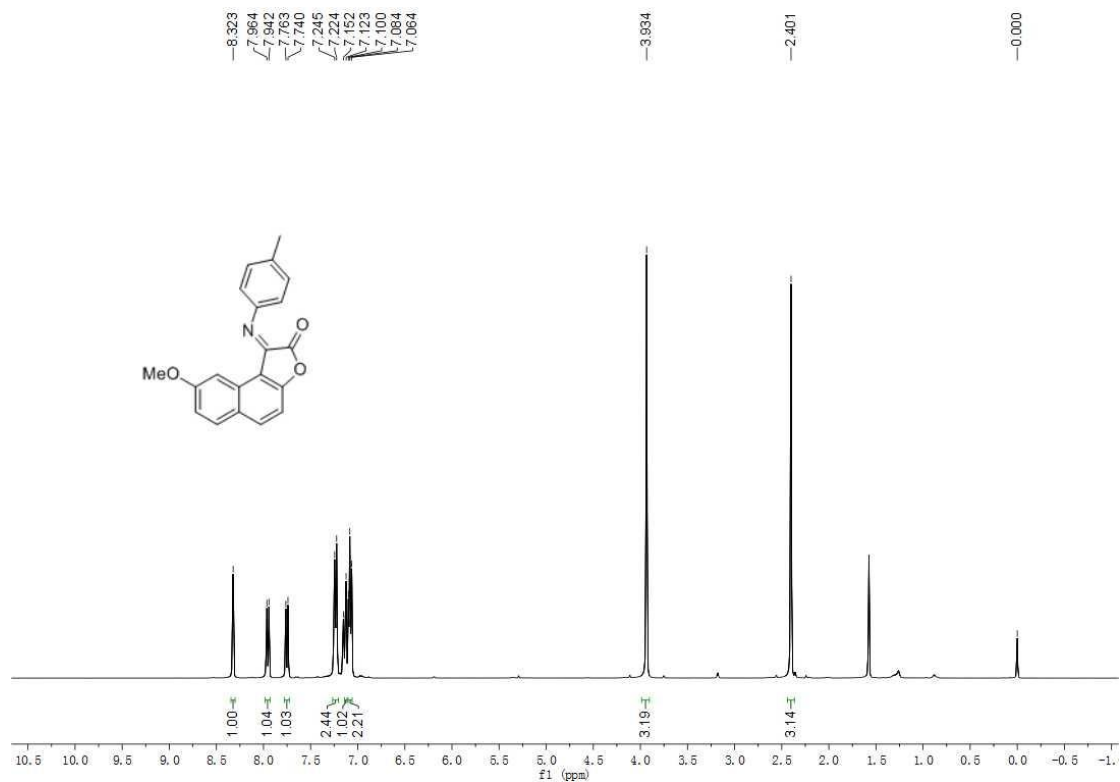
¹H NMR Spectrum of (Z)-7-bromo-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (**4v**)



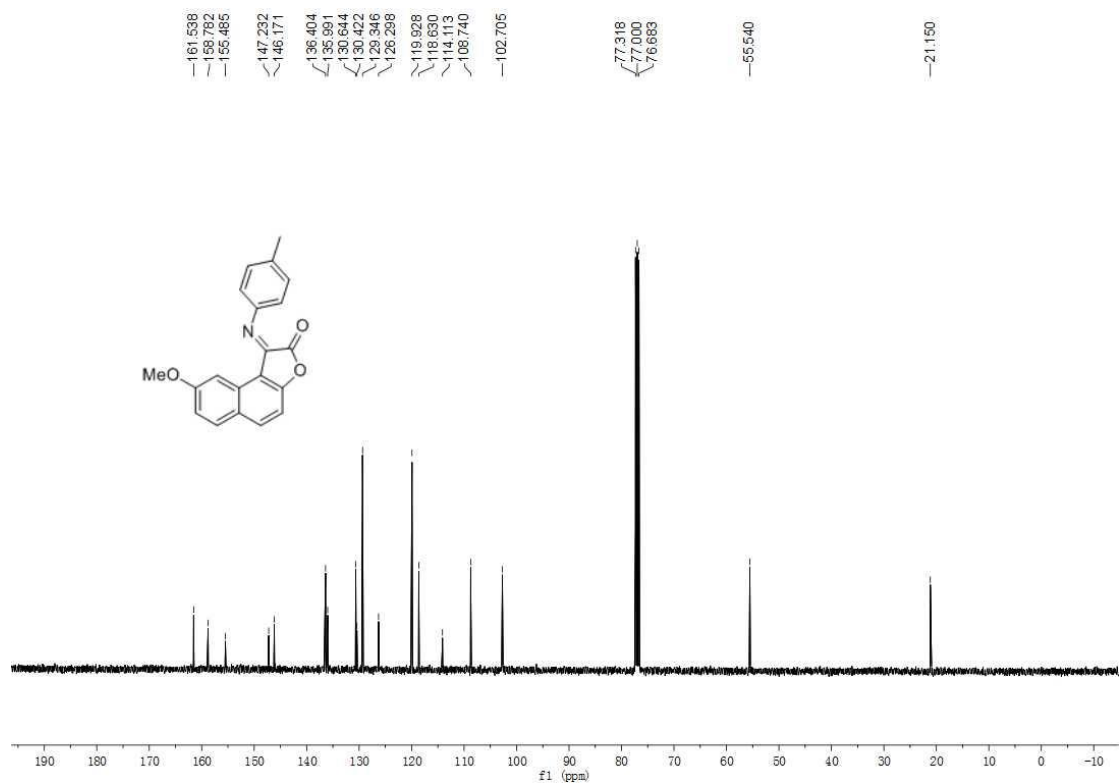
¹³C NMR Spectrum of (Z)-7-bromo-1-(p-tolylimino)naphtho[2,1-b]furan-2(1H)-one (**4v**)



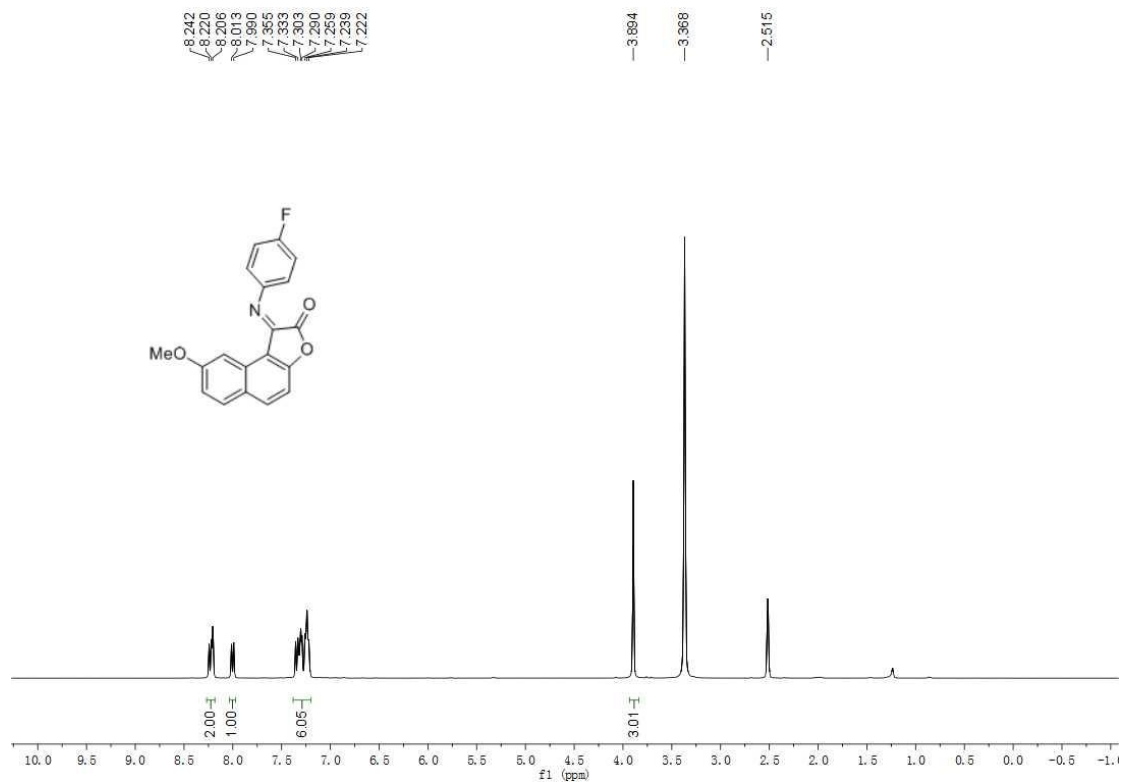
¹H NMR Spectrum of (Z)-8-bromo-1-(*p*-tolylimino)naphtho[2,1-*b*]furan-2(1*H*)-one (**4w**)



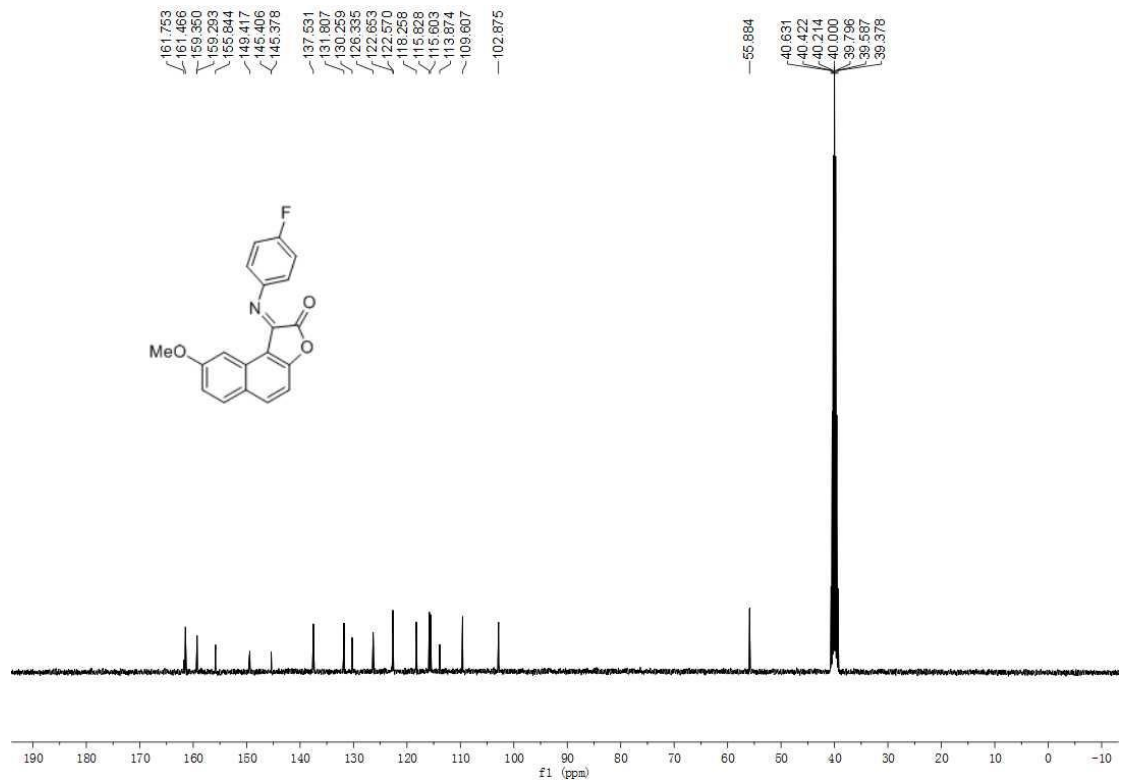
¹³C NMR Spectrum of (Z)-8-bromo-1-(*p*-tolylimino)naphtho[2,1-*b*]furan-2(1*H*)-one (**4w**)



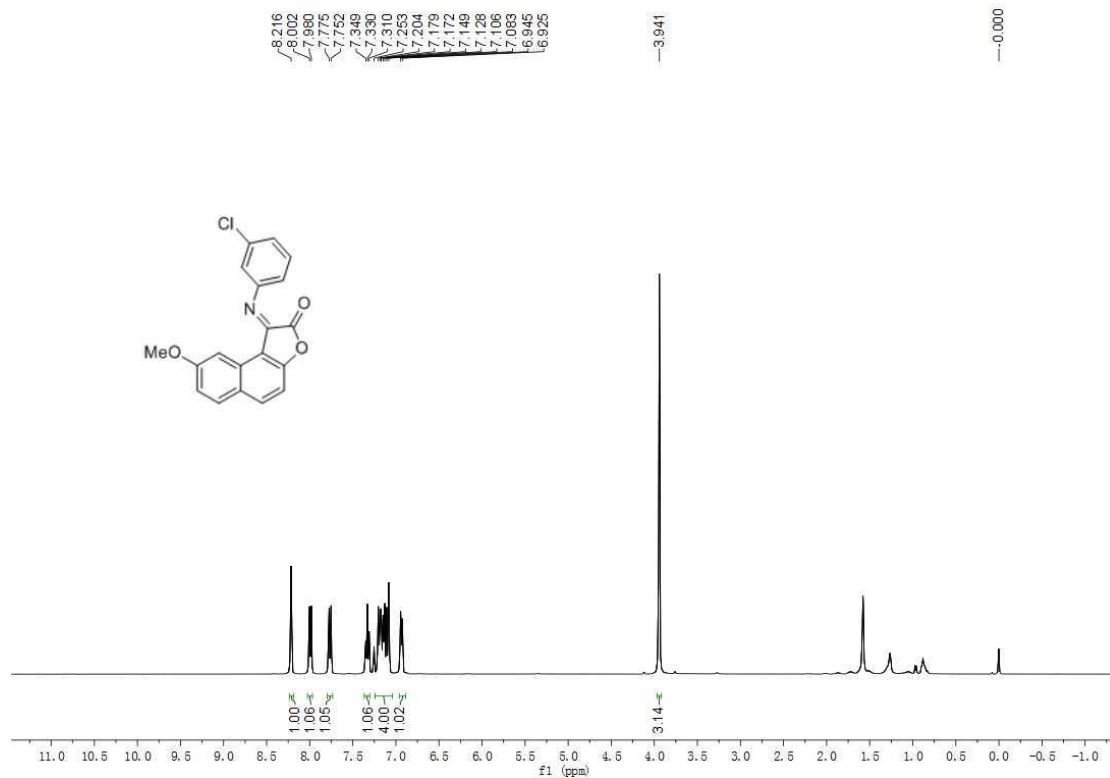
¹H NMR Spectrum of (Z)-1-((4-fluorophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4x)



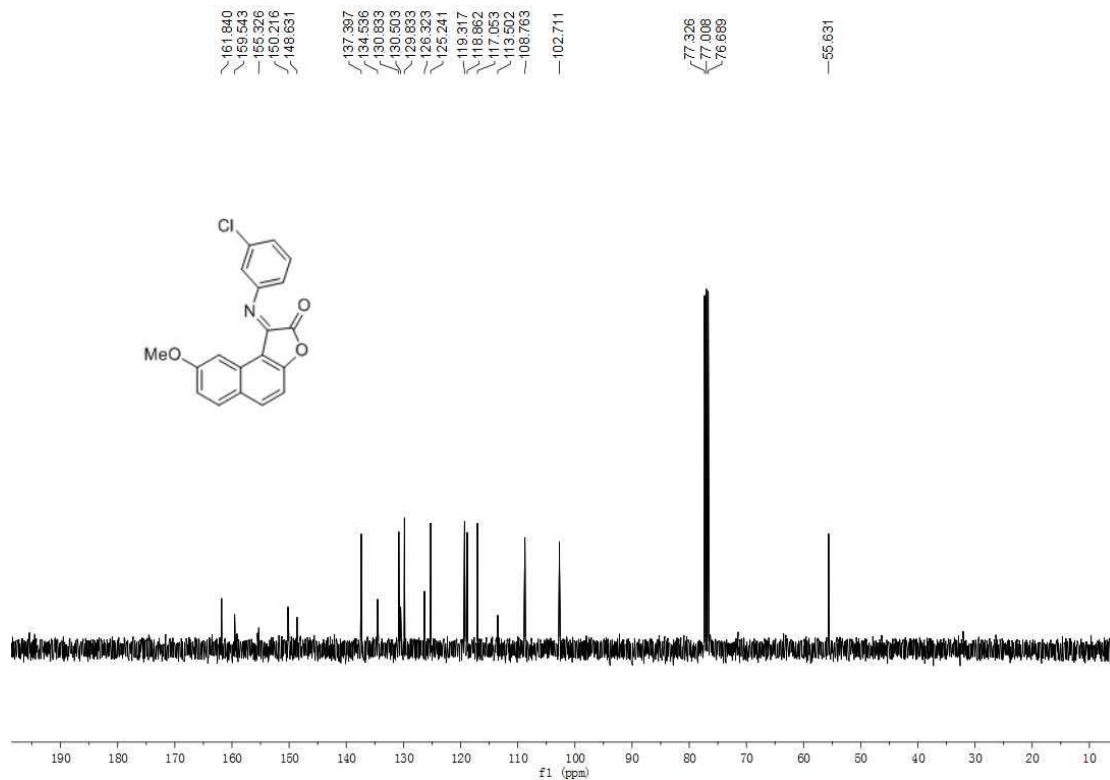
¹³C NMR Spectrum of (Z)-1-((4-fluorophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4x)



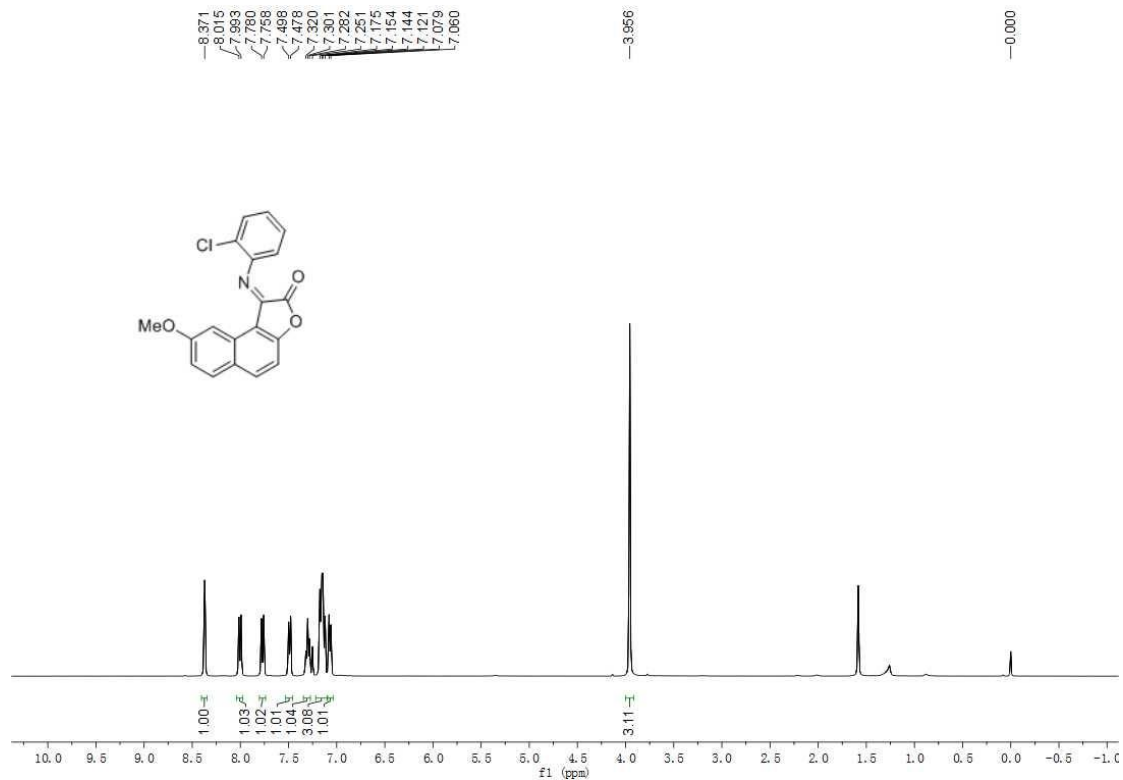
¹H NMR Spectrum of (Z)-1-((3-chlorophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4y)



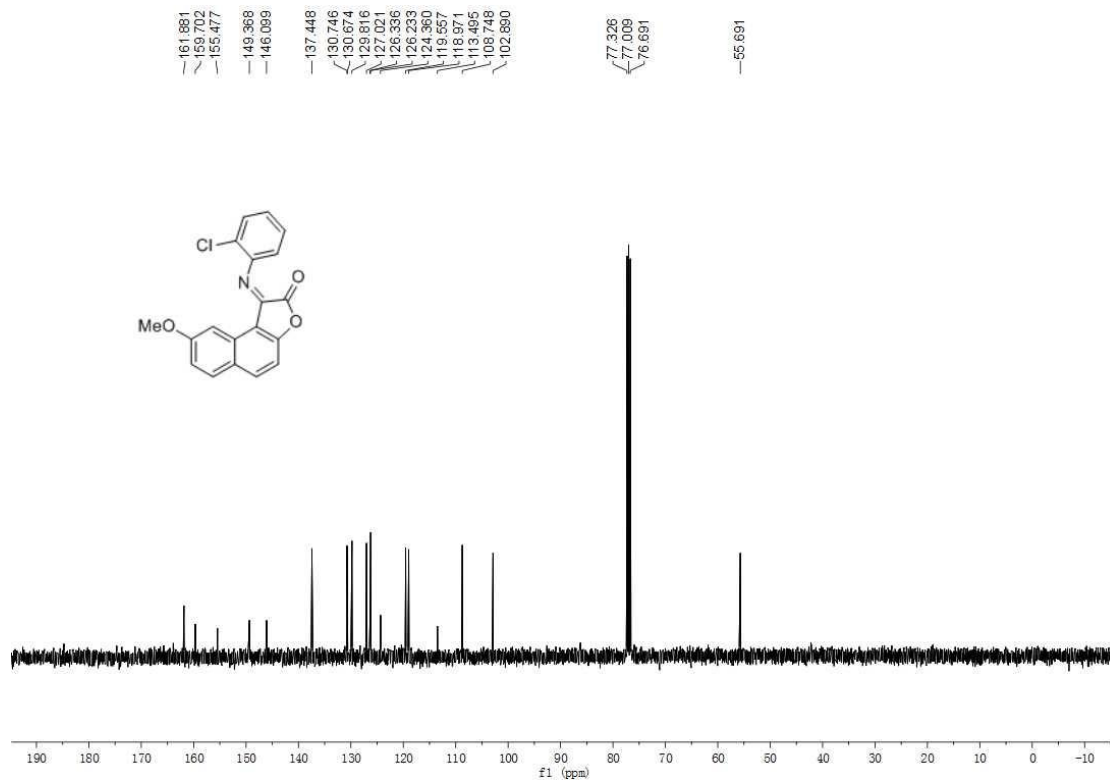
¹³C NMR Spectrum of (Z)-1-((3-chlorophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4y)



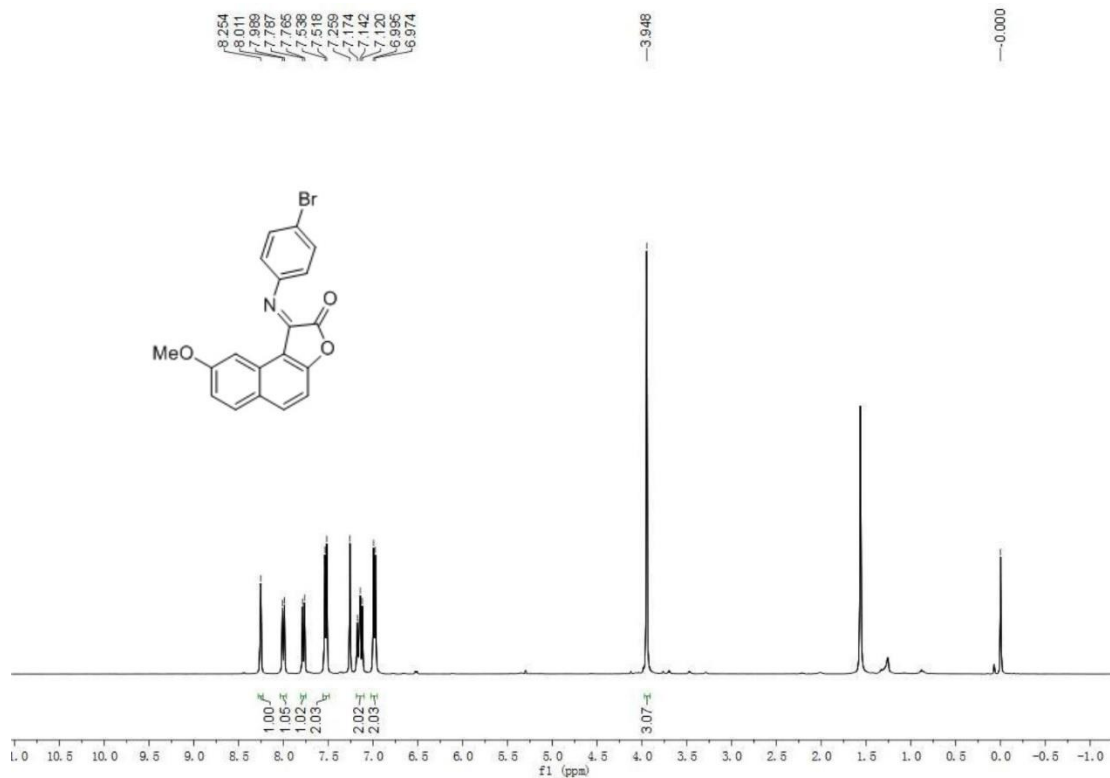
¹H NMR Spectrum of (Z)-1-((2-chlorophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4z)



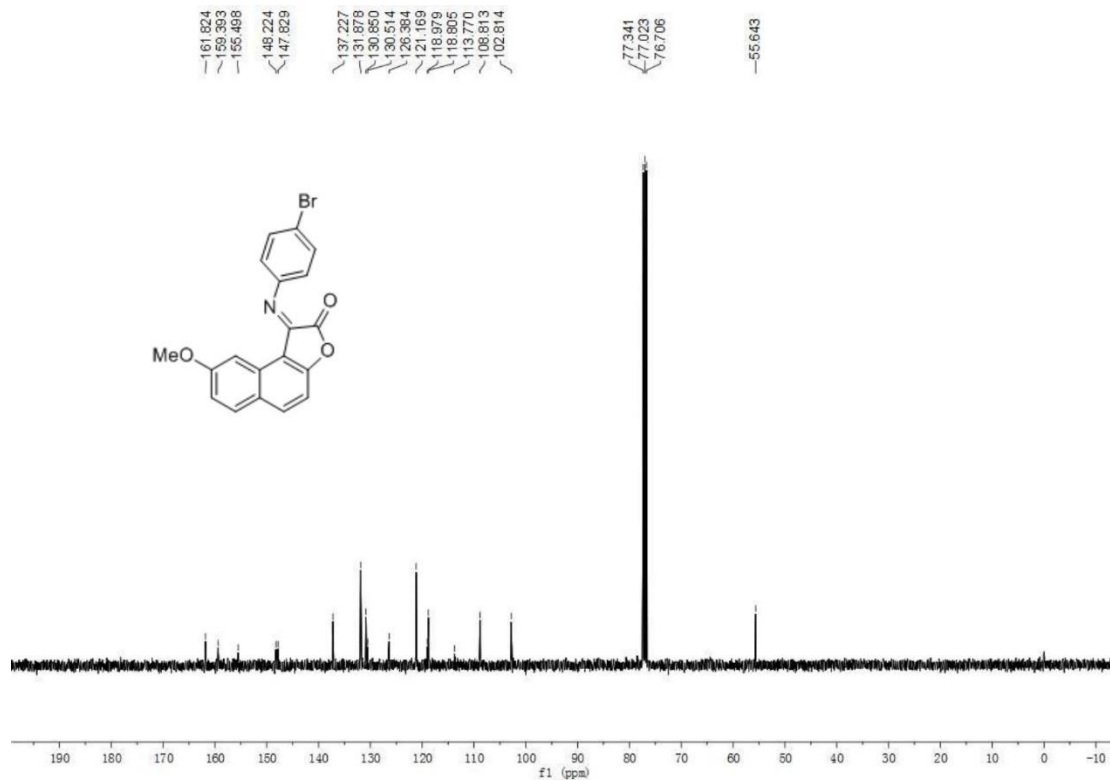
¹³C NMR Spectrum of (Z)-1-((2-chlorophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4z)



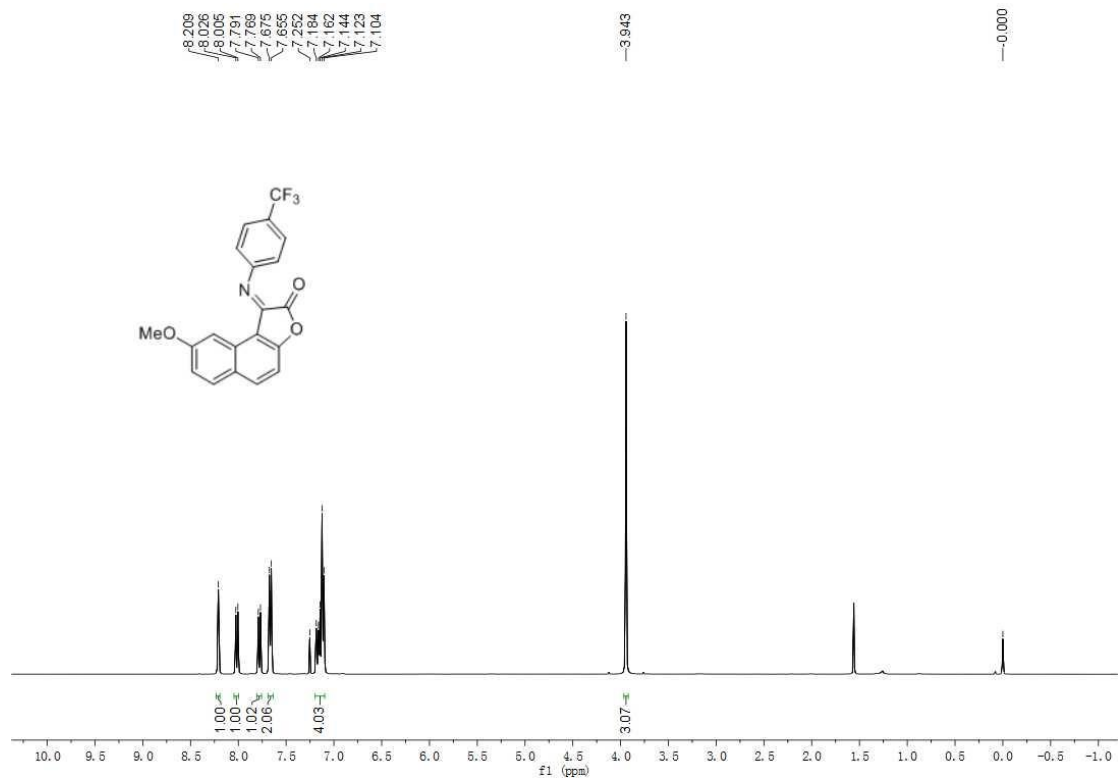
¹H NMR Spectrum of (Z)-1-((4-bromophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4za)



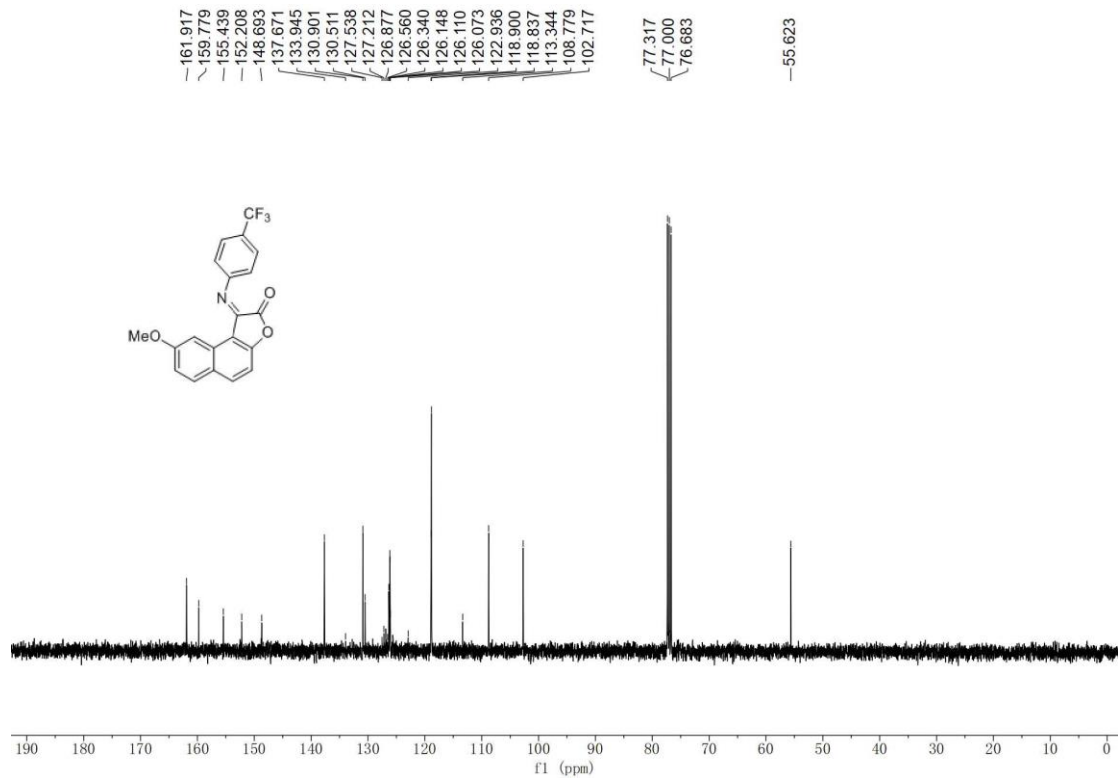
¹³C NMR Spectrum of (Z)-1-((4-bromophenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4za)



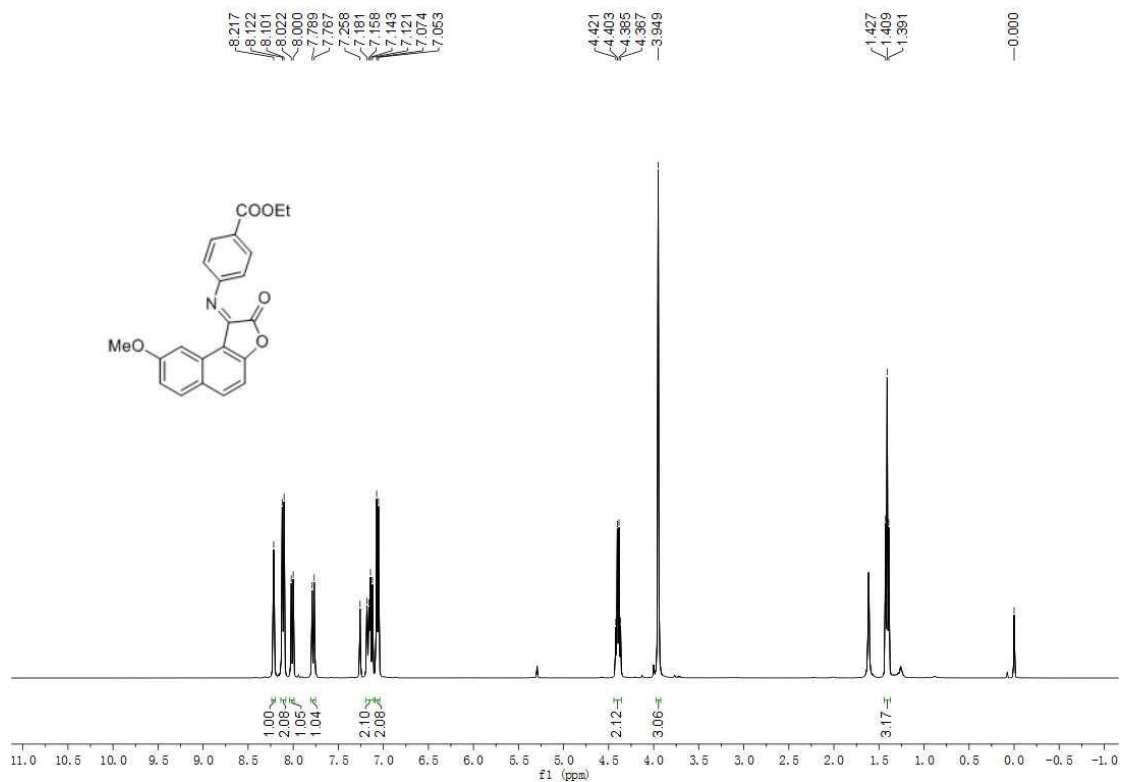
¹H NMR Spectrum of (Z)-8-methoxy-1-((4-(trifluoromethyl)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4zb**)



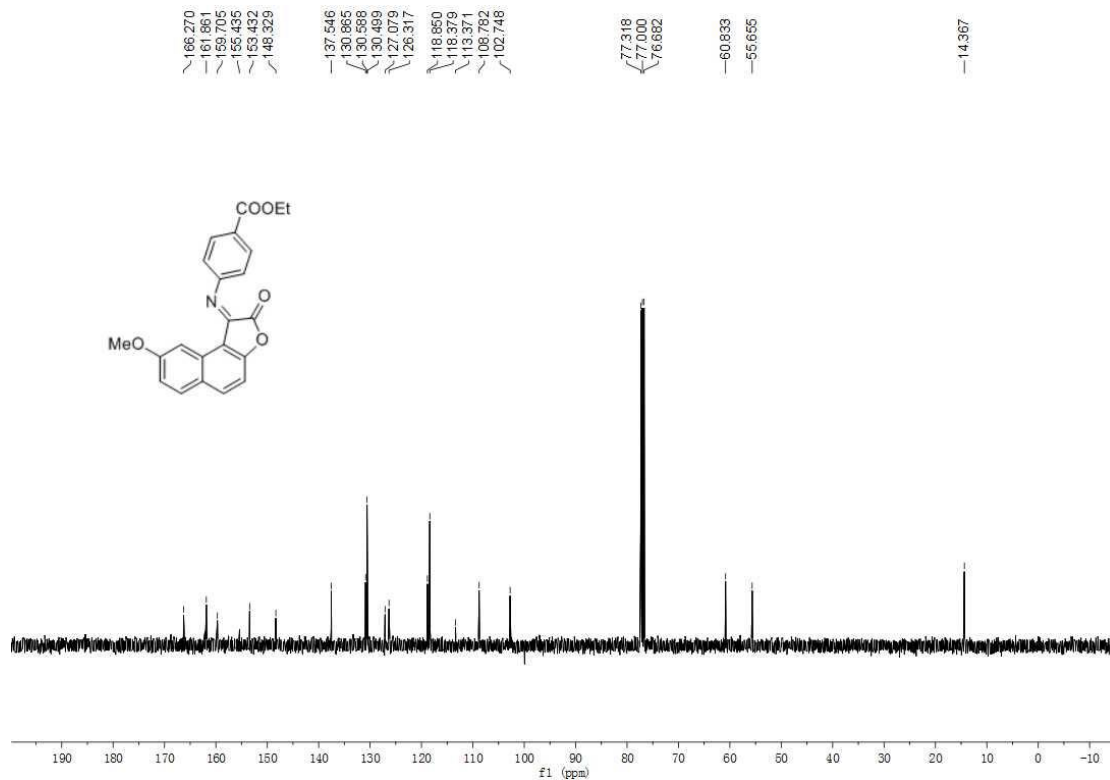
¹³C NMR Spectrum of (Z)-8-methoxy-1-((4-(trifluoromethyl)phenyl)imino)naphtho[2,1-b]furan-2(1H)-one (**4zb**)



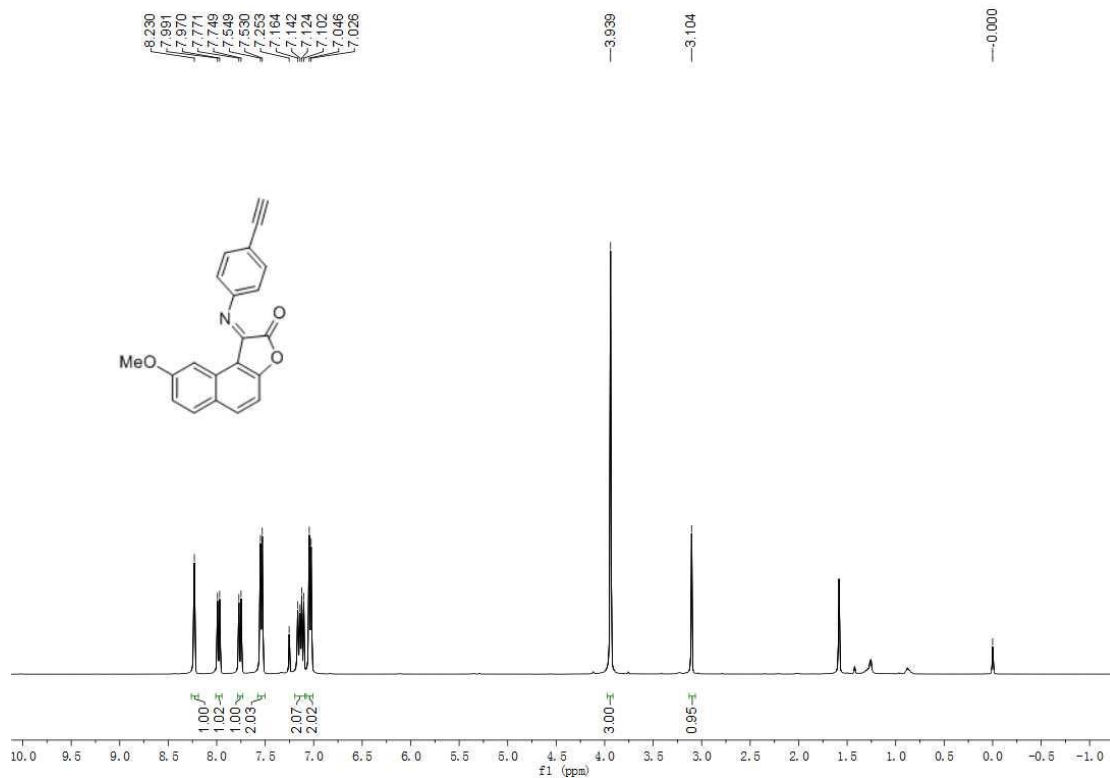
¹H NMR Spectrum of ethyl (Z)-4-((8-methoxy-2-oxonaphtho[2,1-b]furan-1(2H)-ylidene)amino)benzoate (**4zc**)



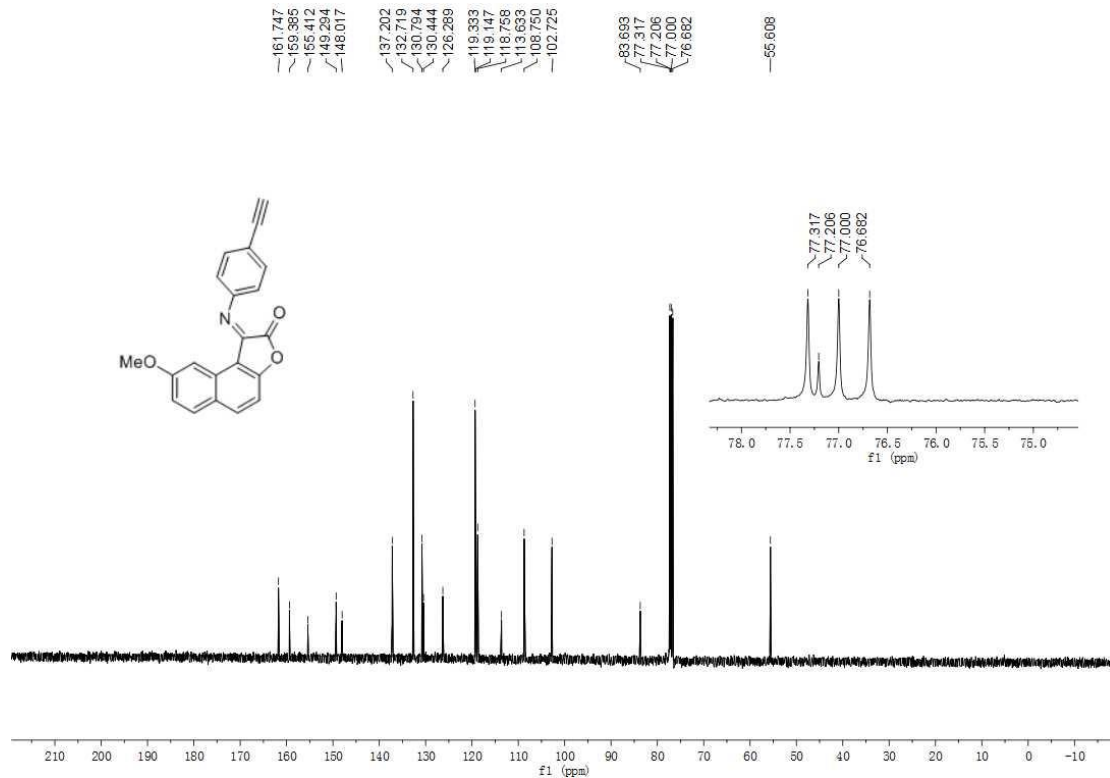
¹³C NMR Spectrum of ethyl (Z)-4-((8-methoxy-2-oxonaphtho[2,1-b]furan-1(2H)-ylidene)amino)benzoate (**4zc**)



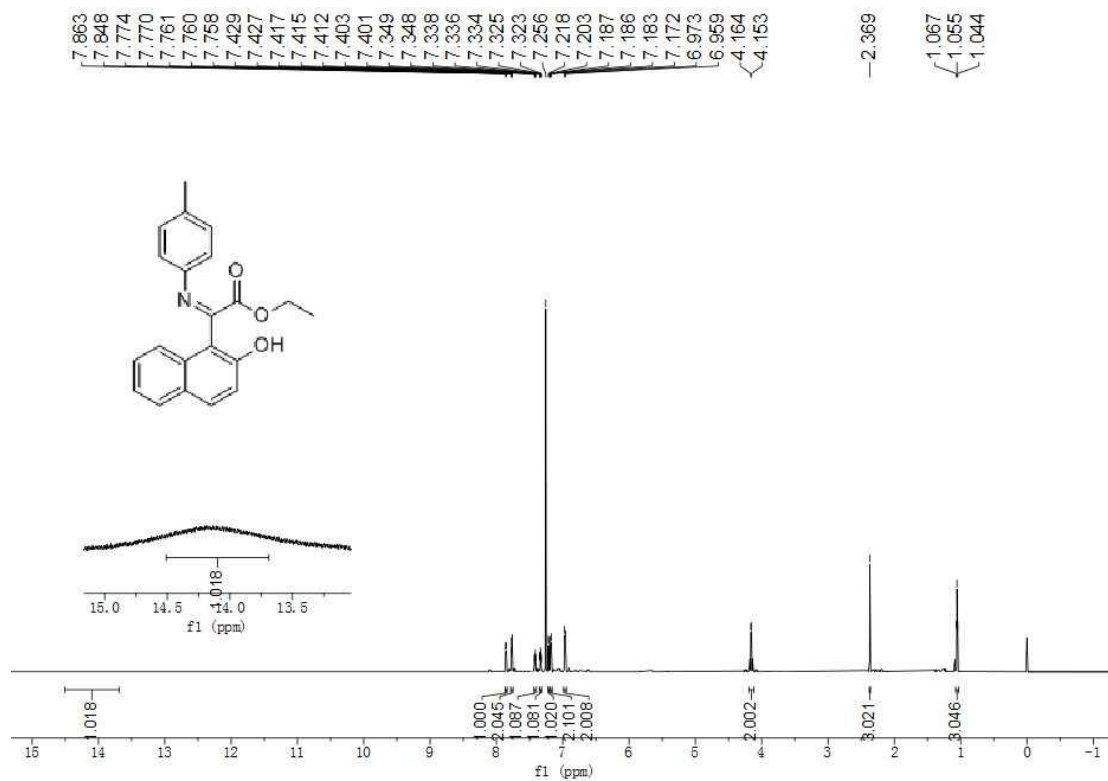
¹H NMR Spectrum of (Z)-1-((4-ethynylphenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4zd)



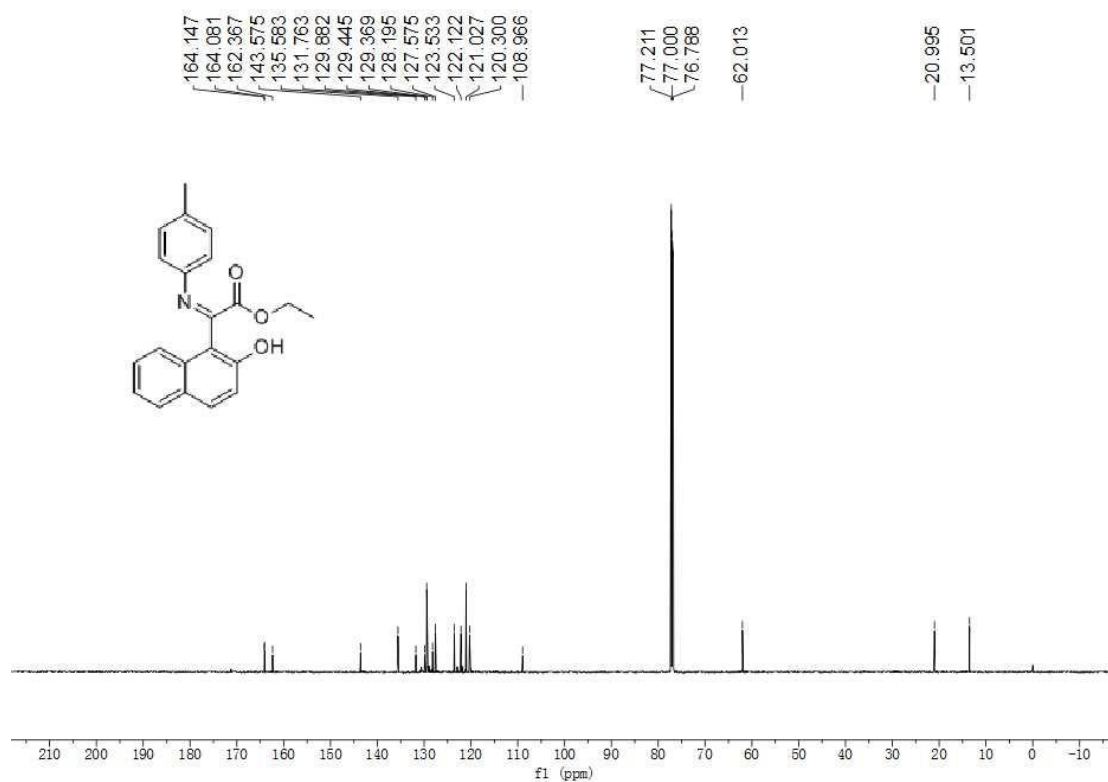
¹³C NMR Spectrum of (Z)-1-((4-ethynylphenyl)imino)-8-methoxynaphtho[2,1-b]furan-2(1H)-one (4zd)



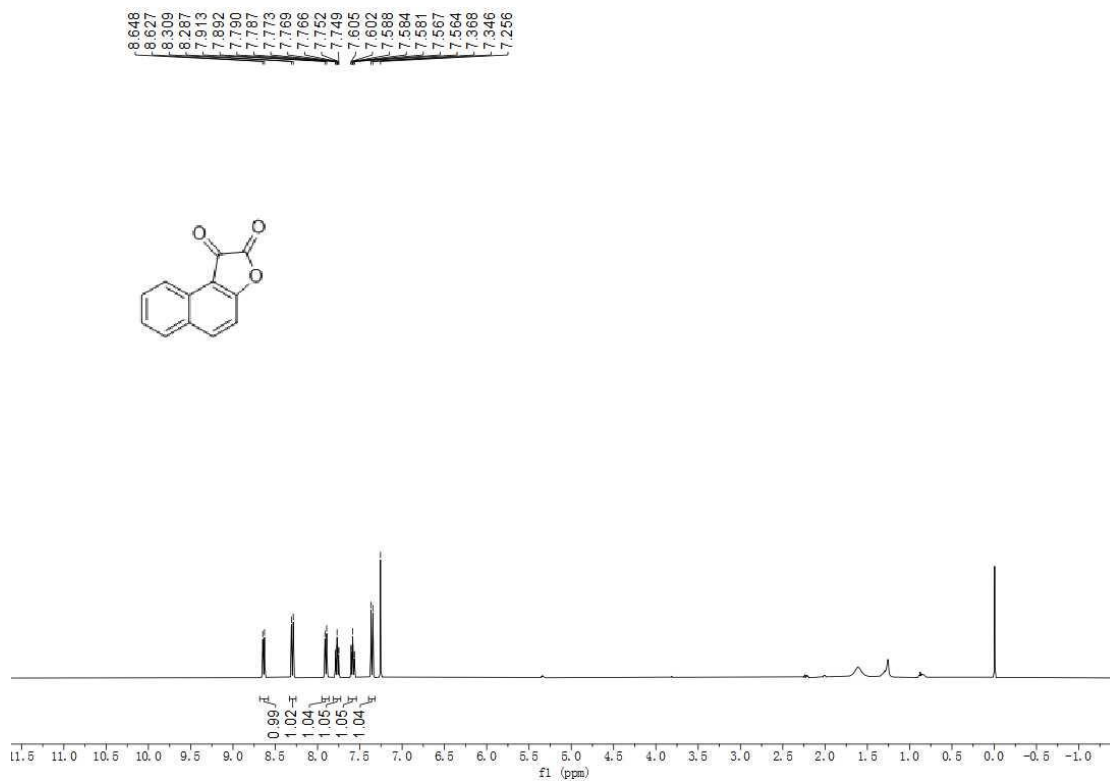
¹H NMR Spectrum of ethyl (Z)-2-(2-hydroxynaphthalen-1-yl)-2-(p-tolylimino)acetate (**5a**)



¹³C NMR Spectrum of ethyl (Z)-2-(2-hydroxynaphthalen-1-yl)-2-(p-tolylimino)acetate (**5a**)



¹H NMR Spectrum of naphtho[2,1-*b*]furan-1,2-dione (**6a**)



¹³C NMR Spectrum of naphtho[2,1-*b*]furan-1,2-dione (**6a**)

