Electronic Supplementary Information

Copper-iodine Co-catalyzed C−H Aminoalkenylation of Indoles via Temperature-Controlled Selectivity Switch: facile synthesis of 2-azolyl-3-alkenylindoles

Xiaoxiang Zhang, Wenhua Yu, Yiyu Nie, Yingying Zhang, Xiaoting Gu, Wanxing Wei, Zhuan Zhang, and Taoyuan Liang*

Guangxi Key Laboratory of Electrochemical Energy Materials, School of Chemistry and Chemical Engineering, Guangxi University, Nanning, Guangxi 530004, People's Republic of China.

*E-mail: taoyuanliang@gxu.edu.cn

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1. General information.

All the obtained products were characterized by melting points (m.p.), ^{1}H-NMR, ^{13}C-NMR. Melting points were measured on an Electrothemal SGW-X4 microscopy digital melting point apparatus and are uncorrected; the ^{1}H NMR (500 MHz) and ^{13}C NMR (125 MHz) spectroscopic data were recorded with $CDCl_3$ or [D6] DMSO as the solvent and TMS as the internal standard. Chemical shifts were reported in parts per million (ppm, δ) downfield from tetramethylsilane. Proton coupling patterns are described as singlet (s), doublet (d), triplet (t), multiplet (m); TLC was performed using commercially prepared 100-400 mesh silica gel plates (GF254), and visualization effected at 254 nm; Unless otherwise stated, all the reagents were purchased from commercial sources, used without further purification.

All the reagents were purchased from Bide Pharmatech Ltd and Energy Chemical. All solvents were purchased from Greagent (Shanghai Titansci incorporated company) and used without further purification. All reactions were heated by metal sand bath (WATTCAS, LAB-500, https://www.wattcas.com). Column chromatography was performed on silica gel (200-300 mesh). Reactions were monitored by using thin layer chromatography (TLC) (Qingdao Jiyida silica gel reagent factory GF254).

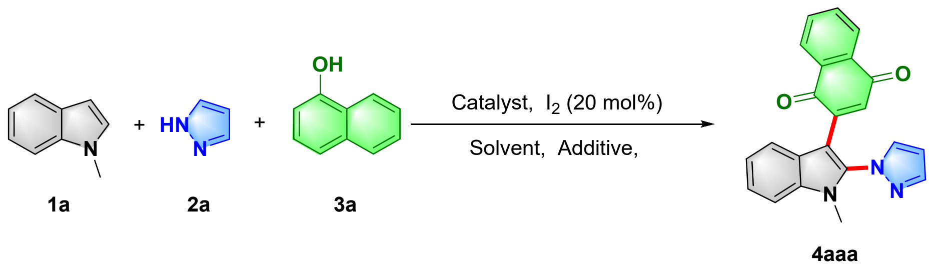
2. Substrates preparation.

General Procedure for the preparation of *N*-substituted indole derivatives (1b-1n)[1]:

Procedure for 4-chloro-1-methyl-1*H*-indole (**1j**): To a suspended solution of NaH (0.55 g, 65% dispersion in mineral oil, 15.0 mmol) in DMF (5.0 mL), 4-chloro-1*H*-indole (1.51 g, 10.0 mmol) in DMF (5.0 mL) was added dropwise at 0 °C. The heterogeneous mixture was stirred at 0 °C for 15 min and 1 h at room temperature. The mixture was then cooled to 0 °C, treated with iodomethane (0.83 mL, 13.0 mmol), and allowed to warm to room temperature. After 30 min, the reaction mixture was cooled to 0 °C, quenched with saturated NH_4Cl (20.0 mL), and extracted with ether (3 × 20.0 mL). The organic layers were combined, washed with brine, dried over anhydrous Na_2SO_4 and concentrated in vacuo. The resulting oil was purified by column chromatography on silica gel (petroleum ether) afforded **1j** as a yellow oil. Similarly, the other *N*-substituted indole derivatives were prepared from their corresponding indoles and halides.

3. Optimization of reaction conditions.

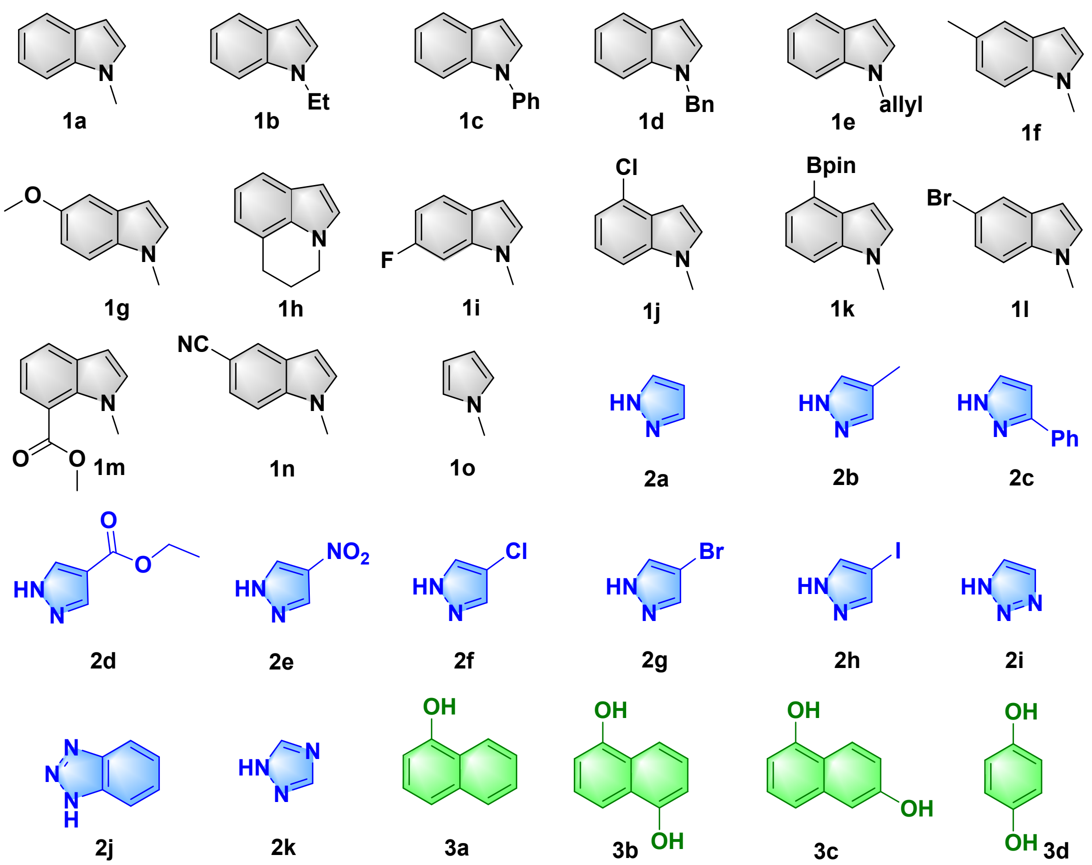
Table S1. Optimization of the reaction conditions.[a]



Entry	Catalyst	Solvent	Additive	**4aaa**, Yield (%)[b]
1.	CuI	CH_3CN	-	63
2.	$CuBr_2$	CH_3CN	-	19
3.	$Cu(OTf)_2$	CH_3CN	-	trace
4.	CuCl	CH_3CN	-	59
5.	CuBr	CH_3CN	-	62
6.	$CuCl_2$	CH_3CN	-	31
7.	CuI	THF	-	26
8.	CuI	DCE	-	39
9.	CuI	DMF	-	55
10.	CuI	DMSO	-	17
11.	CuI	1,4-dioxane	-	69
12.	CuI	1,4-dioxane/CH_3CN	-	(61, 75, 68)[c]
13.	CuI	1,4-dioxane/CH_3CN	-	(41, 65)[d]
14.	CuI	1,4-dioxane/CH_3CN	-	(65, 73)[e]
15.	CuI	1,4-dioxane/CH_3CN	-	(15, 19, 54)[f]
16.	CuI	1,4-dioxane/CH_3CN	$AlCl_3$	51
17.	CuI	1,4-dioxane/CH_3CN	$ZnCl_2$	46
18.	CuI	1,4-dioxane/CH_3CN	BF_3OEt_2	73
19.	CuI	1,4-dioxane/CH_3CN	NaOTf	42
20.	CuI	1,4-dioxane/CH_3CN	$Zn(OTf)_2$	(79, 83, 71)[g]
21.	CuI	1,4-dioxane/CH_3CN	$Zn(OTf)_2$	27[h]

[a]Reaction conditions, unless specified otherwise: 1a (0.25 mmol), 2a (0.75 mmol), 3a (0.375 mmol), and catalyst (20 mol%), I_2 (20 mol%), additive (20 mol%), solvent (3.0 mL) were stirred at 60 °C under O_2 for 12 h; then, the reaction mixture was heated to 100 °C for another 12 h; [b]isolated yield; cthe mixed solutions of 1,4-dioxane and CH_3CN (v/v = 1/1, 2/1, 1/2); [d]yields are with respect to the use of 10 mol% and 30 mol% of catalyst, respectively; [e]yields are with respect to the use of 10 mol% and 30 mol% of I_2, respectively; [f]yields are with respect to the use of 20 mol% NaI, KI and NIS instead of I_2, respectively; [g]yields are with respect to the use of 10 mol%, 20 mol% and 30 mol% of additive, respectively; [h]under air.

Scheme S1. **Substrates employed for the synthesis of 2-azolyl-3-alkenylindoles.**



4. Typical procedure for the synthesis of 4aaa.

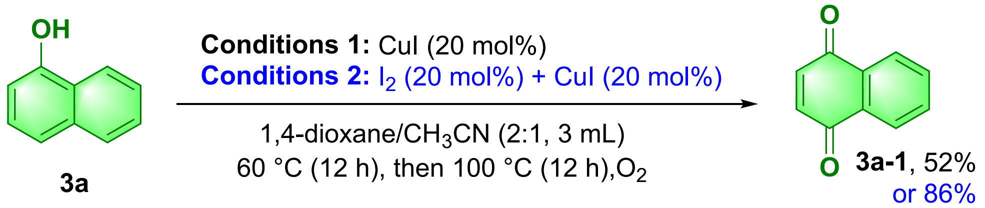
The mixture of 1-methylindole **1a** (32.8 mg, 0.25 mmol), **2a** (51.0 mg, 0.75 mmol), **3a** (54.0 mg, 0.375 mmol), and CuI (20 mol%), I_2 (20 mol%), $Zn(OTf)_2$ (20 mol%), 1,4-dioxane (2.0 mL) and CH_3CN (1.0 mL) were stirred at 60 °C under O_2 for 12 h; then, the reaction mixture was heated to 100 °C for another 12 h. The resulting mixture was concentrated by removing the solvent under vacuum, and the residue was purified by preparative TLC on silica gel by using petroleum ether/dichloromethane (1:1) as the eluent to give **4aaa** as a red solid (73.7 mg, 83% yield).

5. Synthetic utility.

The mixture of 1*H*-indole **1p** (29.3 mg, 0.25 mmol), **2a** (51.0 mg, 0.75 mmol), **3a** (54.0 mg, 0.375 mmol), and CuI (20 mol%), I_2 (20 mol%), $Zn(OTf)_2$ (20 mol%), 1,4-dioxane (2.0 mL) and CH_3CN (1.0 mL) were stirred at 60 °C under O_2 for 12 h; then, the reaction mixture was heated to 100 °C for another 12 h. The resulting mixture was concentrated by removing the solvent under vacuum, and the residue was purified by preparative TLC on silica gel by using petroleum ether/dichloromethane (1:1) as the eluent to give **4paa** as a red solid (48.3 mg, 57% yield).

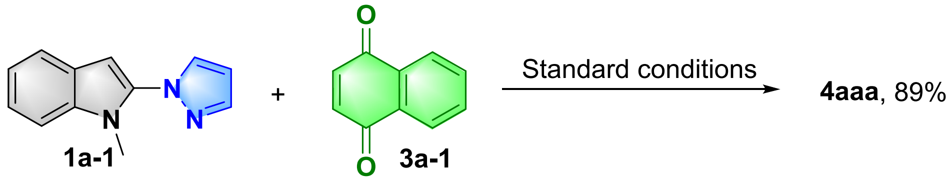
6. Control experiments.

(1) Conditions 1: 1-naphthol **3a** (36.0 mg, 0.25 mmol) and CuI (20 mol%), 1,4-dioxane (2.0 mL) and CH_3CN (1.0 mL) were stirred at 60 °C under O_2 for 12 h, then, the reaction mixture was heated to 100 °C for another 12 h. The reaction mixture was purified by preparative TLC on silica eluting with petroleum ether/dichloromethane (1:1) to give product **3a-1** (20.5 mg, 52% yield). **Conditions 2**: 1-naphthol **3a** (36.0 mg, 0.25 mmol) and CuI (20 mol%), I_2 (20 mol%), 1,4-dioxane (2.0 mL) and CH_3CN (1.0 mL) were stirred at 60 °C under O_2 for 12 h, then, the reaction mixture was heated to 100 °C for another 12 h. The reaction mixture was purified by preparative TLC on silica eluting with petroleum ether/dichloromethane (1:1) to give product **3a-1** (34.0 mg, 86% yield).

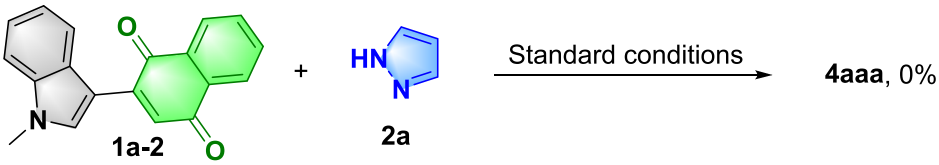


(2) The preparation of **1a-1** was similar to the literature procedures.[2] Under the optimized reaction conditions, the reaction of **1a-1** (49.3 mg, 0.25 mmol) and **3a-1** (39.5 mg, 0.25 mmol) were carried. Then, the reaction mixture was purified by preparative

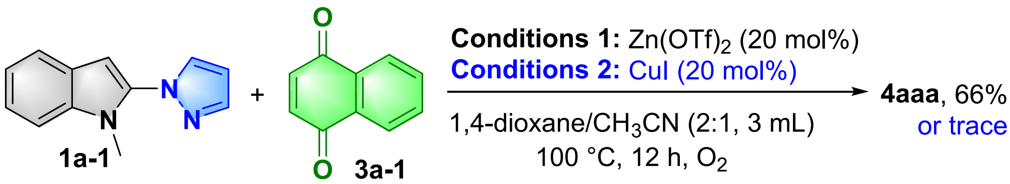
TLC on silica eluting with petroleum ether/dichloromethane (1:1) to give product **4aaa** as red solid (78.6 mg, 89% yield).



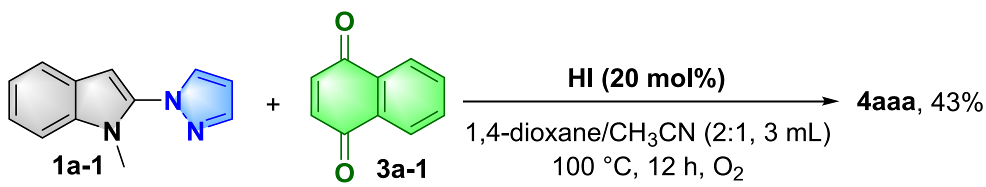
(3) The preparation of **1a-2** was similar to the literature procedures.[3] Under the optimized reaction conditions, the reaction of **1a-2** (71.8 mg, 0.25 mmol) and **2a** (51.0 mg, 0.75 mmol) were carried. Then, the crude reaction mixture was analyzed by TLC, and **4aaa** was not observed.



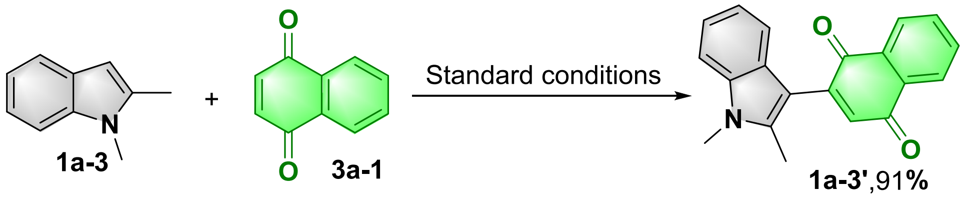
(4) Conditions 1: **1a-1** (49.3 mg, 0.25 mmol), **3a-1** (39.5 mg, 0.25 mmol), and $Zn(OTf)_2$ (20 mol%), 1,4-dioxane (2.0 mL) and CH_3CN (1.0 mL) were stirred at 100 °C under O_2 for 12 h. Then, the reaction mixture was purified by preparative TLC on silica eluting with petroleum ether/dichloromethane (1:1) to give product **4aaa** (58.2 mg, 66% yield). **Conditions 2**: **1a-1** (49.3 mg, 0.25 mmol), **3a-1** (39.5 mg, 0.25 mmol) and CuI (20 mol%), 1,4-dioxane (2.0 mL) and CH_3CN (1.0 mL) were stirred at 100 °C under O_2 for 12 h. Then, the crude reaction mixture was analyzed by TLC, and only a trace of **4aaa** was observed.



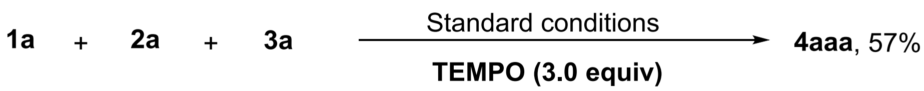
(5) **1a-1** (49.3 mg, 0.25 mmol), **3a-1** (39.5 mg, 0.25 mmol), and HI (20 mol%), 1,4-dioxane (2.0 mL) and CH_3CN (1.0 mL) were stirred at 100 °C under O_2 for 12 h. Then, the reaction mixture was purified by preparative TLC on silica eluting with petroleum ether/dichloromethane (1:1) to give product **4aaa** (37.9 mg, 43% yield).



(6) Under the optimized reaction conditions, the reaction of **1a-3** (36.3 mg, 0.25 mmol) and **3a-1** (39.5 mg, 0.25 mmol) were carried. Then, the reaction mixture was purified by preparative TLC on silica eluting with petroleum ether/dichloromethane (1:1) to give product **1a-3'** as purple solid (68.5 mg, 91% yield).



(7) Under the optimized reaction conditions, the reaction of **1a** (33.3 mg, 0.25 mmol) and **2a** (51.0 mg, 0.75 mmol), **3a** (54.0 mg, 0.375 mmol) and **TEMPO** (117.2 mg, 0.75 mmol) were carried. Then, the reaction mixture was purified by preparative TLC on silica eluting with petroleum ether/dichloromethane (1:1) to give product **4aaa** as red solid (50.3 mg, 57% yield).



7. Single crystal X-ray diffraction of 4jaa.

Red block-like single crystals of **4jaa** were grown by layering a dichlormethane solution with *n*-hexane at ambient temperature. X-Ray diffraction data of one these crystals were collected on a R-AXIS SPIDER diffractometer. The measurements were performed with Mo-Kα radiation (λ = 0.71073 Å). Data were collected at 153(2) K, using the *ω*- and *φ*- scans to a maximum θ value of 25.242°. The data were refined by full-matrix least-squares techniques on F^2 with SHELXTL-2014. And the structures were solved by direct methods SHELXS-2014. All the non-hydrogen atoms were refined anisotropically. The hydrogen atoms were included at geometrically idealized positions. And an ORTEP representation of the structure is shown below.

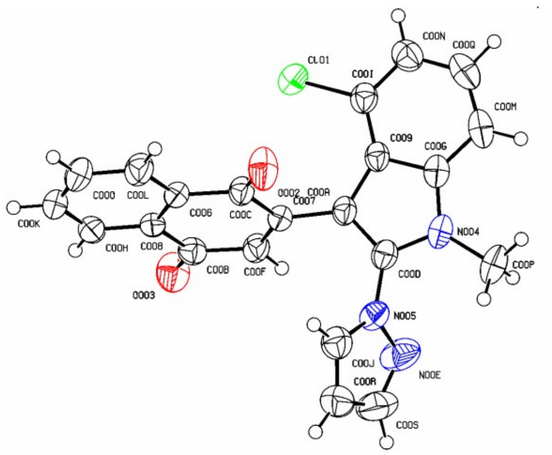


Figure S1. ORTEP drawing of **4jaa** with the numbering scheme.

Table S2. Crystal data and structure refinement for **4jaa**

Identification code	4jaa	
Empirical formula	$C_{22}H_{14}ClN_3O_2$	
Formula weight	387.81	
Temperature	153(2) K	
Crystal system	Monoclinic	
Space group	$P2_1/n$	
Unit cell dimensions	a = 12.4017(8) Å	α = 90°.
	b = 10.1700(9) Å	β = 101.867(6)°.
	c = 14.9975(10) Å	γ = 90°.
Volume	1851.1(2) $Å^3$	
Z	4	
F(000)	800.0	
Crystal size	0.15x 0.11 x 0.08 mm^3	
2^{Θ} range for data collection	1.945 to 29.544°	
Index ranges	-16<=h<=9, -12<=k<=10, -14<=l<=19	
Reflections collected	9344	
Independent reflections	4308 [R(int) = 0.0261]	
Data / restraints / parameters	4308 / 0 / 254	
Goodness-of-fit on F^2	1.045	
Final R indices [I>=2^{σ} (I)]	R1 = 0.0571, wR2 = 0.1150	
Final R indices (all data)	R1 = 0.0910, wR2 = 0.1330	
Largest diff. peak and hole	0.240 and -0.290 e.$Å^{-3}$	

Table S3. Fractional Atomic Coordinates (×10^4) and Equivalent Isotropic Displacement Parameters ($Å^2$×10^3) for **4jaa**. U(eq) is defined as 1/3 of of the trace of the orthogonalised U^{ij} tensor.

Atom	*x*	*y*	*z*	U(eq)
Cl(01)	8100(1)	2518(1)	4345(1)	56(1)
O(002)	7817(1)	5893(2)	4261(1)	67(1)
O(003)	8068(2)	4250(2)	939(1)	64(1)
N(004)	4332(1)	4401(2)	3652(1)	46(1)
N(005)	4561(2)	5753(2)	2376(1)	48(1)
C(006)	8891(2)	5802(2)	3132(1)	38(1)
C(007)	6982(2)	4760(2)	2926(1)	37(1)
C(008)	8932(2)	5438(2)	2240(1)	37(1)
C(009)	6046(2)	3584(2)	4113(1)	39(1)
C(00A)	6039(2)	4418(2)	3343(1)	39(1)
C(00B)	8005(2)	4697(2)	1680(1)	41(1)
C(00C)	7902(2)	5505(2)	3513(1)	40(1)
C(00D)	4984(2)	4875(2)	3085(1)	41(1)
N(00E)	3734(2)	5374(3)	1675(2)	72(1)
C(00F)	7031(2)	4452(2)	2068(1)	42(1)
C(00G)	4971(2)	3606(2)	4290(1)	43(1)
C(00H)	9851(2)	5765(2)	1887(2)	46(1)
C(00I)	6812(2)	2731(2)	4626(1)	44(1)
C(00J)	4926(2)	6968(3)	2241(2)	56(1)
C(00K)	10716(2)	6435(3)	2416(2)	56(1)
C(00L)	9772(2)	6476(3)	3656(2)	54(1)
C(00M)	4681(2)	2871(3)	4991(2)	55(1)
C(00N)	6538(2)	1989(3)	5313(2)	53(1)
C(00O)	10677(2)	6788(3)	3295(2)	61(1)
C(00P)	3170(2)	4705(3)	3605(2)	62(1)
C(00Q)	5484(2)	2091(3)	5493(2)	60(1)
C(00R)	4338(2)	7415(3)	1440(2)	67(1)
C(00S)	3619(3)	6398(4)	1121(2)	77(1)

Table S4. Anisotropic Displacement Parameters (Å^2×10^3) for **4jaa**. The Anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2a^{*2}U_{11}+2hka^*b^*U_{12}+\ldots]$.

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Cl(01)	39(1)	68(1)	58(1)	5(1)	7(1)	3(1)
O(002)	59(1)	94(2)	51(1)	-29(1)	22(1)	-26(1)

O(003)	66(1)	80(1)	50(1)	-21(1)	21(1)	-8(1)
N(004)	32(1)	52(1)	56(1)	-10(1)	14(1)	-5(1)
N(005)	38(1)	52(1)	50(1)	-2(1)	1(1)	-1(1)
C(006)	31(1)	40(1)	41(1)	3(1)	6(1)	0(1)
C(007)	32(1)	39(1)	39(1)	3(1)	6(1)	-1(1)
C(008)	34(1)	36(1)	40(1)	8(1)	9(1)	5(1)
C(009)	35(1)	43(1)	39(1)	-5(1)	9(1)	-9(1)
C(00A)	33(1)	44(1)	40(1)	-3(1)	8(1)	-7(1)
C(00B)	44(1)	42(1)	38(1)	-1(1)	9(1)	1(1)
C(00C)	35(1)	46(2)	40(1)	-3(1)	8(1)	-5(1)
C(00D)	35(1)	43(2)	44(1)	-5(1)	6(1)	-5(1)
N(00E)	61(1)	71(2)	70(2)	-4(1)	-18(1)	-5(1)
C(00F)	36(1)	49(2)	41(1)	-3(1)	4(1)	-7(1)
C(00G)	40(1)	46(2)	47(1)	-10(1)	14(1)	-7(1)
C(00H)	42(1)	50(2)	50(1)	13(1)	16(1)	10(1)
C(00I)	40(1)	50(2)	42(1)	-6(1)	7(1)	-7(1)
C(00J)	52(2)	54(2)	63(2)	2(1)	10(1)	-4(1)
C(00K)	33(1)	69(2)	67(2)	21(1)	13(1)	1(1)
C(00L)	41(1)	67(2)	52(1)	-3(1)	5(1)	-9(1)
C(00M)	54(2)	58(2)	59(2)	-9(1)	29(1)	-16(1)
C(00N)	59(2)	51(2)	50(1)	2(1)	10(1)	-8(1)
C(00O)	35(1)	75(2)	70(2)	5(1)	1(1)	-14(1)
C(00P)	35(1)	72(2)	83(2)	-12(2)	19(1)	-1(1)
C(00Q)	76(2)	59(2)	50(1)	2(1)	26(1)	-15(2)
C(00R)	64(2)	68(2)	71(2)	18(2)	18(1)	10(2)
C(00S)	70(2)	92(3)	61(2)	10(2)	-6(1)	18(2)

Table S5. Bond Lengths for **4jaa**.

Atom	Atom	Length/Å	Atom	Atom	Length/Å
Cl(01)	C(00I)	1.748(2)	C(00F)	H(00F)	0.9500
O(002)	C(00C)	1.214(2)	C(00G)	C(00M)	1.395(3)
O(003)	C(00B)	1.219(2)	C(00H)	H(00H)	0.9500
N(004)	C(00D)	1.376(3)	C(00H)	C(00K)	1.377(3)
N(004)	C(00G)	1.373(3)	C(00I)	C(00N)	1.376(3)
N(004)	C(00P)	1.461(3)	C(00J)	H(00J)	0.9500

N(005)	C(00D)	1.405(3)	C(00J)	C(00R)	1.349(4)
N(005)	N(00E)	1.365(3)	C(00K)	H(00K)	0.9500
N(005)	C(00J)	1.345(3)	C(00K)	C(00O)	1.376(4)
C(006)	C(008)	1.400(3)	C(00L)	H(00L)	0.9500
C(006)	C(00C)	1.486(3)	C(00L)	C(00O)	1.379(3)
C(006)	C(00L)	1.389(3)	C(00M)	C(00M)	0.9500
C(007)	C(00A)	1.477(3)	C(00M)	C(00M)	1.371(4)
C(007)	C(00C)	1.496(3)	C(00N)	C(00N)	0.9500
C(007)	C(00F)	1.338(3)	C(00N)	C(00N)	1.392(4)
C(008)	C(00B)	1.482(3)	C(00O)	C(00O)	0.9500
C(008)	C(00H)	1.391(3)	C(00P)	C(00P)	0.9800
C(009)	C(00A)	1.431(3)	C(00P)	C(00P)	0.9800
C(009)	C(00G)	1.412(3)	C(00P)	C(00P)	0.9800
C(009)	C(00I)	1.395(3)	C(00Q)	C(00Q)	0.9500
C(00A)	C(00D)	1.367(3)	C(00R)	C(00R)	0.9500
C(00B)	C(00F)	1.466(3)	C(00R)	C(00R)	1.385(4)
N(00E)	C(00S)	1.322(4)	C(00S)	C(00S)	0.9500

Table S6. Bond Angles for **4jaa**.

Atom	Atom	Atom	Angle/°	Atom	Atom	Atom	Angle/°
C(00D)	-N(004)	C(00P)	126.5(2)	C(00K)	C(00H)	C(008)	120.1(2)
C(00G)	N(004)	C(00D)	107.98(18)	C(00K)	C(00H)	H(00H)	120.0
C(00G)	N(004)	C(00P)	125.5(2)	C(009)	C(00I)	Cl(01)	119.82(17)
N(00E)	N(005)	C(00D)	120.5(2)	C(00N)	C(00I)	Cl(01)	119.41(19)
C(00J)	N(005)	C(00D)	127.8(2)	C(00N)	C(00I)	C(009)	120.6(2)
C(00J)	N(005)	N(00E)	111.5(2)	N(005)	C(00J)	H(00J)	126.1
C(008)	C(006)	C(00C)	120.60(18)	N(005)	C(00J)	C(00R)	107.9(2)
C(00L)	C(006)	C(008)	119.47(19)	C(00R)	C(00J)	H(00J)	126.1
C(00L)	C(006)	C(00C)	119.91(19)	C(00H)	C(00K)	H(00K)	119.9
C(00A)	C(007)	C(00C)	116.46(17)	C(00O)	C(00K)	C(00H)	120.3(2)
C(00F)	C(007)	C(00A)	123.26(19)	C(00O)	C(00K)	H(00K)	119.9
C(00F)	C(007)	C(00C)	120.27(19)	C(006)	C(00L)	H(00L)	120.0
C(006)	C(008)	C(00B)	119.95(18)	C(00O)	C(00L)	C(006)	120.0(2)
C(00H)	C(008)	C(006)	119.6(2)	C(00O)	C(00L)	H(00L)	120.0
C(00H)	C(008)	C(00B)	120.41(19)	C(00G)	C(00M)	H(00M)	121.5
C(00G)	C(009)	C(00A)	107.07(19)	C(00Q)	C(00M)	C(00G)	117.0(2)
C(00I)	C(009)	C(00A)	134.69(19)	C(00Q)	C(00M)	H(00M)	121.5
C(00I)	C(009)	C(00G)	117.9(2)	C(00I)	C(00N)	H(00N)	120.3

C(009)	C(00A)	C(007)	127.40(19)	C(00I)	C(00N)	C(00Q)	119.4(2)
C(00D)	C(00A)	C(007)	126.5(2)	C(00Q)	C(00N)	H(00N)	120.3
C(00D)	C(00A)	C(009)	106.02(18)	C(00K)	C(00O)	C(00L)	120.5(2)
O(003)	C(00B)	C(008)	121.3(2)	C(00K)	C(00O)	H(00O)	119.7
O(003)	C(00B)	C(00F)	120.7(2)	C(00L)	C(00O)	H(00O)	119.7
C(00F)	C(00B)	C(008)	117.91(18)	N(004)	C(00P)	H(00A)	109.5
O(002)	C(00C)	C(006)	121.63(19)	N(004)	C(00P)	H(00B)	109.5
O(002)	C(00C)	C(007)	120.47(19)	N(004)	C(00P)	H(00C)	109.5
C(006)	C(00C)	C(007)	117.84(18)	H(00A)	C(00P)	H(00B)	109.5
N(004)	C(00D)	N(005)	121.0(2)	H(00A)	C(00P)	H(00C)	109.5
C(00A)	C(00D)	N(004)	110.9(2)	H(00B)	C(00P)	H(00C)	109.5
C(00A)	C(00D)	N(005)	128.1(2)	C(00M)	C(00Q)	C(00N)	122.8(2)
C(00S)	N(00E)	N(005)	103.2(2)	C(00M)	C(00Q)	H(00Q)	118.6
C(007)	C(00F)	C(00B)	123.1(2)	C(00N)	C(00Q)	H(00Q)	118.6
C(007)	C(00F)	H(00F)	118.5	C(00J)	C(00R)	H(00R)	127.8
C(00B)	C(00F)	H(00F)	118.5	C(00J)	C(00R)	C(00S)	104.3(3)
N(004)	C(00G)	C(009)	108.05(19)	C(00S)	C(00R)	H(00R)	127.8
N(004)	C(00G)	C(00M)	129.7(2)	N(00E)	C(00S)	C(00R)	113.1(3)
C(00M)	C(00G)	C(009)	122.2(2)	N(00E)	C(00S)	H(00S)	123.4
C(008)	C(00H)	H(00H)	120.0	C(00R)	C(00S)	H(00S)	123.4

Table S7. Torsion Angles for **4jaa**.

A	**B**	**C**	**D**	**Angle/°**	**A**	**B**	**C**	**D**	**Angle/°**
Cl(01)	C(00I)	C(00N)	C(00Q)	175.42(19)	C(00D)	N(005)	N(00E)	C(00S)	175.4(2)
O(003)	C(00B)	C(00F)	C(007)	170.4(2)	C(00D)	N(005)	C(00J)	C(00R)	175.0(2)
N(004)	C(00G)	C(00M)	C(00Q)	175.8(2)	N(00E)	N(005)	C(00D)	N(004)	63.6(3)
N(005)	N(00E)	C(00S)	C(00R)	0.0(3)	N(00E)	N(005)	C(00D)	C(00A)	119.3(3)
N(005)	C(00J)	C(00R)	C(00S)	0.1(3)	N(00E)	N(005)	C(00J)	C(00R)	0.1(3)
C(006)	C(008)	C(00B)	O(003)	172.3(2)	C(00F)	C(007)	C(00A)	C(009)	117.9(3)
C(006)	C(008)	C(00B)	C(00F)	4.3(3)	C(00F)	C(007)	C(00A)	C(00D)	64.5(3)
C(006)	C(008)	C(00H)	C(00K)	0.4(3)	C(00F)	C(007)	C(00C)	O(002)	173.1(2)
C(006)	C(00L)	C(00O)	C(00K)	0.0(4)	C(00F)	C(007)	C(00C)	C(006)	4.2(3)
C(007)	C(00A)	C(00D)	N(004)	177.0(2)	C(00G)	N(004)	C(00D)	N(005)	178.1(2)
C(007)	C(00A)	C(00D)	N(005)	0.3(4)	C(00G)	N(004)	C(00D)	C(00A)	0.6(3)
C(008)	C(006)	C(00C)	O(002)	174.7(2)	C(00G)	C(009)	C(00A)	C(007)	176.9(2)
C(008)	C(006)	C(00C)	C(007)	2.5(3)	C(00G)	C(009)	C(00A)	C(00D)	1.1(2)
C(008)	C(006)	C(00L)	C(00O)	0.3(4)	C(00G)	C(009)	C(00I)	Cl(01)	172.32(17)
C(008)	C(00B)	C(00F)	C(007)	6.2(3)	C(00G)	C(009)	C(00I)	C(00N)	3.3(3)
C(008)	C(00H)	C(00K)	C(00O)	0.1(4)	C(00G)	C(00M)	C(00Q)	C(00N)	1.7(4)
C(009)	C(00A)	C(00D)	N(004)	1.0(3)	C(00H)	C(008)	C(00B)	O(003)	7.1(3)

C(009)	C(00A)	C(00D)	N(005)	178.4(2)	C(00H)	C(008)	C(00B)	C(00F)	176.3(2)
C(009)	C(00G)	C(00M)	C(00Q)	1.6(4)	C(00H)	C(00K)	C(00O)	C(00L)	0.1(4)
C(009)	C(00I)	C(00N)	C(00Q)	0.3(4)	C(00I)	C(009)	C(00A)	C(007)	9.8(4)
C(00A)	C(007)	C(00C)	O(002)	6.0(3)	C(00I)	C(009)	C(00A)	C(00D)	172.2(2)
C(00A)	C(007)	C(00C)	C(006)	176.75(19)	C(00I)	C(009)	C(00G)	N(004)	173.84(19)
C(00A)	C(007)	C(00F)	C(00B)	174.8(2)	C(00I)	C(009)	C(00G)	C(00M)	4.1(3)
C(00A)	C(009)	C(00G)	N(004)	0.7(3)	C(00I)	C(00N)	C(00Q)	C(00M)	2.4(4)
C(00A)	C(009)	C(00G)	C(00M)	178.6(2)	C(00J)	N(005)	C(00D)	N(004)	121.9(3)
C(00A)	C(009)	C(00I)	Cl(01)	0.4(4)	C(00J)	N(005)	C(00D)	C(00A)	55.1(4)
C(00A)	C(009)	C(00I)	C(00N)	176.0(2)	C(00J)	N(005)	N(00E)	C(00S)	0.1(3)
C(00B)	C(008)	C(00H)	C(00K)	179.0(2)	C(00J)	C(00R)	C(00S)	N(00E)	0.0(4)
C(00C)	C(006)	C(008)	C(00B)	2.8(3)	C(00L)	C(006)	C(008)	C(00B)	178.9(2)
C(00C)	C(006)	C(008)	C(00H)	177.9(2)	C(00L)	C(006)	C(008)	C(00H)	0.5(3)
C(00C)	C(006)	C(00L)	C(00O)	178.1(2)	C(00L)	C(006)	C(00C)	O(002)	3.6(4)
C(00C)	C(007)	C(00A)	C(009)	63.1(3)	C(00L)	C(006)	C(00C)	C(007)	179.1(2)
C(00C)	C(007)	C(00A)	C(00D)	114.5(2)	C(00P)	N(004)	C(00D)	N(005)	0.6(3)
C(00C)	C(007)	C(00F)	C(00B)	6.1(4)	C(00P)	N(004)	C(00D)	C(00A)	178.2(2)
C(00D)	N(004)	C(00G)	C(009)	0.1(3)	C(00P)	N(004)	C(00G)	C(009)	178.9(2)
C(00D)	N(004)	C(00G)	C(00M)	177.8(2)	C(00P)	N(004)	C(00G)	C(00M)	3.4(4)

Table S8. Hydrogen Atom Coordinates (Å×10^4) and Isotropic Displacement Parameters (Å^2×10^3) for **4jaa**.

Atom	***x***	***y***	***z***	**U(eq)**
H(00F)	6405	4056	1693	51
H(00H)	9881	5525	1280	56
H(00J)	5496	7429	2638	68
H(00K)	11342	6656	2174	68
H(00L)	9752	6723	4263	65
H(00M)	3960	2909	5114	66
H(00N)	7064	1412	5662	64
H(00O)	11278	7248	3656	74
H(00A)	3017	5603	3378	93
H(00B)	3003	4630	4214	93
H(00C)	2711	4086	3191	93
H(00Q)	5313	1599	5985	71
H(00R)	4403	8240	1158	81
H(00S)	3096	6433	561	93

8. Reference.

1. S. K. Banjare, T. Nanda, B. V. Pati, G. K. D. Adhikari, J. Dutta and P. C. Ravikumar,

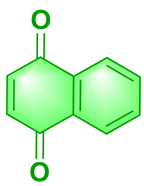
ACS Catal., 2021, **11**, 11579–11587.

2. D. Beukeaw, K. Udomsasporn and S. Yotphan, *J. Org. Chem.,* 2015, **80**, 3447–3454.

3. J. Jiang, S. S. K. Boominathan, W. Hu, C. Chen, J. K. Vandavasi, Y. Lin and J. Wang, *Eur. J. Org. Chem.,* 2016, **2016**, 2284–2289.

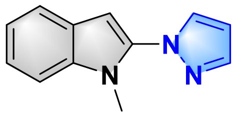
9. Analytic data of the obtained compounds.

(1) naphthalene-1,4-dione (3a-1)



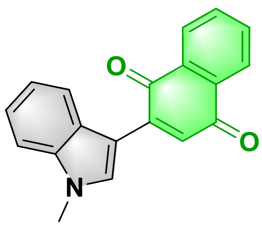
Known compound, ^{1}H NMR (500 MHz, Chloroform-d) δ 7.94–7.92 (m, 2H), 7.66–7.63 (m, 2H), 6.88 (d, *J* = 1.0 Hz, 2H).

(2) 1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-indole (1a-1)



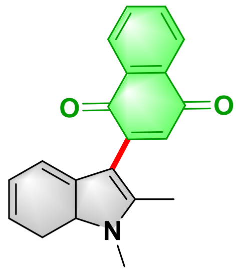
Known compound, ^{1}H NMR (500 MHz, Chloroform-d) δ 7.90 (d, *J* = 1.6 Hz, 1H), 7.79 (d, *J* = 2.3 Hz, 1H), 7.73 (d, *J* = 7.9 Hz, 1H), 7.43 (d, *J* = 8.1 Hz, 1H), 7.39 (t, *J* = 8.1 Hz, 1H), 7.28 (t, *J* = 8.0 Hz, 1H), 6.60 (s, 1H), 6.54 (t, *J* = 8.0 Hz, 1H), 3.75 (s, 3H).

(3) 2-(1-methyl-1*H*-indol-3-yl)naphthalene-1,4-dione (1a-2)



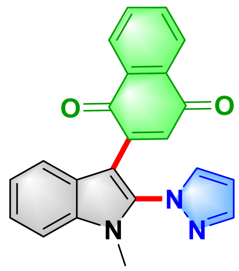
Known compound, ^{1}H NMR (500 MHz, Chloroform-d) δ 8.18–8.15 (m, 2H), 8.13 (d, *J* = 8.7 Hz, 1H), 8.01 (d, *J* = 6.5 Hz, 1H), 7.76–7.74 (m, 2H), 7.43 (s, 1H), 7.41 (d, *J* = 7.4 Hz, 1H), 7.35–7.31 (m, 2H), 3.89 (s, 3H).

(4) 2-(1,2-dimethyl-1*H*-indol-3-yl)naphthalene-1,4-dione (1a-3')



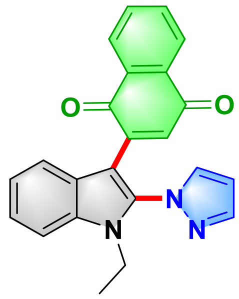
Known compound, ^{1}H NMR (500 MHz, Chloroform-d) δ 8.21 (d, *J* = 8.7 Hz, 1H), 8.16 (d, *J* = 8.6 Hz, 1H), 7.80 – 7.77 (m, 2H), 7.58 (d, *J* = 7.8 Hz, 1H), 7.34 (d, *J* = 8.1 Hz, 1H), 7.25 (t, *J* = 7.1 Hz, 1H), 7.19 (t, *J* = 7.0 Hz, 1H), 7.10 (s, 1H), 3.75 (s, 3H), 2.46 (s, 3H).

(5) 2-(1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4aaa)



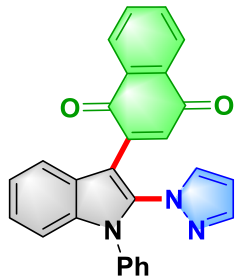
Red solid, (73.3 mg, 83% yield); m.p.: 148-150 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.09 (d, *J* = 8.7 Hz, 1H), 7.98 (d, *J* = 8.7 Hz, 1H), 7.80 (d, *J* = 1.6 Hz, 1H), 7.73–7.65 (m, 4H), 7.43 (d, *J* = 8.1 Hz, 1H), 7.39 (t, *J* = 7.6 Hz, 1H), 7.29 (t, *J* = 8.0 Hz, 1H), 6.98 (s, 1H), 6.46–6.43 (m, 1H), 3.68 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.87, 183.49, 142.70, 142.34, 135.30, 135.07, 135.05, 133.65, 133.48, 133.19, 132.57, 132.20, 126.87, 125.97, 125.12, 123.91, 121.89, 120.55, 110.23, 107.67, 104.04, 30.03. HRMS (ESI): Calcd. for $C_{22}H_{15}N_3O_2$ $[M+H]^+$: 354.1237; found: 354.1232.

(6) 2-(1-ethyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4baa)



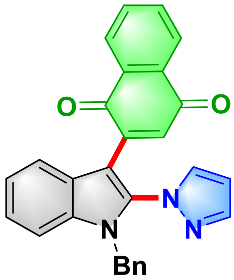
Red solid, (72.5 mg, 79% yield); m.p.: 153-155 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.09 (d, *J* = 8.8 Hz, 1H), 8.01 (d, *J* = 8.8 Hz, 1H), 7.83 (d, *J* = 1.6 Hz, 1H), 7.75–7.69 (m, 3H), 7.67 (d, *J* = 2.3 Hz, 1H), 7.47 (d, *J* = 8.3 Hz, 1H), 7.39 (t, *J* = 8.2 Hz, 1H), 7.31–7.28 (m, 1H), 6.97 (s, 1H), 6.47–6.43 (m, 1H), 4.14 (q, *J* = 7.2 Hz, 2H), 1.39 (t, *J* = 7.2 Hz, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.86, 183.49, 142.66, 142.27, 135.04, 134.51, 134.24, 133.66, 133.49, 133.19, 132.57, 132.18, 126.87, 125.95, 125.32, 123.84, 121.75, 120.79, 110.34, 107.60, 104.31, 38.71, 15.30. HRMS (ESI): Calcd. for $C_{23}H_{17}N_3O_2$ $[M+H]^+$: 368.1393; found: 368.1389.

(7) 2-(1-phenyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4caa)



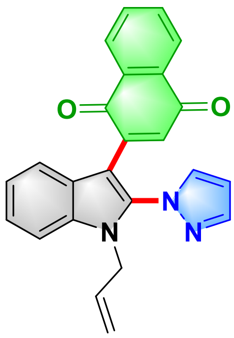
Red solid, (74.7 mg, 72% yield), m.p.: 184-186 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.14 (d, *J* = 7.7 Hz, 1H), 7.96 (d, *J* = 8.7 Hz, 1H), 7.79–7.74 (m, 2H), 7.72 (d, *J* = 7.5 Hz, 1H), 7.54 (d, *J* = 1.6 Hz, 1H), 7.49–7.44 (m, 3H), 7.39 (d, *J* = 2.3 Hz, 1H), 7.36–7.33 (m, 4H), 7.32 (d, *J* = 1.2 Hz, 1H), 7.25 (s, 1H), 6.27–6.25 (m, 1H). ^{13}C NMR (125 MHz, Chloroform-d) δ 138.18, 136.83, 136.43, 135.98, 129.50, 128.88, 128.62, 127.84, 127.71, 127.18, 126.91, 126.04, 122.56, 122.52, 120.66, 120.53, 120.35, 118.10, 110.40, 110.19, 104.28, 50.34, 48.06. HRMS (ESI): Calcd. for $C_{27}H_{17}N_3O_2$ $[M+H]^+$: 416.1393; found: 416.1388.

(8) 2-(1-benzyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4daa)



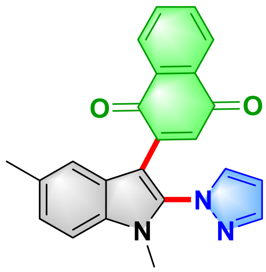
Red solid, (62.2 mg, 58% yield), m.p.: 148-150 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.11 (d, J = 7.0 Hz, 1H), 8.00 (d, J = 7.0 Hz, 1H), 7.79 (d, J = 1.6 Hz, 1H), 7.75–7.73 (m, 2H), 7.70 (d, J = 7.1 Hz, 1H), 7.52 (d, J = 2.3 Hz, 1H), 7.39 (d, J = 7.7 Hz, 1H), 7.35 (d, J = 6.3 Hz, 1H), 7.30–7.25 (m, 4H), 7.10 (d, J = 6.9 Hz, 2H), 7.06 (s, 1H), 6.39–6.37 (m, 1H), 5.35 (s, 2H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.86, 183.36, 142.62, 142.35, 136.40, 135.29, 134.97, 133.67, 133.52, 133.26, 132.56, 132.20, 128.86, 127.77, 126.87, 126.64, 125.99, 125.37, 124.11, 122.02, 120.63, 110.97, 107.69, 47.20. HRMS (ESI): Calcd. for $C_{28}H_{19}N_3O_2$ $[M+H]^+$: 430.1550; found: 430.1546.

(9) 2-(1-allyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4eaa)



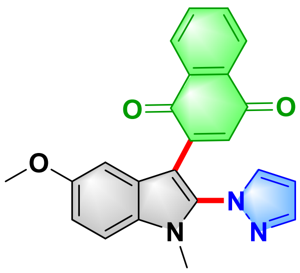
Red solid, (57.8 mg, 61% yield); m.p.: 136-138 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.10 (d, J = 7.2 Hz, 1H), 7.99 (d, J = 7.1 Hz, 1H), 7.78 (d, J = 1.1 Hz, 1H), 7.75–7.67 (m, 4H), 7.45–7.28 (m, 3H), 7.03 (s, 1H), 6.47–6.41 (m, 1H), 6.01–5.91 (m, 1H), 5.14 (d, J = 79.6 Hz, 2H), 4.70 (d, J = 10.0 Hz, 2H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.85, 183.37, 142.67, 142.27, 135.14, 134.74, 133.65, 133.50, 133.27, 132.56, 132.55, 132.18, 126.84, 125.96, 125.29, 123.96, 121.94, 120.61, 117.62, 110.78, 107.62, 104.39, 46.06. HRMS (ESI): Calcd. for $C_{24}H_{17}N_3O_2$ $[M+H]^+$: 380.1393; found: 380.1390.

(10) 2-(1,5-dimethyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4faa)



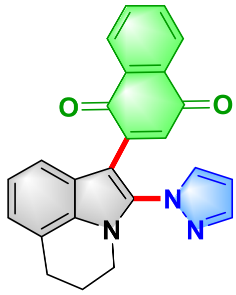
Red solid, (69.7 mg, 76% yield), m.p.: 211-214 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.09 (d, *J* = 8.8 Hz, 1H), 7.98 (d, *J* = 8.8 Hz, 1H), 7.80 (d, *J* = 1.7 Hz, 1H), 7.75–7.68 (m, 3H), 7.49 (s, 1H), 7.31 (d, *J* = 8.4 Hz, 1H), 7.21 (d, *J* = 9.6 Hz, 1H), 7.01 (s, 1H), 6.45 (t, *J* = 2.1 Hz, 1H), 3.66 (s, 3H), 2.50 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.93, 183.45, 142.93, 142.21, 135.08, 134.76, 133.69, 133.59, 133.43, 133.21, 132.60, 132.21, 131.40, 126.84, 125.92, 125.53, 125.24, 120.03, 109.96, 107.57, 30.06, 21.69. HRMS (ESI): Calcd. for $C_{23}H_{17}N_3O_2$ $[M+H]^+$: 368.1393; found: 368.1387.

(11) 2-(5-methoxy-1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4gaa)



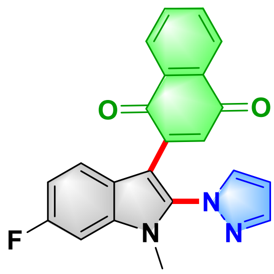
Red solid, (70.9 mg, 74% yield), m.p.: 192-194 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.09 (d, *J* = 7.3 Hz, 1H), 7.98 (d, *J* = 8.7 Hz, 1H), 7.79 (d, *J* = 1.6 Hz, 1H), 7.75–7.69 (m, 2H), 7.67 (d, *J* = 2.3 Hz, 1H), 7.33 (d, *J* = 8.9 Hz, 1H), 7.13 (d, *J* = 2.4 Hz, 1H), 7.04 (d, *J* = 11.4 Hz, 1H), 6.98 (s, 1H), 6.46–6.44 (m, 1H), 3.88 (s, 3H), 3.66 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.89, 183.46, 155.72, 142.93, 142.24, 135.30, 134.60, 133.62, 133.44, 133.15, 132.58, 132.20, 130.40, 126.85, 125.93, 125.56, 114.19, 111.17, 107.61, 103.78, 102.18, 55.95, 30.14. HRMS (ESI): Calcd. for $C_{23}H_{17}N_3O_3$ $[M+H]^+$: 384.1342; found: 384.1338.

(12) 2-(2-(1*H*-pyrazol-1-yl)-5,6-dihydro-4*H*-pyrrolo[3,2,1-*ij*]quinolin-1-yl)naphthalene-1,4-dione (4haa)



Red solid, (80.5 mg, 85% yield); m.p.: 167-169 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.10 (d, J = 7.3 Hz, 1H), 7.98 (d, J = 6.0 Hz, 1H), 7.79 (s, 1H), 7.76 –7.67 (m, 3H), 7.54 (d, J = 8.1 Hz, 1H), 7.20 (t, J = 7.9 Hz, 1H), 7.09 (d, J = 6.0 Hz, 1H), 7.03 (s, 1H), 6.47–6.43 (m, 1H), 4.14 (t, J = 20.0 Hz, 2H), 3.05 (t, J = 20.0 Hz, 2H), 2.30–2.23 (m, 2H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.90, 183.64, 143.28, 142.14, 134.29, 134.09, 133.58, 133.39, 132.68, 132.56, 132.26, 126.83, 125.90, 123.46, 122.59, 122.03, 120.88, 117.89, 107.67, 102.81, 42.60, 24.66, 22.45. HRMS (ESI): Calcd. for $C_{24}H_{17}N_3O_2$ $[M+H]^+$: 380.1393; found: 380.1389.

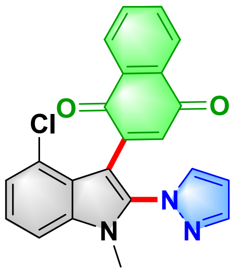
(13) 2-(6-fluoro-1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4iaa)



Red solid, (59.4 mg, 64% yield), m.p.: 225-227 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.09 (d, J = 7.2 Hz, 1H), 8.02 (d, J = 7.3 Hz, 1H), 7.82 (d, J = 1.7 Hz, 1H), 7.77–7.71 (m, 2H), 7.66 (d, J = 2.4 Hz, 1H), 7.61 (dd, J = 8.8, 5.2 Hz, 1H), 7.10 (d, J = 9.3 Hz, 1H), 7.07–7.02 (m, 1H), 6.91 (s, 1H), 6.46 (t, J = 2.1 Hz, 1H), 3.63 (s, 3H). ^{13}C

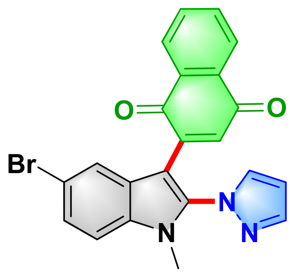
NMR (125 MHz, Chloroform-d) δ 184.75, 183.49, 142.47, 142.24, 135.39 (d, J = 12.5 Hz,), 135.29, 135.14 (d, J = 3.75 Hz), 133.77, 133.58, 133.17, 132.45, 132.11, 126.89, 126.01, 122.02 (d, J = 10.0 Hz) , 121.48, 110.74, 110.55, 107.77, 104.38, 96.71 (d, J = 26.3 Hz), 30.14. ^{19}F NMR (470 MHz, Chloroform-d) δ -116.96. HRMS (ESI): Calcd. for $C_{22}H_{14}FN_3O_2$ $[M+H]^+$: 372.1142; found: 372.1139.

(14) 2-(4-chloro-1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4jaa)



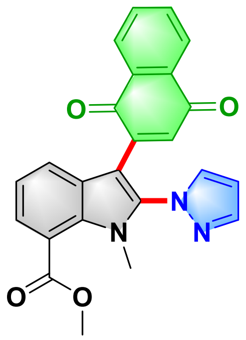
Red solid, (52.2 mg, 54% yield), m.p.: 216-218 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.16 (d, J = 9.0 Hz, 1H), 8.10 (d, J = 9.0 Hz, 1H), 7.82 (d, J = 1.7 Hz, 1H), 7.78–7.75 (m, 2H), 7.56 (d, J = 2.4 Hz, 1H), 7.35 (d, J = 7.8 Hz, 1H), 7.30 (d, J = 7.7 Hz, 1H), 7.21 (d, J = 7.5 Hz, 1H), 6.81 (s, 1H), 6.42 (t, J = 2.1 Hz, 1H), 3.64 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 185.05, 184.73, 143.24, 142.56, 137.14, 136.02, 134.60, 133.74, 133.72, 133.06, 132.41, 132.27, 127.02, 126.42, 126.13, 124.24, 123.13, 122.26, 109.00, 107.63, 104.49, 30.11. HRMS (ESI): Calcd. for $C_{22}H_{14}ClN_3O_2$ $[M+H]^+$: 388.0847; found: 388.0844.

(15) 2-(5-bromo-1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4laa)



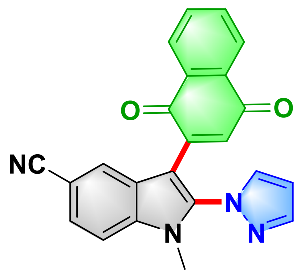
Red solid, (48.5 mg, 45% yield), m.p.: 242-244 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.09 (d, *J* = 7.4 Hz, 1H), 8.00 (d, *J* = 7.2 Hz, 1H), 7.81 (d, *J* = 1.7 Hz, 1H), 7.80 (d, *J* = 1.8 Hz, 1H), 7.76–7.70 (m, 2H), 7.68 (d, *J* = 2.4 Hz, 1H), 7.46 (d, *J* = 8.7 Hz, 1H), 7.30 (d, *J* = 8.7 Hz, 1H), 6.91 (s, 1H), 6.47 (t, *J* = 2.1 Hz, 1H), 3.68 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.70, 183.29, 142.56, 142.04, 135.72, 135.33, 133.88, 133.79, 133.60, 133.12, 132.36, 132.09, 126.93, 126.84, 126.61, 126.01, 123.11, 115.13, 111.76, 107.92, 103.53, 30.23. HRMS (ESI): Calcd. for $C_{22}H_{14}BrN_3O_2$ $[M+H]^+$: 432.0342; found: 432.0340.

(16) methyl3-(1,4-dioxo-1,4-dihydronaphthalen-2-yl)-1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-indole-7-carboxylate (4maa)



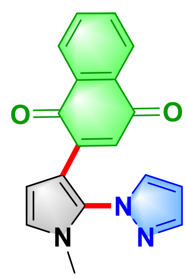
Red solid, (44.2 mg, 43% yield), m.p.: 205-207 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.09 (d, *J* = 8.7 Hz, 1H), 7.98 (d, *J* = 7.1 Hz, 1H), 7.84–7.80 (m, 2H), 7.77 (d, *J* = 1.8 Hz, 1H), 7.76 (d, *J* = 2.5 Hz, 1H), 7.75–7.68 (m, 2H), 7.31–7.28 (m, 1H), 7.01 (s, 1H), 6.49 (t, *J* = 2.7 Hz, 1H), 4.01 (s, 3H), 3.62 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.74, 183.21, 167.41, 142.47, 142.25, 136.99, 135.64, 133.73, 133.57, 133.38, 133.19, 132.42, 132.12, 127.30, 126.91, 126.87, 126.02, 124.62, 120.90, 117.26, 107.98, 104.66, 52.43, 34.02. HRMS (ESI): Calcd. for $C_{24}H_{17}N_3O_4$ $[M+H]^+$: 412.1291; found: 412.1286.

(17) 3-(1,4-dioxo-1,4-dihydronaphthalen-2-yl)-1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-indole-5-carbonitrile (4naa)



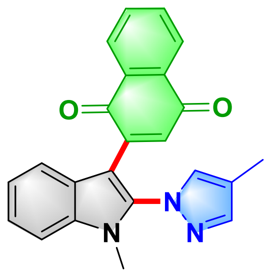
Red solid, (24.6 mg, 26% yield), m.p.: 267-268 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.11 (d, J = 9.0 Hz, 1H), 8.07 (d, J = 9.0 Hz, 1H), 8.02–8.00 (m, 1H), 7.87 (d, J = 1.4 Hz, 1H), 7.80–7.76 (m, 2H), 7.67 (s, 1H), 7.63 (d, J = 10.0 Hz, 1H), 7.52 (d, J = 8.6 Hz, 1H), 6.84 (s, 1H), 6.50 (s, 1H), 3.74 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.48, 183.36, 142.98, 141.21, 136.70, 136.64, 136.08, 134.04, 133.83, 133.07, 132.20, 132.03, 127.04, 126.57, 126.51, 126.16, 125.04, 119.92, 111.21, 108.29, 105.09, 104.85, 30.38. HRMS (ESI): Calcd. for $C_{23}H_{14}N_4O_2$ $[M+H]^+$: 379.1189; found: 379.1186.

(18) 2-(1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-pyrrol-3-yl)naphthalene-1,4-dione (4oaa)



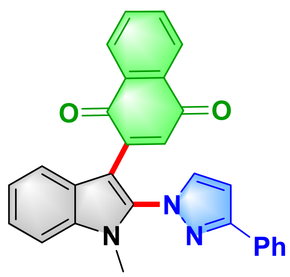
Black solid, (37.9 mg, 50% yield); m.p.: 234-236 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.19 (d, J = 8.9 Hz, 1H), 8.14 (d, J = 8.8 Hz, 1H), 7.81–7.79 (m, 3H), 7.69 (d, J = 2.1 Hz, 1H), 6.97 (s, 1H), 6.68 (d, J = 4.0 Hz, 1H), 6.49–6.48 (m, 1H), 6.35 (d, J = 4.0 Hz, 1H), 3.48 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.64, 184.10, 141.88, 139.43, 135.06, 133.99, 133.73, 132.77, 132.41, 132.30, 132.13, 127.07, 126.09, 126.02, 115.16, 106.96, 104.89, 33.50. HRMS (ESI): Calcd. for $C_{18}H_{13}N_3O_2$ $[M+H]^+$: 304.1080; found: 304.1076.

(19) 2-(1-methyl-2-(4-methyl-1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4aba)



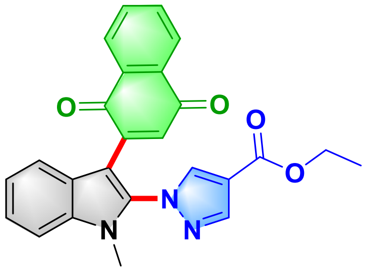
Red solid, (66.1 mg, 72% yield), m.p.: 190-192 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.10 (d, *J* = 7.1 Hz, 1H), 8.01 (d, *J* = 8.8 Hz, 1H), 7.76–7.71 (m, 2H), 7.69 (d, *J* = 8.0 Hz, 1H), 7.62 (s, 1H), 7.42 (d, *J* = 11.4 Hz, 2H), 7.38 (t, *J* = 7.0 Hz, 1H), 7.29 (t, *J* = 6.8 Hz, 1H), 6.99 (s, 1H), 3.69 (s, 3H), 2.13 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.92, 183.57, 143.25, 142.92, 135.45, 135.25, 134.79, 133.60, 133.45, 132.69, 132.24, 131.37, 126.82, 125.94, 125.16, 123.75, 121.80, 120.54, 118.23, 110.20, 103.76, 30.03, 8.86. HRMS (ESI): Calcd. for $C_{23}H_{17}N_3O_2$ $[M+H]^+$: 368.1393; found: 368.1390.

(20) 2-(1-methyl-2-(3-phenyl-1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4aca)



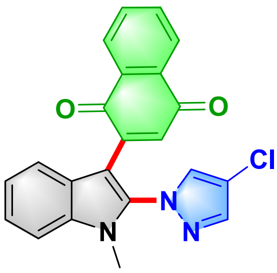
Red solid, (67.6 mg, 63% yield), m.p.: 198-203 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.11 (d, *J* = 7.6 Hz, 1H), 7.95 (d, *J* = 7.7 Hz, 1H), 7.85 (d, *J* = 7.1 Hz, 2H), 7.76–7.70 (m, 3H), 7.67–7.63 (m, 1H), 7.46 (d, *J* = 8.2 Hz, 1H), 7.42 (t, *J* = 8.0 Hz, 3H), 7.37 (d, *J* = 7.3 Hz, 1H), 7.32 (t, *J* = 7.0 Hz, 1H), 7.11 (s, 1H), 6.78 (d, *J* = 2.5 Hz, 1H), 3.79 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.91, 183.56, 154.14, 142.98, 135.42, 135.17, 134.93, 134.56, 133.59, 133.45, 132.68, 132.39, 132.19, 128.68, 128.44, 126.92, 126.02, 125.92, 125.16, 123.91, 121.90, 120.45, 110.24, 105.10, 30.21. HRMS (ESI): Calcd. for $C_{28}H_{19}N_3O_2$ $[M+H]^+$: 429.1550; found: 429.1481.

(21) ethyl1-(3-(1,4-dioxo-1,4-dihydronaphthalen-2-yl)-1-methyl-1*H*-indol-2-yl)-1*H*-pyrazole-4-carboxylate (4ada)



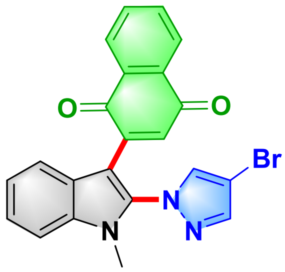
Red solid, (66.9 mg, 63% yield), m.p.: 179-182 °C; ^{1}H NMR (500 MHz,) δ 8.20 (s, 1H), 8.17 (s, 1H), 8.07 (d, *J* = 7.4 Hz, 1H), 7.96 (d, *J* = 8.6 Hz, 1H), 7.74–7.67 (m, 3H), 7.42–7.36 (m, 2H), 7.28 (t, *J* = 8.0 Hz, 1H), 7.06 (s, 1H), 4.31 (q, *J* = 7.1 Hz, 2H), 3.66 (s, 3H), 1.34 (t, *J* = 7.1 Hz, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.72, 183.30, 162.35, 143.05, 142.01, 136.45, 135.53, 135.27, 133.77, 133.69, 133.59, 132.37, 132.11, 126.92, 125.98, 124.88, 124.32, 122.11, 120.55, 117.08, 110.36, 104.43, 60.65, 30.10, 14.35. HRMS (ESI): Calcd. for $C_{25}H_{19}N_3O_4$ $[M+H]^+$: 426.1448; found: 426.1445.

(22) 2-(2-(4-chloro-1*H*-pyrazol-1-yl)-1-methyl-1*H*-indol-3-yl)naphthalene-1,4-dione (4afa)



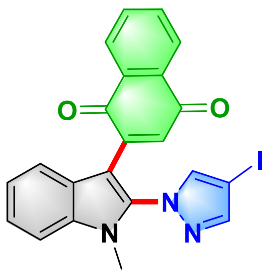
Red solid, (54.2 mg, 56% yield), m.p.: 157-159 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.11 (d, *J* = 5.2 Hz, 1H), 8.02 (d, *J* = 6.0 Hz, 1H), 7.78–7.69 (m, 5H), 7.45–7.40 (m, 2H), 7.31 (t, *J* = 6.4 Hz, 1H), 7.08 (s, 1H), 3.68 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.78, 183.40, 142.23, 140.89, 135.42, 135.24, 134.08, 133.78, 133.62, 132.48, 132.17, 130.86, 126.93, 126.03, 124.92, 124.24, 122.07, 120.52, 112.48, 110.31, 104.29, 30.03. HRMS (ESI): Calcd. for $C_{22}H_{14}ClN_3O_2$ $[M+H]^+$: 388.0847; found: 388.0843.

(23) 2-(2-(4-bromo-1*H*-pyrazol-1-yl)-1-methyl-1*H*-indol-3-yl)naphthalene-1,4-dione (4aga)



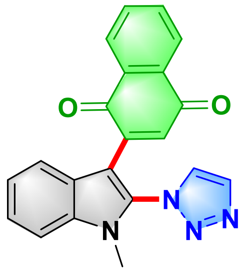
Red solid, (60.3 mg, 56% yield), m.p.: 137-139 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.12 (d, *J* = 6.0 Hz, 1H), 8.02 (d, *J* = 7.7 Hz, 1H), 7.79–7.72 (m, 4H), 7.71 (d, *J* = 8.1 Hz, 1H), 7.46–7.40 (m, 2H), 7.32 (t, *J* = 6.5 Hz, 1H), 7.08 (s, 1H), 3.69 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.79, 183.41, 142.94, 142.23, 135.44, 135.26, 133.98, 133.78, 133.63, 133.01, 132.49, 132.18, 126.95, 126.04, 124.92, 124.25, 122.08, 120.52, 110.30, 104.33, 95.71, 30.04. HRMS (ESI): Calcd. for $C_{22}H_{14}BrN_3O_2$ $[M+H]^+$: 432.0342; found: 432.0337.

(24) 2-(2-(4-iodo-1*H*-pyrazol-1-yl)-1-methyl-1*H*-indol-3-yl)naphthalene-1,4-dione (4aha)



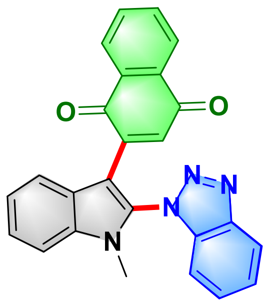
Red solid, (70.7 mg, 59% yield), m.p.: 141-143 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.11 (d, *J* = 7.3 Hz, 1H), 8.00 (d, *J* = 7.7 Hz, 1H), 7.80 (s, 1H), 7.79 –7.71 (m, 3H), 7.70 (d, *J* = 8.1 Hz, 1H), 7.46–7.37 (m, 2H), 7.33–7.28 (m, 1H), 7.07 (s, 1H), 3.67 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.80, 183.43, 147.28, 142.28, 137.27, 135.40, 135.28, 133.84, 133.77, 133.63, 132.52, 132.19, 126.96, 126.05, 124.92, 124.24, 122.07, 120.52, 110.29, 104.33, 58.95, 30.06. HRMS (ESI): Calcd. for $C_{22}H_{14}IN_3O_2$ $[M+H]^+$: 480.0203; found: 480.0199.

(25) 2-(1-methyl-2-(1*H*-1,2,3-triazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4aia)



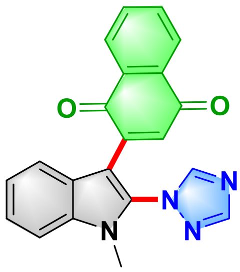
Red solid, (58.4 mg, 66% yield), m.p.: 212-214 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.07 (d, *J* = 7.5 Hz, 1H), 8.02 (s, 1H), 7.91 (d, *J* = 7.4 Hz, 1H), 7.86 (s, 1H), 7.73 (d, *J* = 7.6 Hz, 2H), 7.69 (t, *J* = 7.4 Hz, 1H), 7.44 (q, *J* = 8.3 Hz, 2H), 7.32 (t, *J* = 7.2 Hz, 1H), 7.14 (s, 1H), 3.63 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.60, 183.13, 141.53, 136.06, 135.52, 133.89, 133.78, 133.66, 132.22, 132.05, 130.66, 127.62, 126.89, 126.03, 124.88, 124.64, 122.30, 120.44, 110.48, 105.09, 30.04. HRMS (ESI): Calcd. for $C_{21}H_{14}N_4O_2$ $[M+H]^+$: 355.1189; found: 355.1184.

(26) 2-(2-(1*H*-benzo[*d*][1,2,3]triazol-1-yl)-1-methyl-1*H*-indol-3-yl)naphthalene-1,4-dione (4aja)



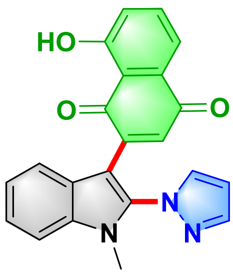
Red solid, (72.7 mg, 72% yield), m.p.: 176-178 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.15 (d, *J* = 8.3 Hz, 1H), 8.00 (d, *J* = 7.7 Hz, 1H), 7.79 (d, *J* = 8.0 Hz, 1H), 7.70–7.64 (m, 2H), 7.58 (t, *J* = 7.5 Hz, 1H), 7.51 (t, *J* = 7.5 Hz, 2H), 7.44 (td, *J* = 13.3, 12.7, 7.5 Hz, 3H), 7.36 (t, *J* = 7.5 Hz, 1H), 7.13 (s, 1H), 3.66 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.69, 182.92, 145.33, 142.00, 135.92, 135.25, 134.90, 133.71, 133.53, 132.12, 131.87, 130.09, 129.33, 126.72, 125.88, 125.26, 124.80, 124.56, 122.28, 120.58, 120.50, 110.57, 109.98, 105.55, 30.38. HRMS (ESI): Calcd. for $C_{25}H_{16}N_4O_2$ $[M+H]^+$: 405.1346; found: 405.1342.

(27) 2-(1-methyl-2-(1*H*-1,2,4-triazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4aka)



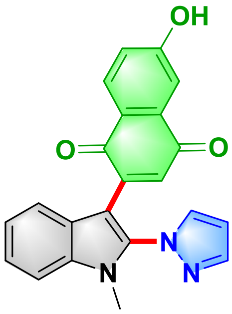
Red solid, (61.1 mg, 69% yield), m.p.: 207-209 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 8.43 (s, 1H), 8.20 (s, 1H), 8.08 (d, *J* = 8.7 Hz, 1H), 7.96 (d, *J* = 6.6 Hz, 1H), 7.76–7.72 (m, 1H), 7.70 (t, *J* = 7.6 Hz, 2H), 7.46–7.40 (m, 2H), 7.31 (t, *J* = 7.3 Hz, 1H), 7.12 (s, 1H), 3.67 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.59, 183.25, 141.49, 136.14, 135.50, 133.94, 133.72, 132.18, 132.03, 130.64, 127.03, 126.04, 124.81, 124.60, 122.25, 120.43, 110.40, 105.00, 30.04. HRMS (ESI): Calcd. for $C_{21}H_{14}N_4O_2$ $[M+H]^+$: 355.1189; found: 355.1184.

(28) 8-hydroxy-2-(1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4aab)



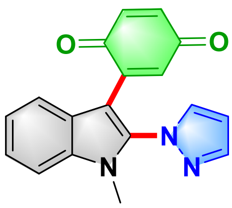
Red solid, (65.5 mg, 71% yield), m.p.: 253-255 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 11.79 (s, 1H), 7.83 (d, *J* = 1.5 Hz, 1H), 7.71 (d, *J* = 8.0 Hz, 1H), 7.68 (d, *J* = 2.4 Hz, 1H), 7.64 (q, *J* = 4.0, 3.6 Hz, 2H), 7.45 (d, *J* = 8.1 Hz, 1H), 7.41 (t, *J* = 7.0 Hz, 1H), 7.31 (t, *J* = 6.9 Hz, 1H), 7.25 (dd, *J* = 6.6, 3.0 Hz, 1H), 6.99 (s, 1H), 6.49 –6.46 (m, 1H), 3.72 (s, 3H). ^{13}C NMR (125 MHz, Chloroform-d) δ 188.62, 184.09, 161.56, 142.48, 142.43, 136.44, 136.03, 135.30, 135.15, 133.13, 132.28, 125.02, 124.05, 124.01, 121.99, 120.37, 118.62, 115.32, 110.31, 107.79, 103.00, 30.12. HRMS (ESI): Calcd. for $C_{22}H_{15}N_3O_3$ $[M+H]^+$: 370.1186; found: 370.1183.

(29) 6-hydroxy-2-(1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4aac)



Red solid, (58.1 mg, 63% yield), m.p.: 272-274 °C; ^{1}H NMR (500 MHz, DMSO-d6) δ 8.19 (d, *J* = 2.3 Hz, 1H), 7.86 (d, *J* = 8.1 Hz, 1H), 7.82 (d, *J* = 2.3 Hz, 1H), 7.66–7.63 (m, 2H), 7.37 (t, *J* = 7.7 Hz, 1H), 7.24 (t, *J* = 7.6 Hz, 1H), 7.18 (s, 1H), 7.16 (d, *J* = 2.2 Hz, 1H), 6.70 (s, 1H), 6.56–6.55 (m, 1H), 3.67 (s, 3H). ^{13}C NMR (125 MHz, DMSO-d6) δ 183.59, 183.51, 163.03, 142.49, 142.29, 135.40, 135.08, 134.99, 134.72, 134.66, 128.87, 125.00, 124.28, 123.83, 121.80, 121.21, 120.91, 112.67, 111.27, 108.08, 103.65, 30.38. HRMS (ESI): Calcd. for $C_{22}H_{15}N_3O_3$ $[M+H]^+$: 370.1186; found: 370.1183.

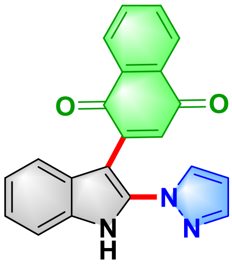
(30) 2-(1-methyl-2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)cyclohexa-2,5-diene-1,4-dione(4aad)



Black solid, (43.2 mg, 57% yield), m.p.: 153-156 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 10.19 (s, 1H), 8.18 (d, *J* = 7.5 Hz, 1H), 8.06 (d, *J* = 8.6 Hz, 1H), 7.81 –7.74 (m, 2H), 7.72 (d, *J* = 1.5 Hz, 1H), 7.65–7.62 (m, 2H), 7.42 (d, *J* = 7.6 Hz, 1H), 7.28 (s, 1H), 7.28–7.22 (m, 2H), 6.37 (t, *J* = 10 Hz, 1H). ^{13}C NMR (125 MHz, Chloroform-d) δ

187.36, 185.35, 142.31, 140.61, 136.75, 136.48, 135.28, 134.98, 133.17, 132.59, 124.86, 124.01, 121.97, 120.24, 110.28, 107.72, 103.42, 30.00. HRMS (ESI): Calcd. for $C_{18}H_{13}N_3O_2$ $[M+H]^+$: 304.1080; found: 304.1076.

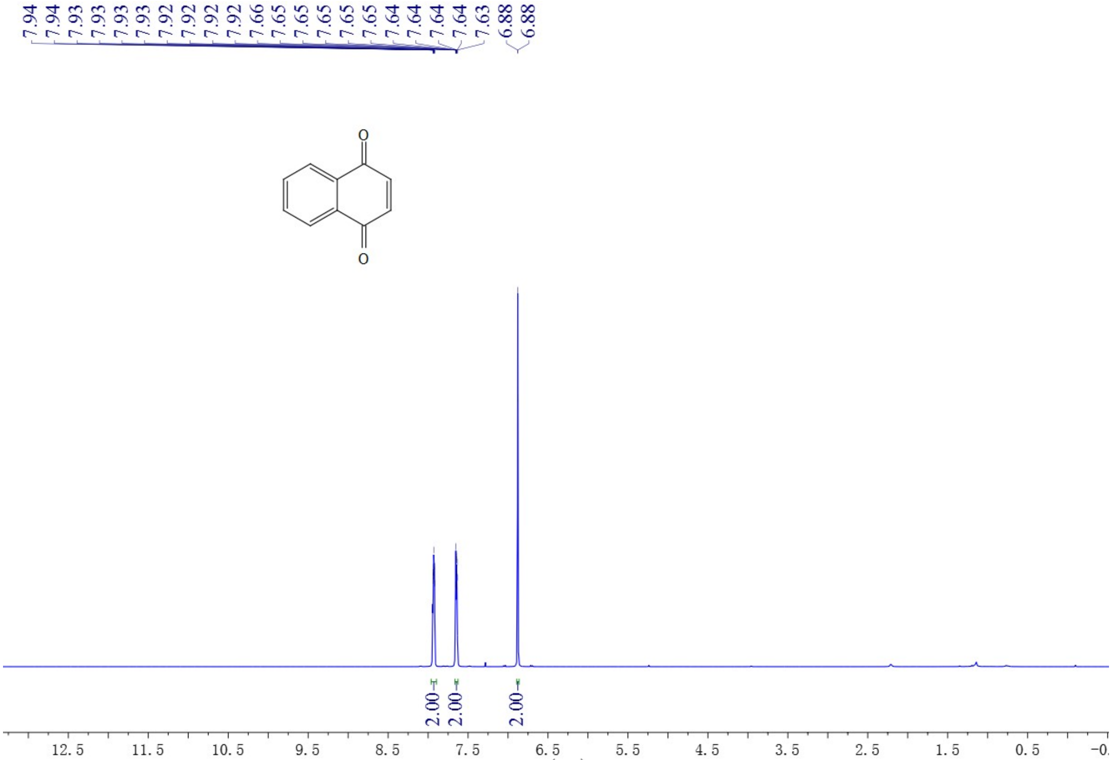
(31) 2-(2-(1*H*-pyrazol-1-yl)-1*H*-indol-3-yl)naphthalene-1,4-dione (4paa)



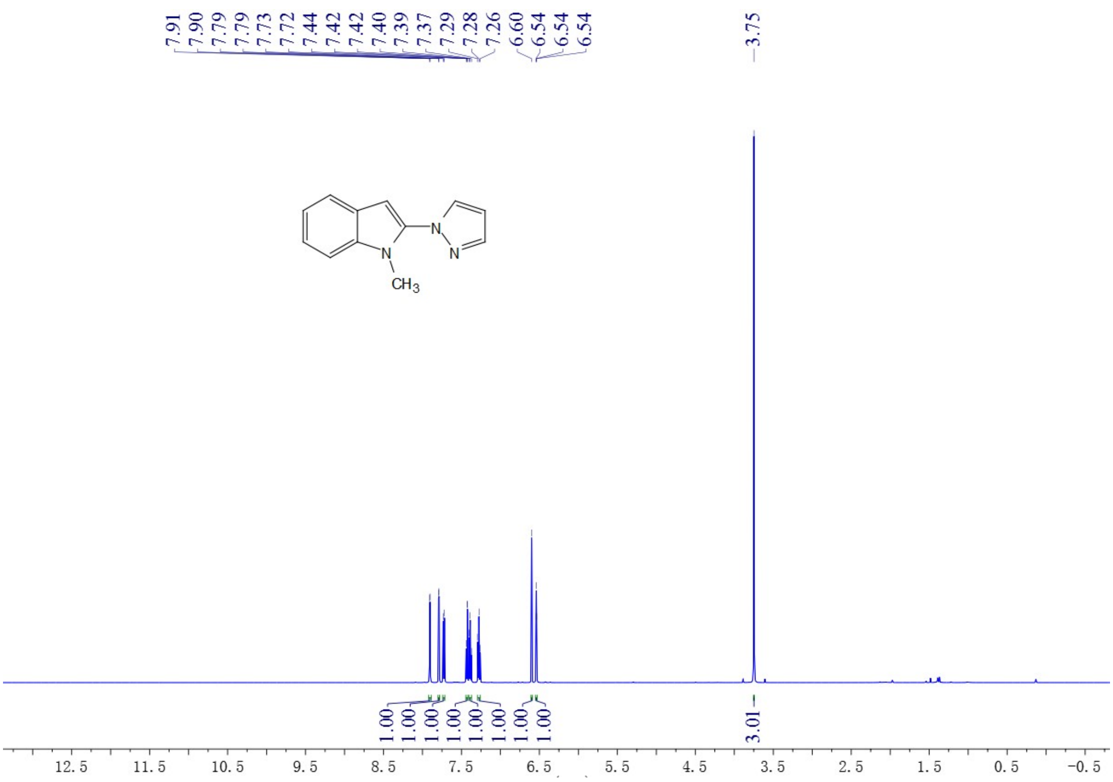
Red solid, (48.3 mg, 57% yield), m.p.: 149-151 °C; ^{1}H NMR (500 MHz, Chloroform-d) δ 7.76 (d, J = 8.8 Hz, 1H), 7.60 (d, J = 7.9 Hz, 1H), 7.39 (d, J = 8.3 Hz, 1H), 7.32–7.27 (m, 4H), 7.24 (d, J = 7.1 Hz, 1H), 7.12–7.09 (m, 3H), 7.05–7.01 (m, 2H), 6.91–6.84 (m, 4H), 5.31 (s, 4H). ^{13}C NMR (125 MHz, Chloroform-d) δ 184.95, 183.51, 143.18, 141.67, 136.07, 134.50, 133.91, 133.71, 133.47, 132.61, 132.28, 130.37, 127.13, 126.68, 126.14, 123.54, 121.76, 119.26, 111.74, 108.19, 97.70. HRMS (ESI): Calcd. for $C_{21}H_{13}N_3O_2$ $[M+H]^+$: 340.1080; found: 340.1077.

10. NMR spectra of the obtained compounds.

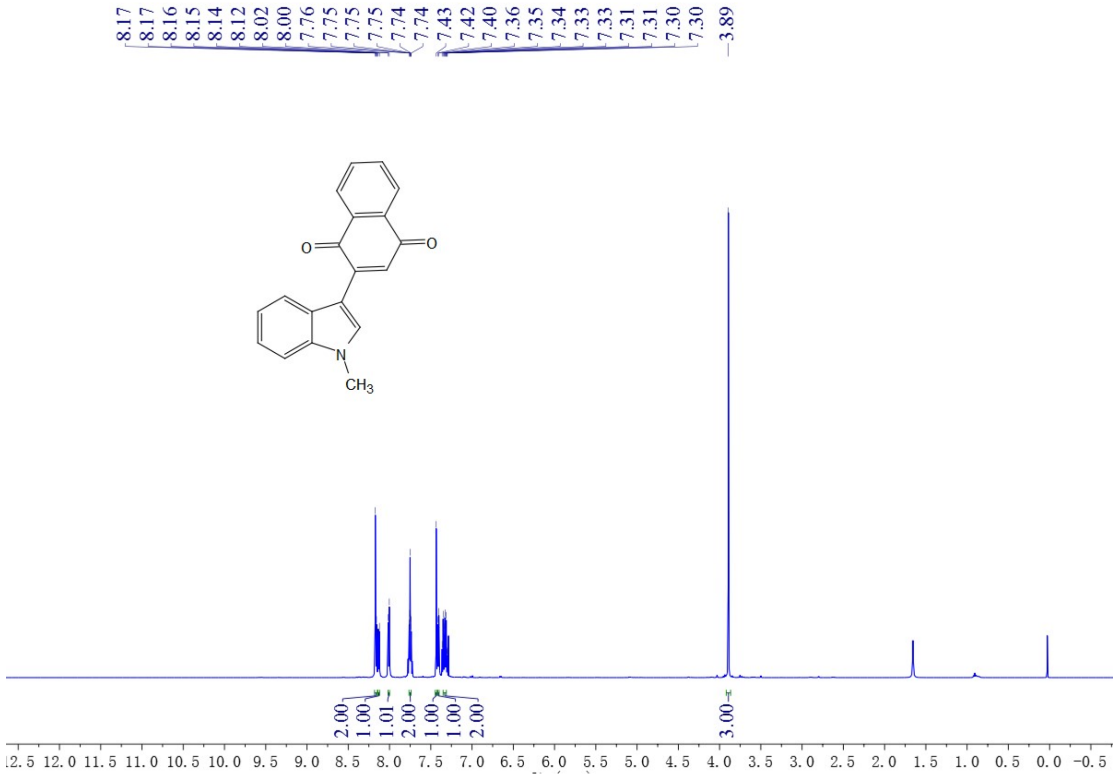
(1) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 3a-1



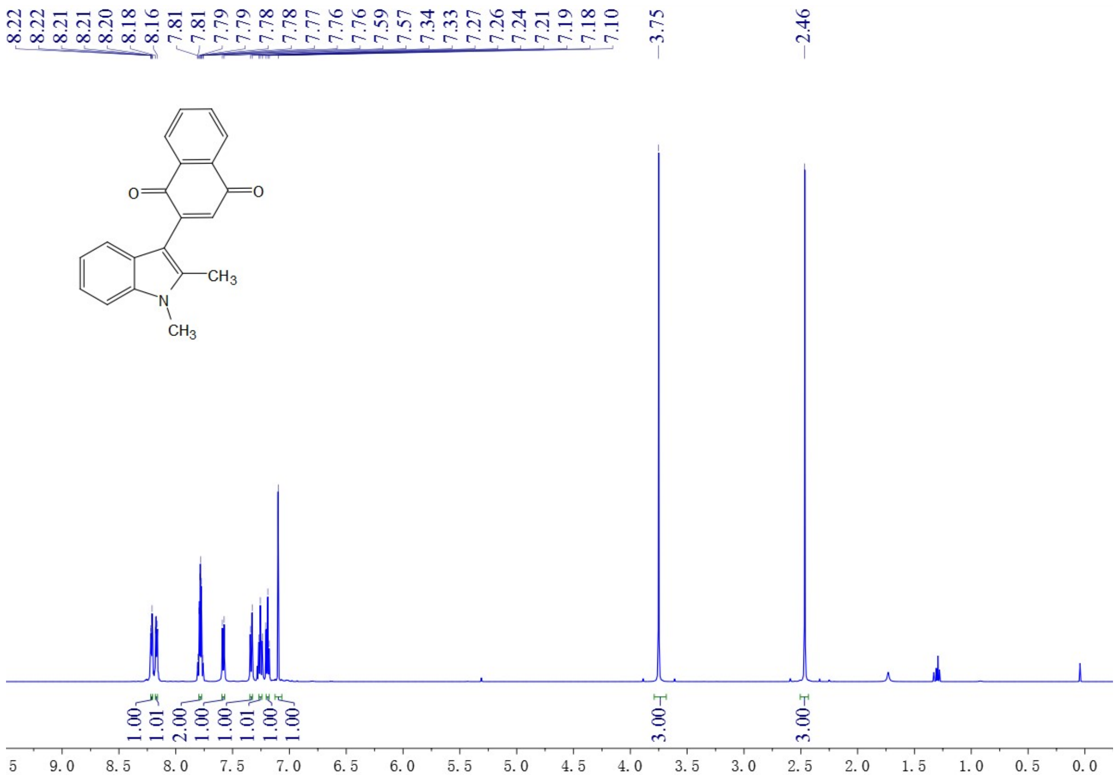
(2) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 1a-1



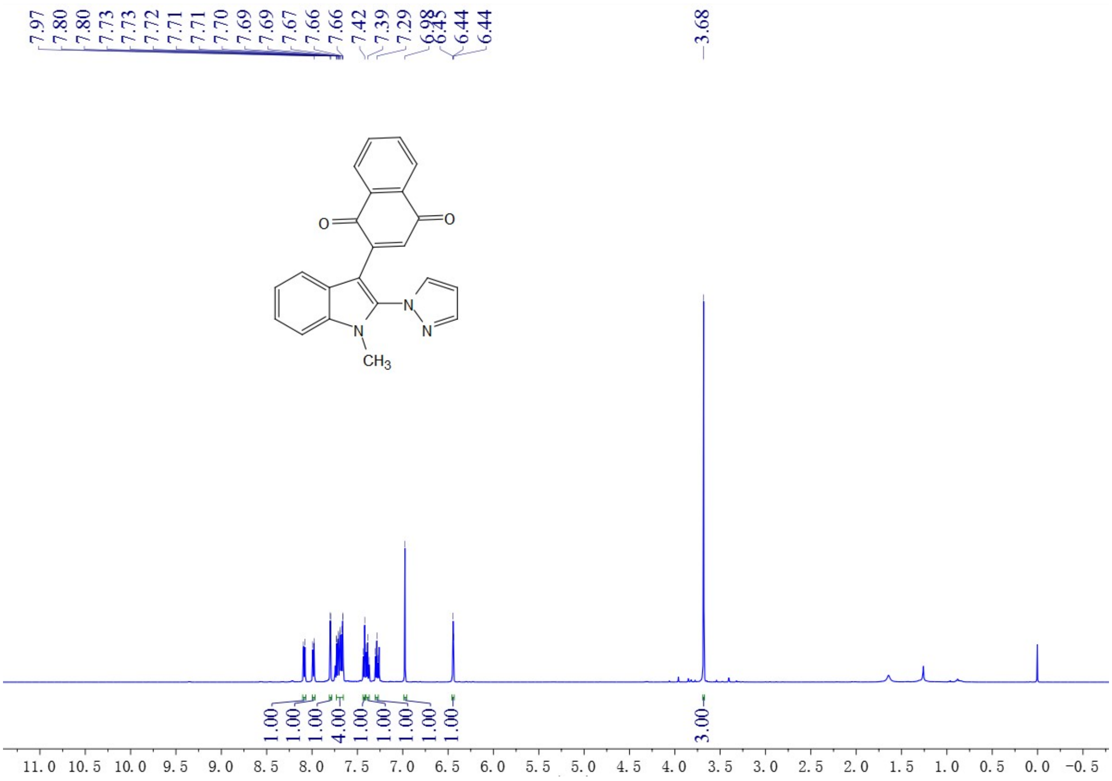
(3) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 1a-2



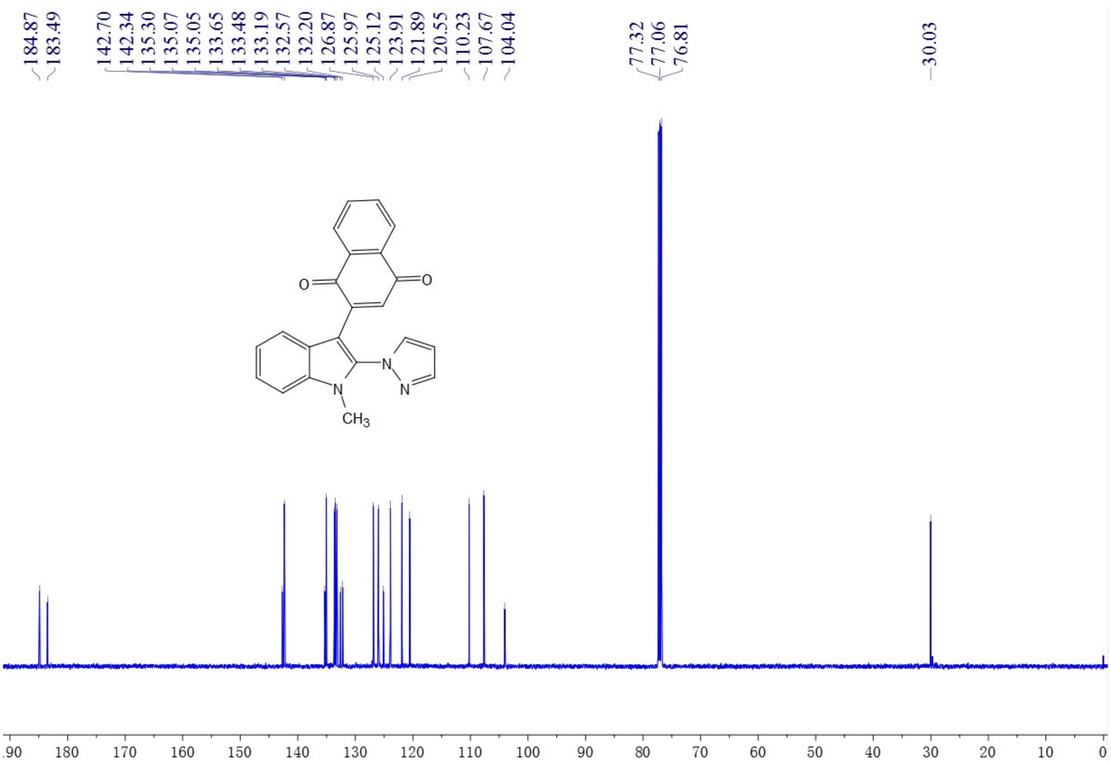
(4) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 1a-3'



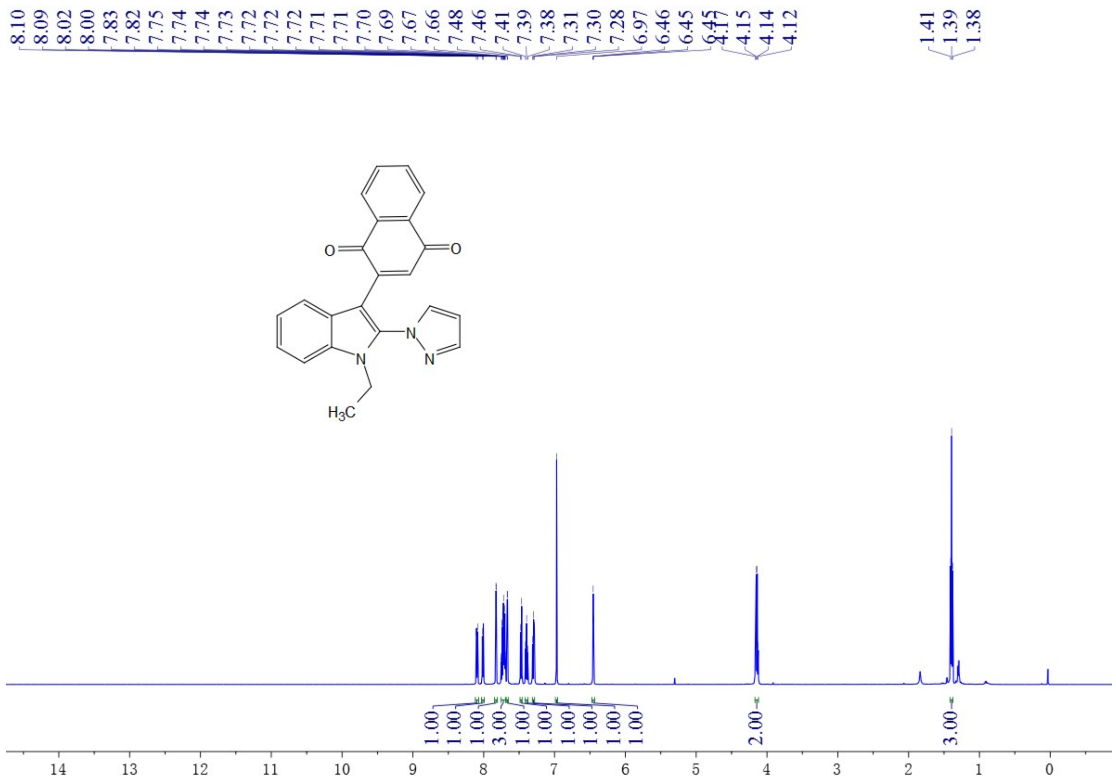
(5) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4aaa



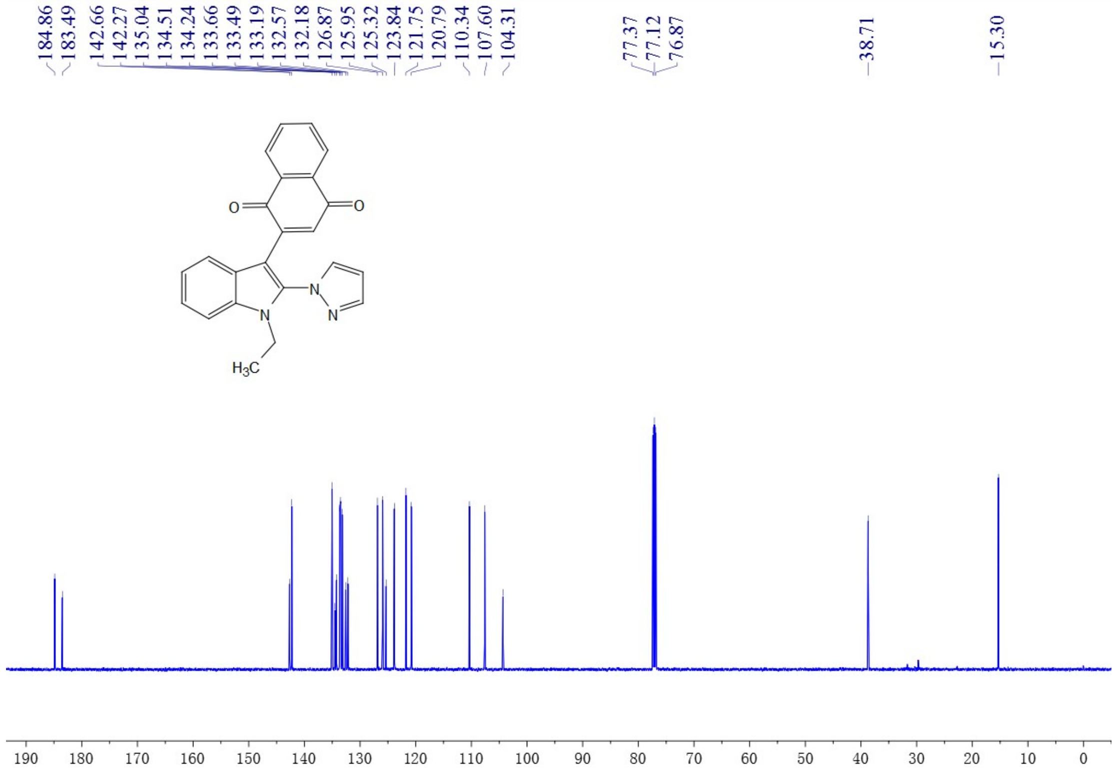
(6) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4baa



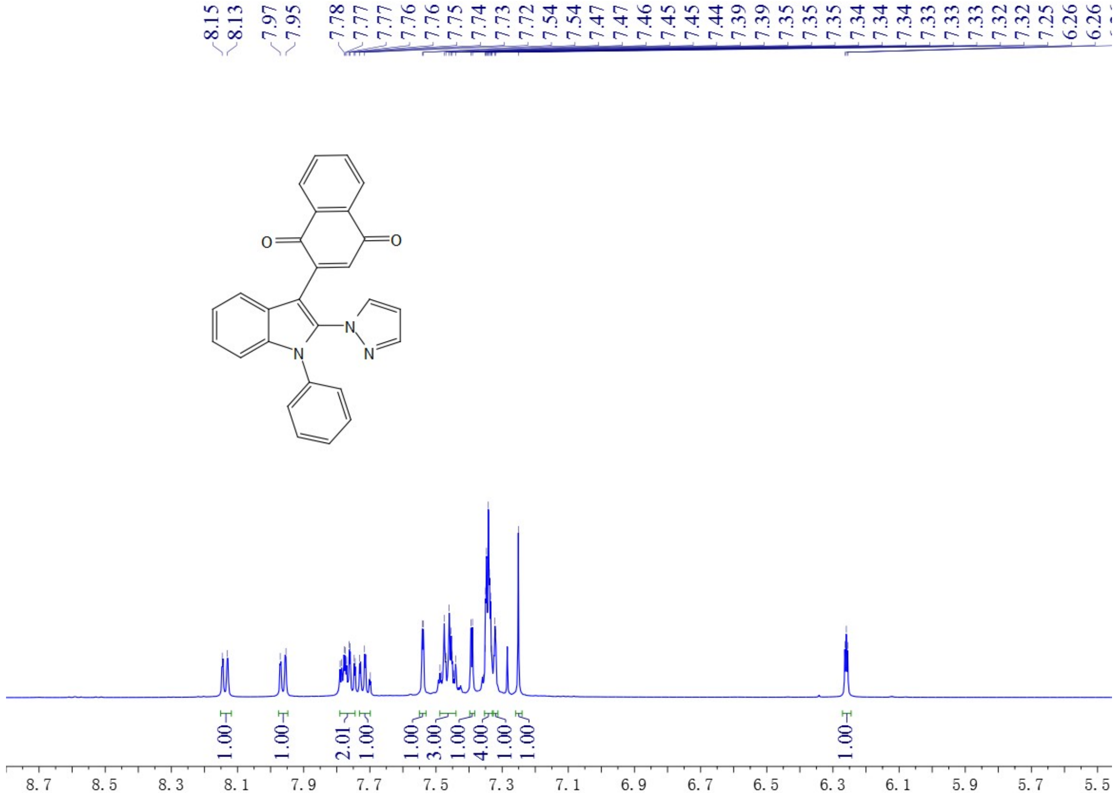
(7) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4baa



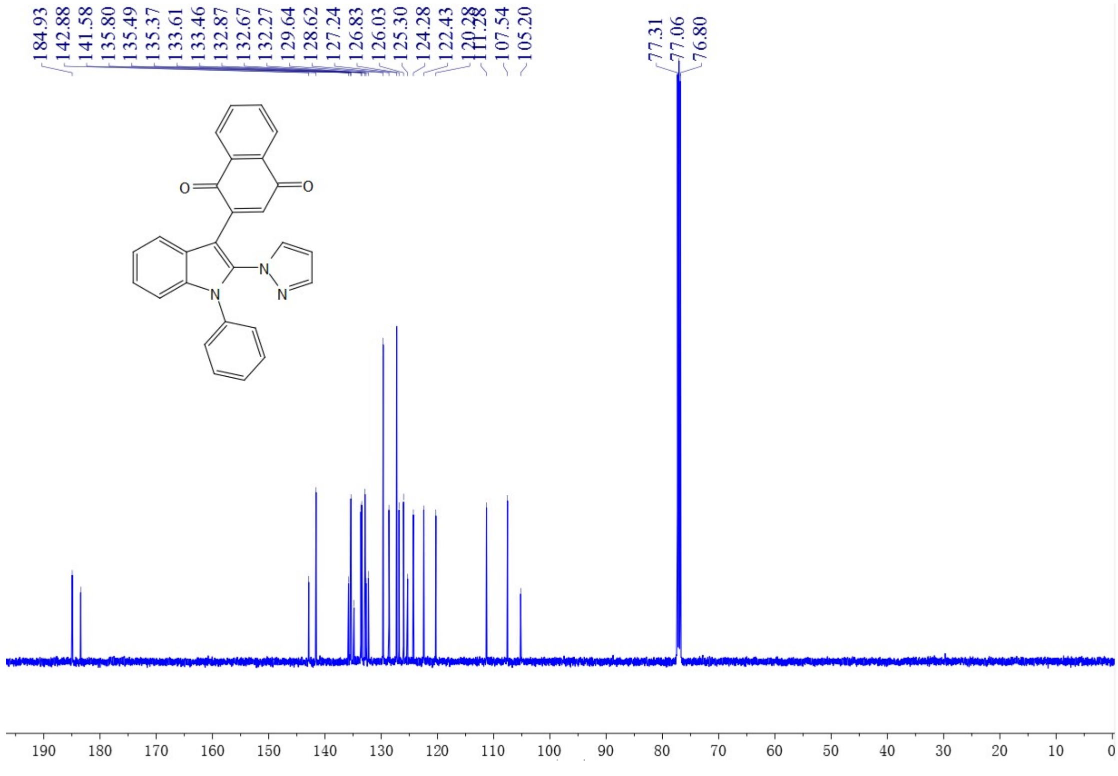
(8) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4baa



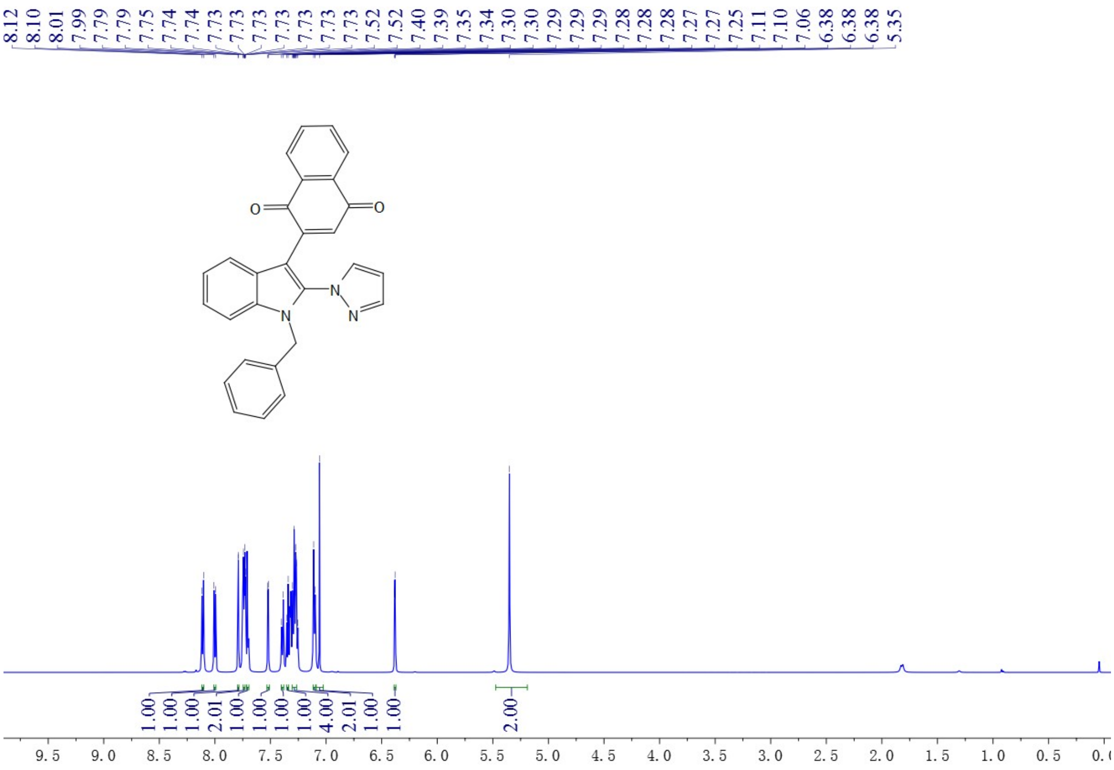
(9) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4caa



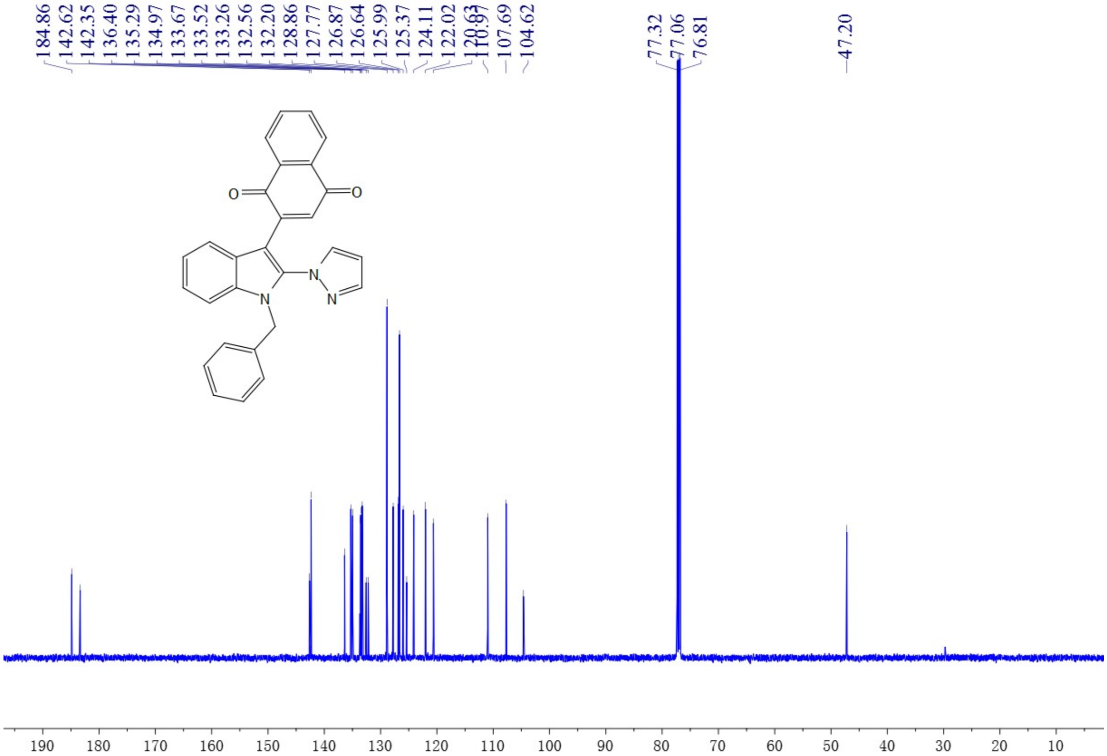
(10) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4caa



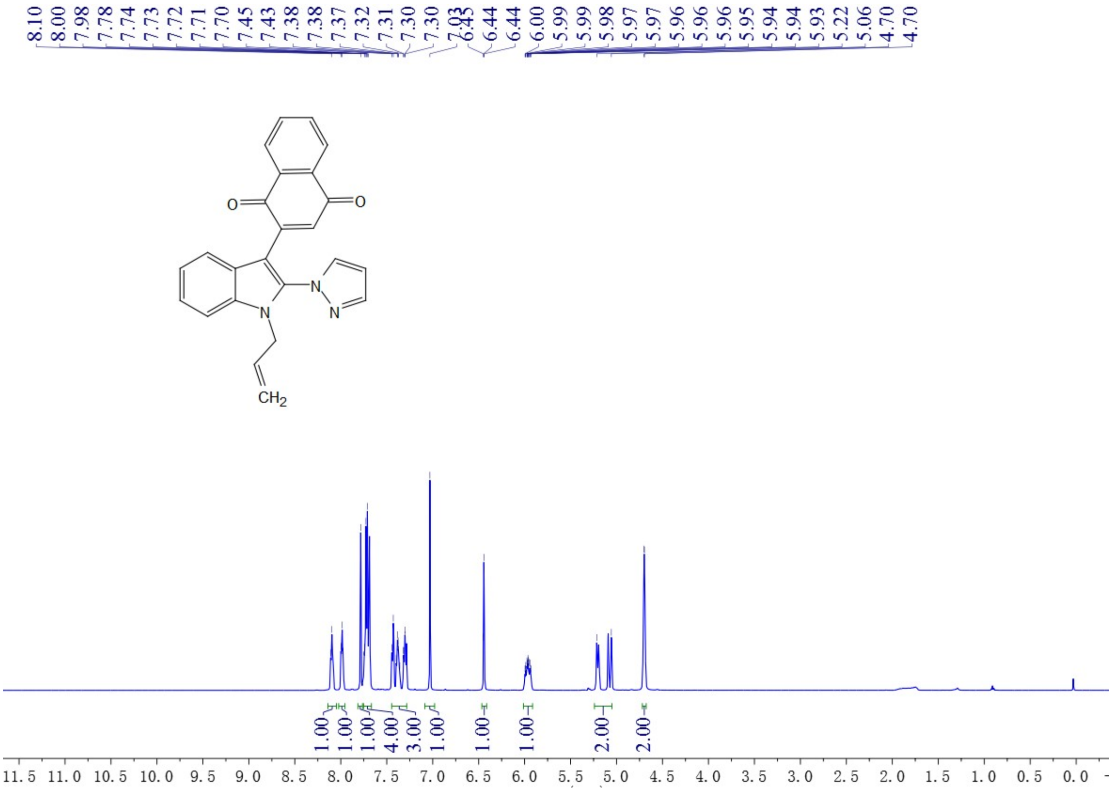
(11) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4daa



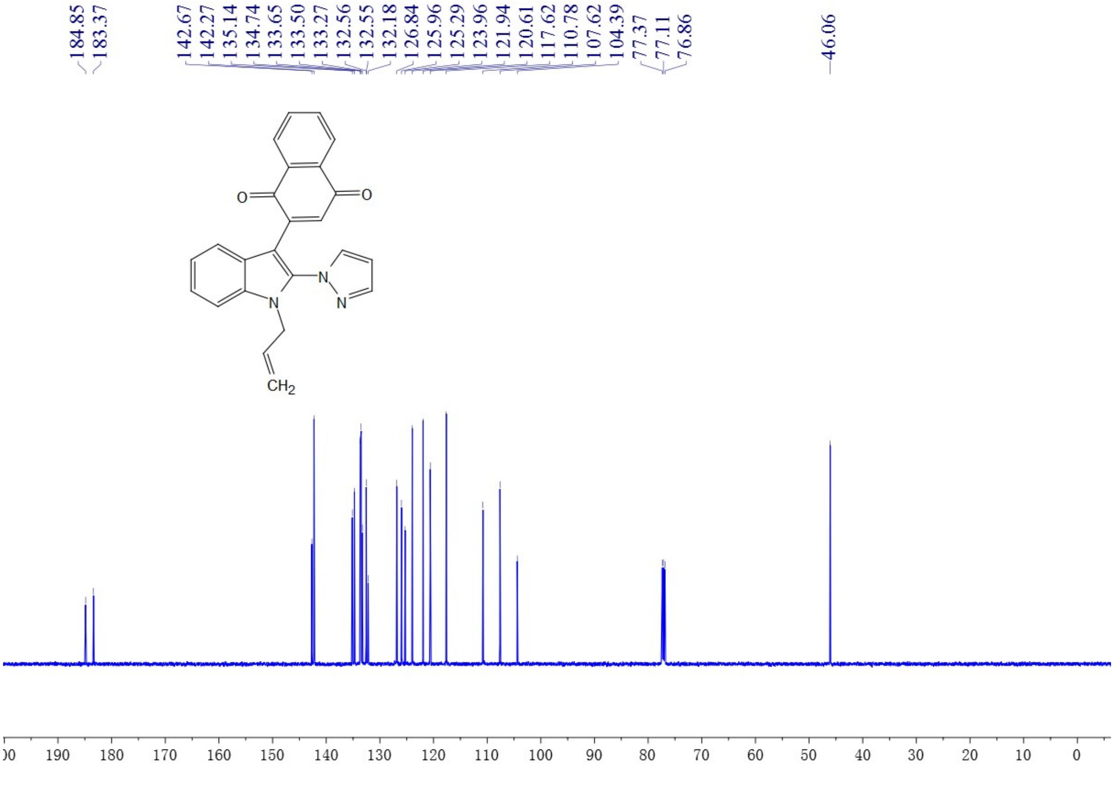
(12) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4daa



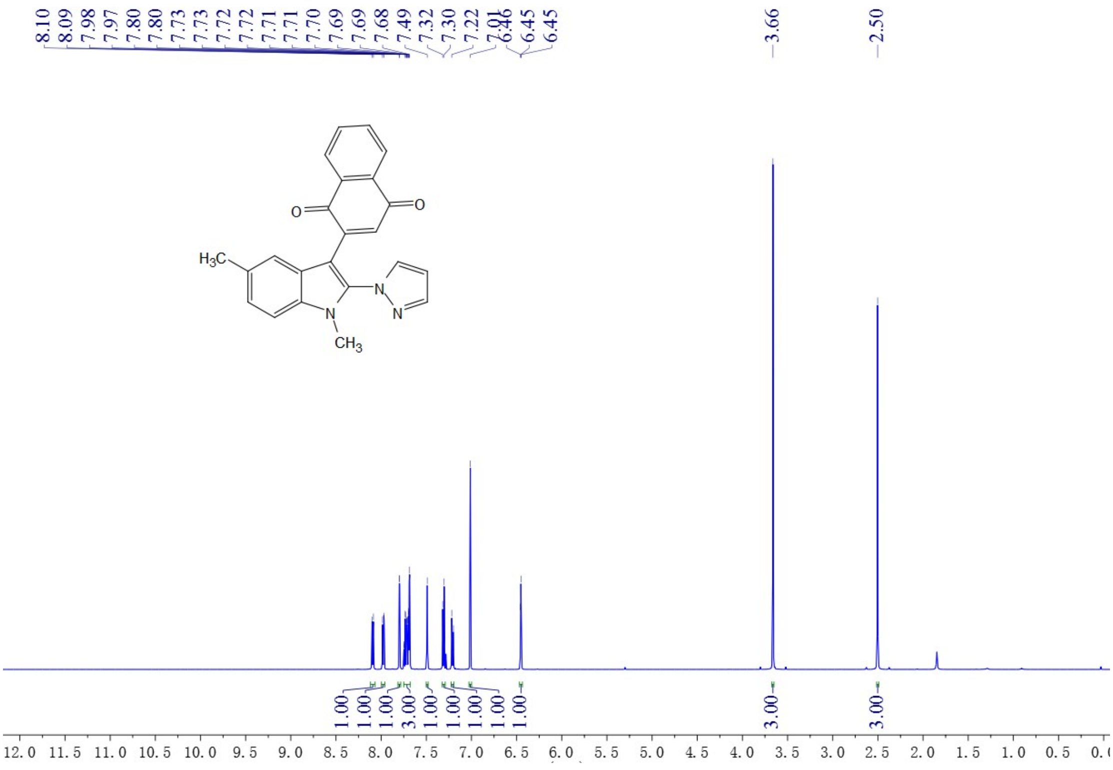
(13) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4eaa



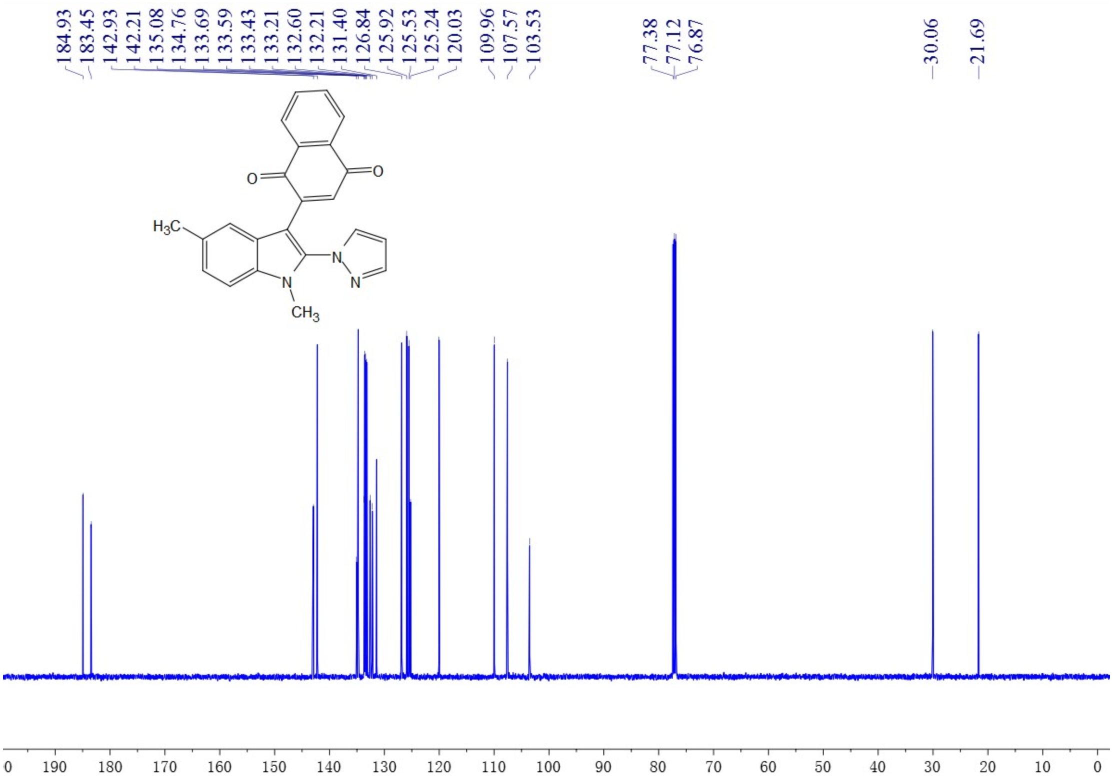
(14) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4eaa



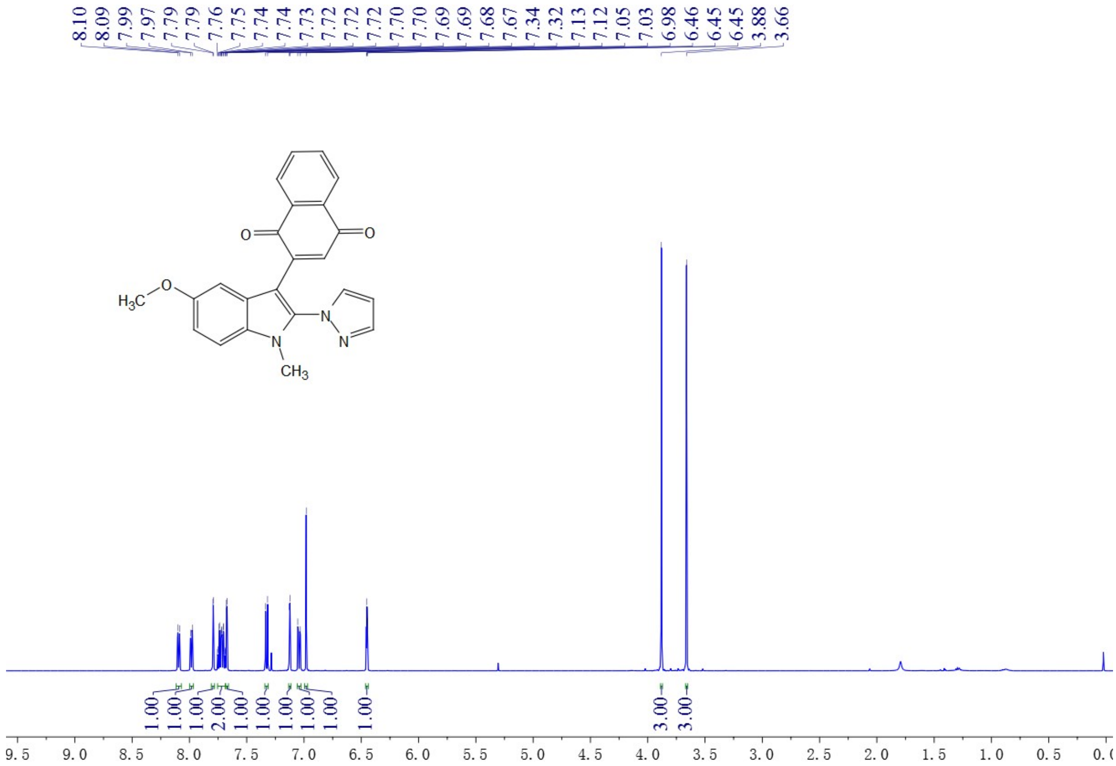
(15) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4faa



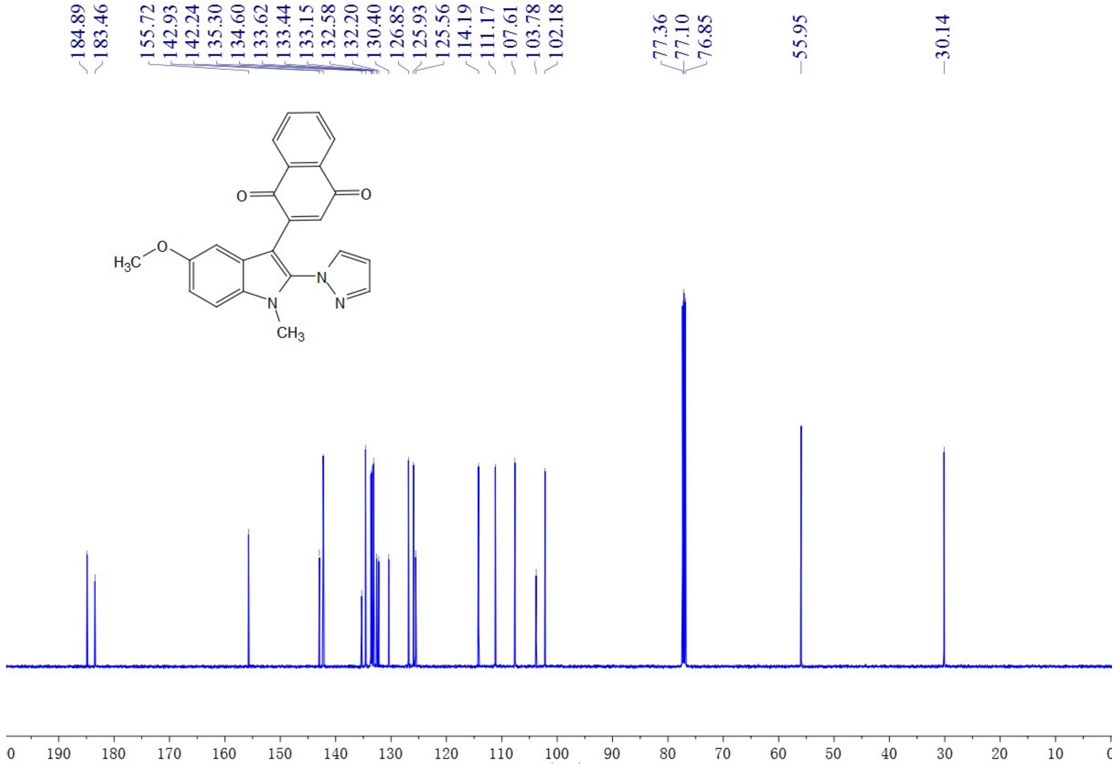
(16) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4faa



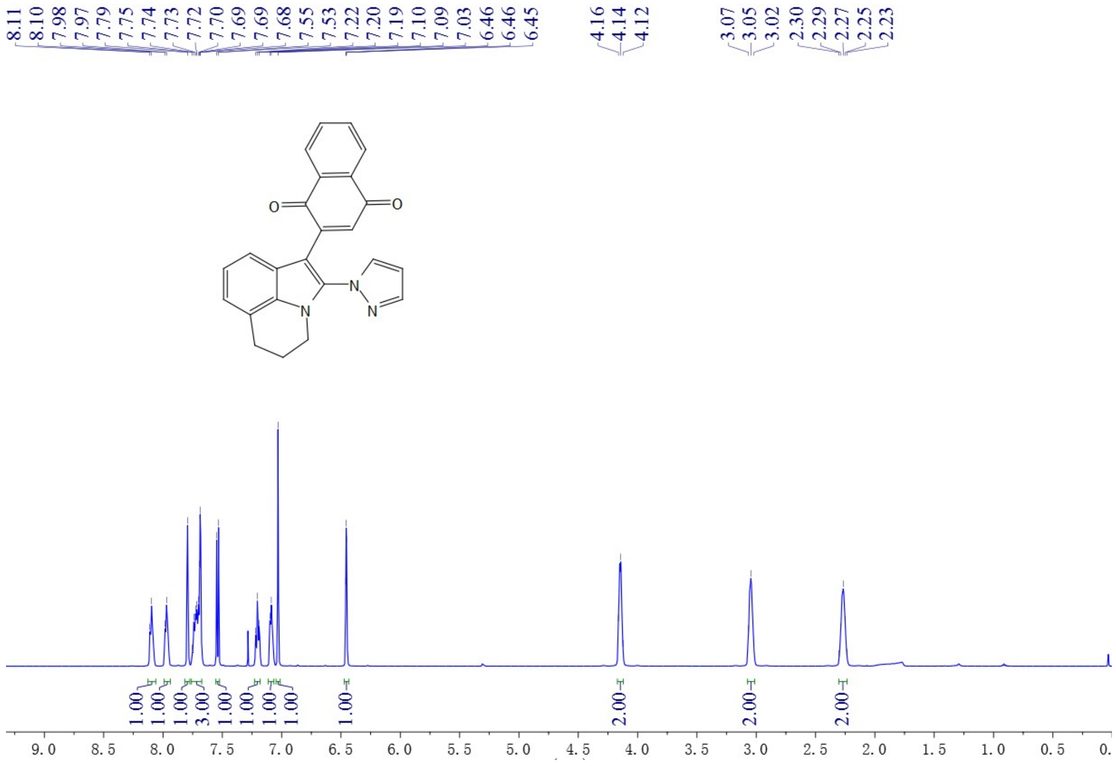
(17) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4gaa



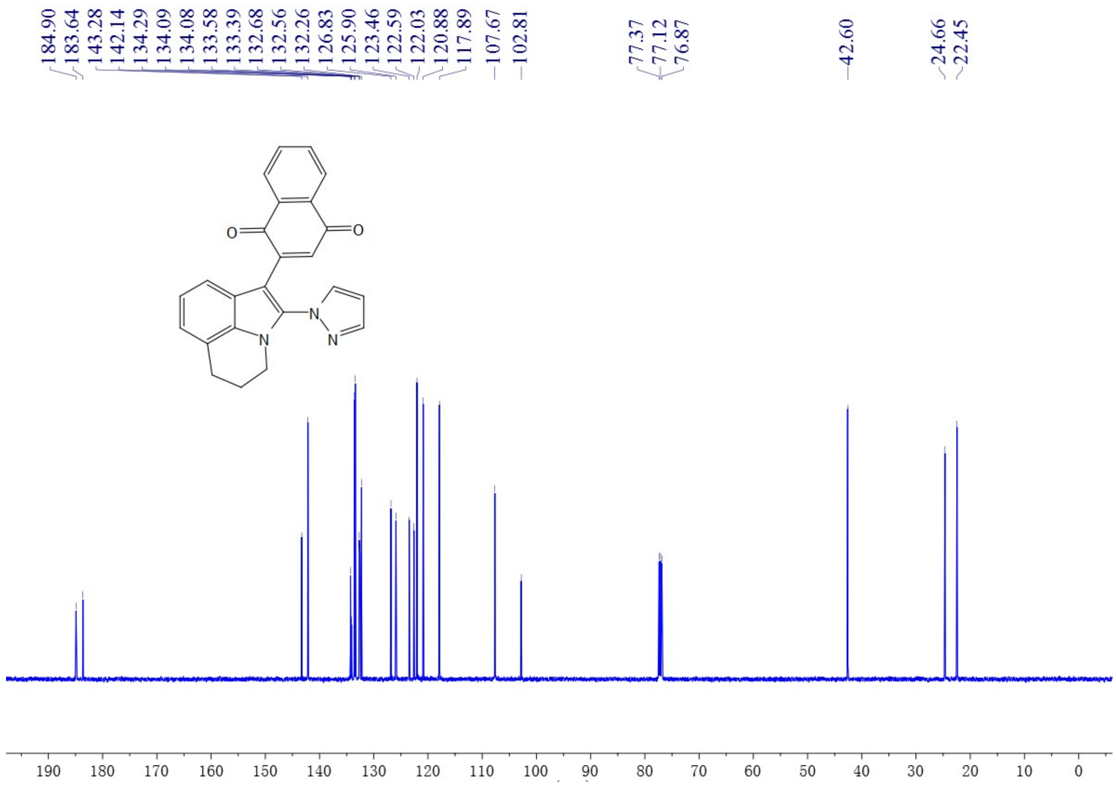
(18) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4gaa



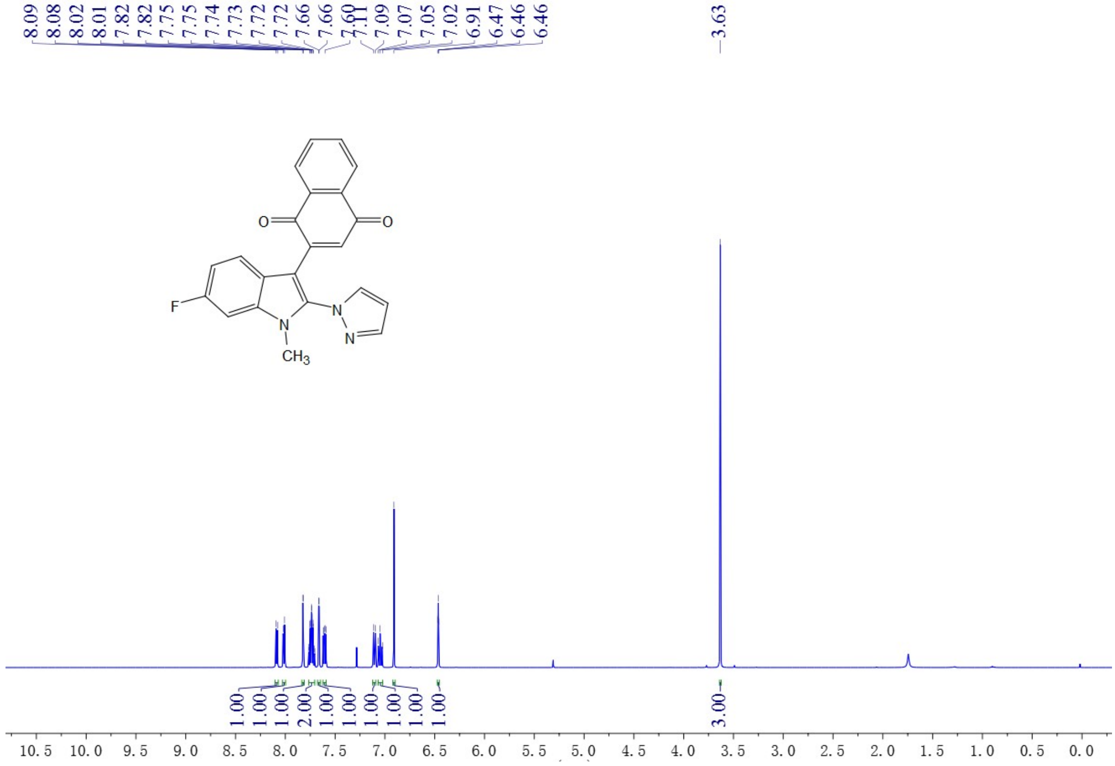
(19) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4haa



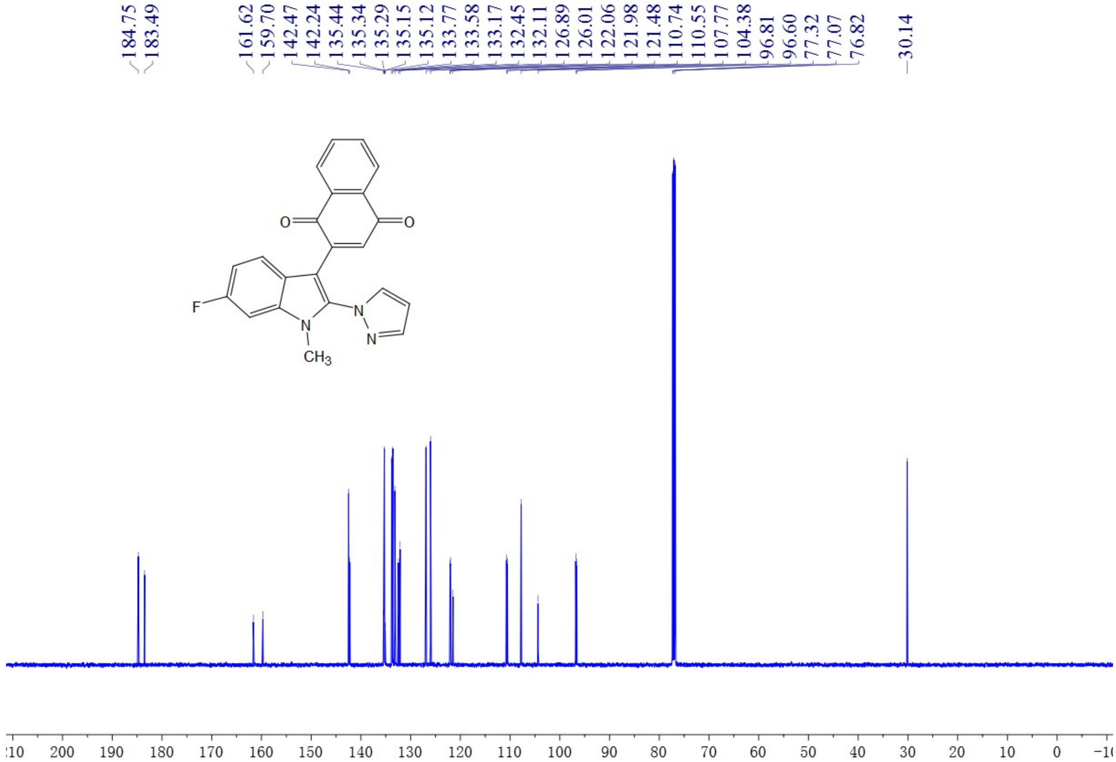
(20) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4haa



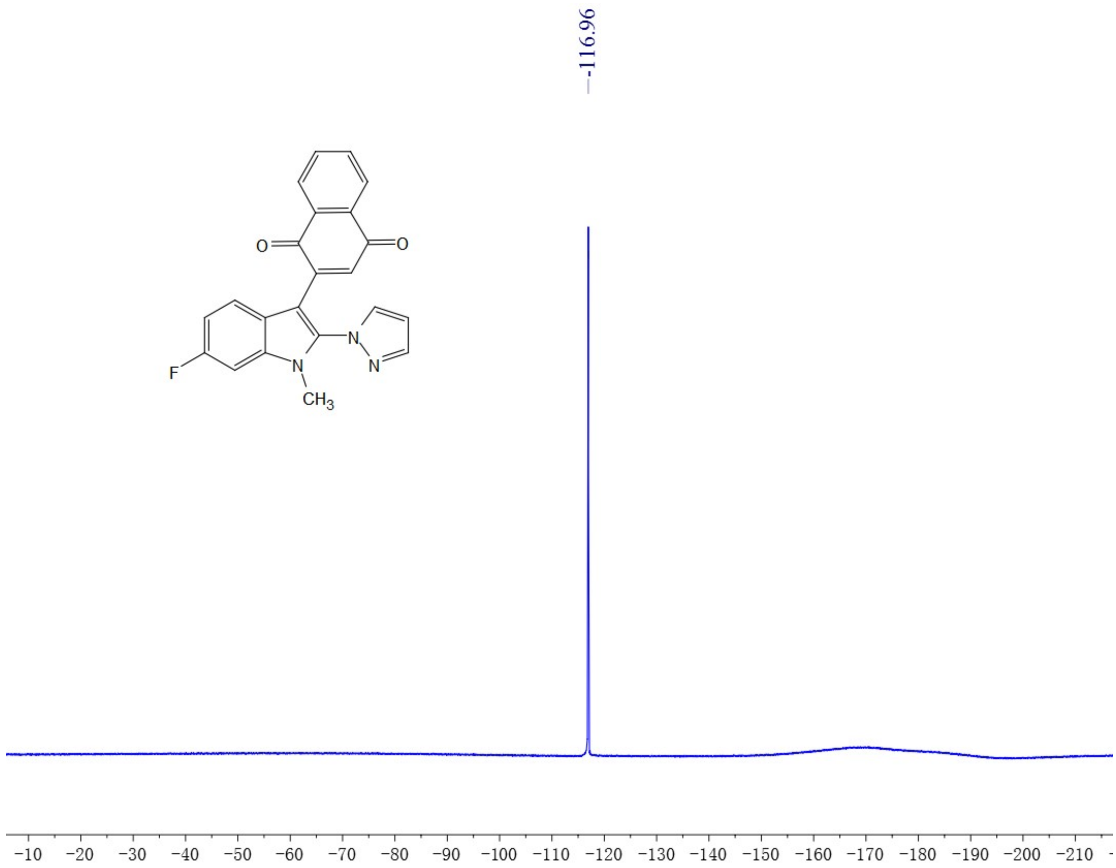
(21) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4iaa



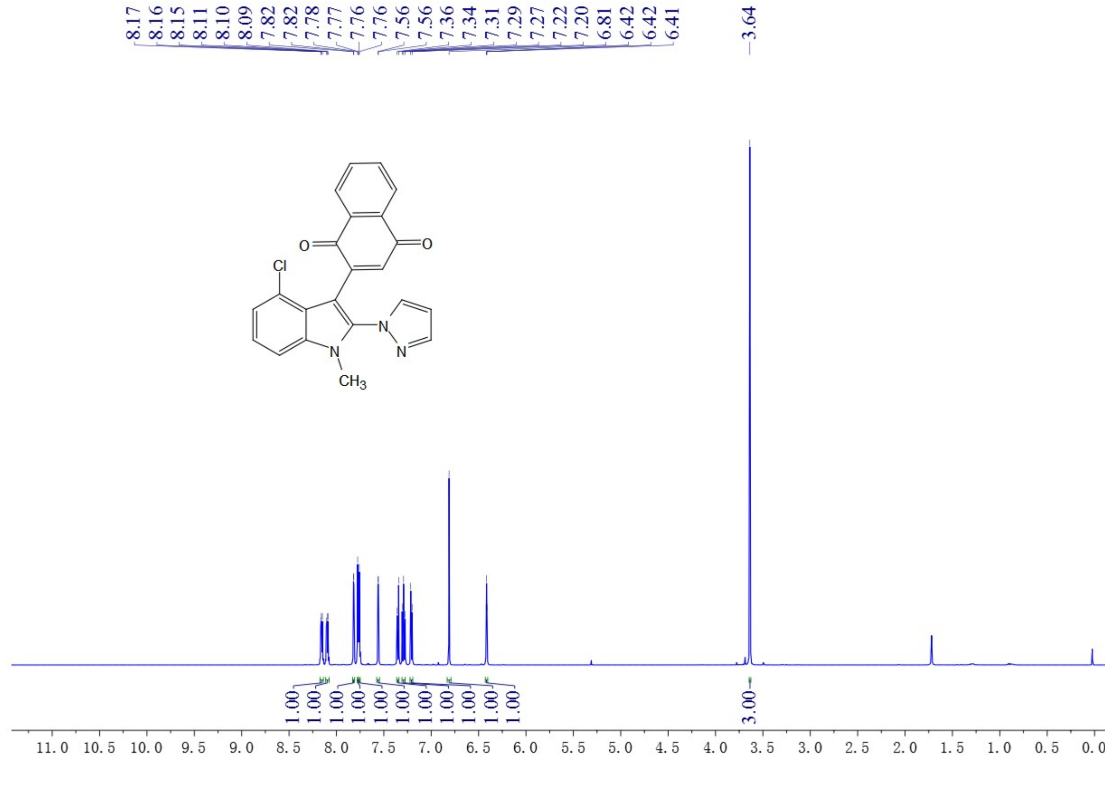
(22) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4iaa



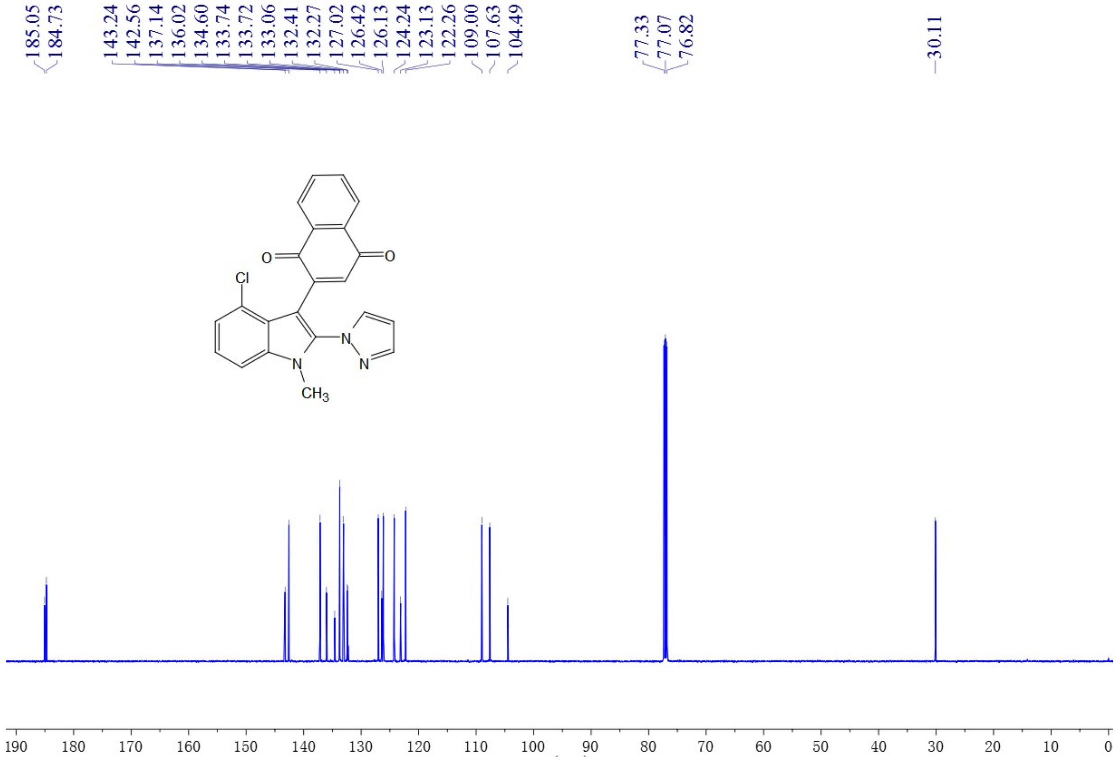
(23) ^{19}F-NMR (470 MHz, $CDCl_3$) spectrum of 4iaa



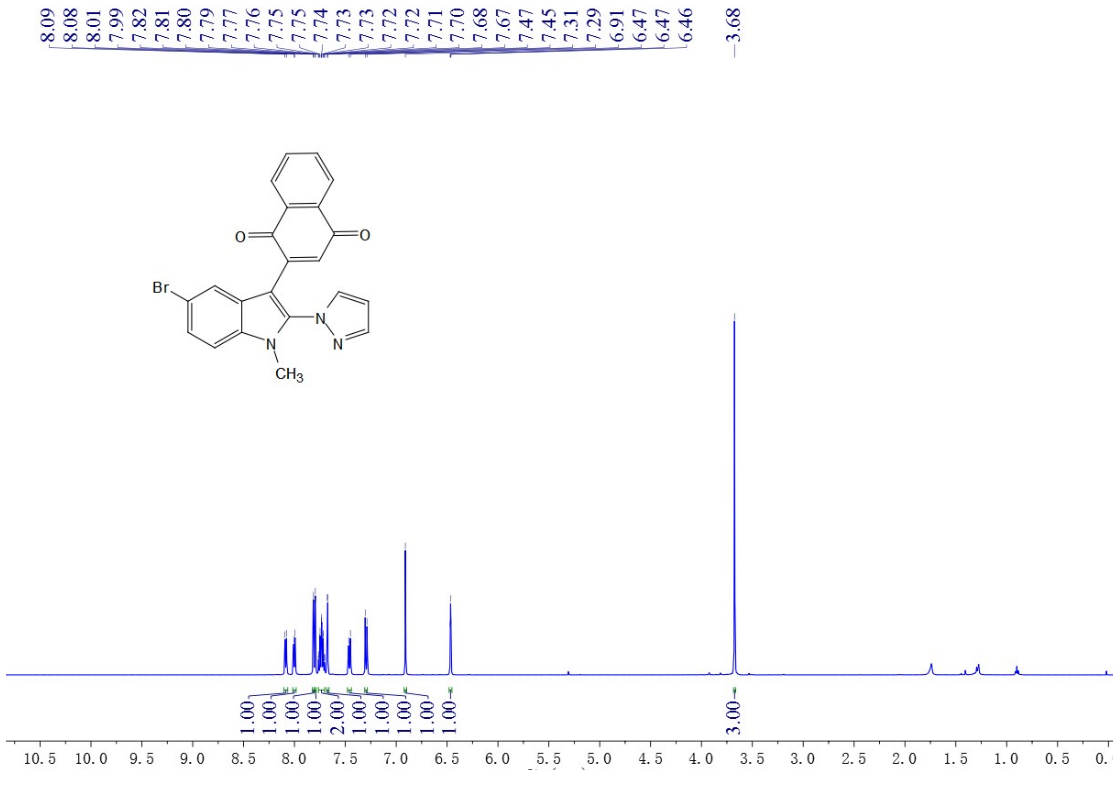
(24) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4jaa



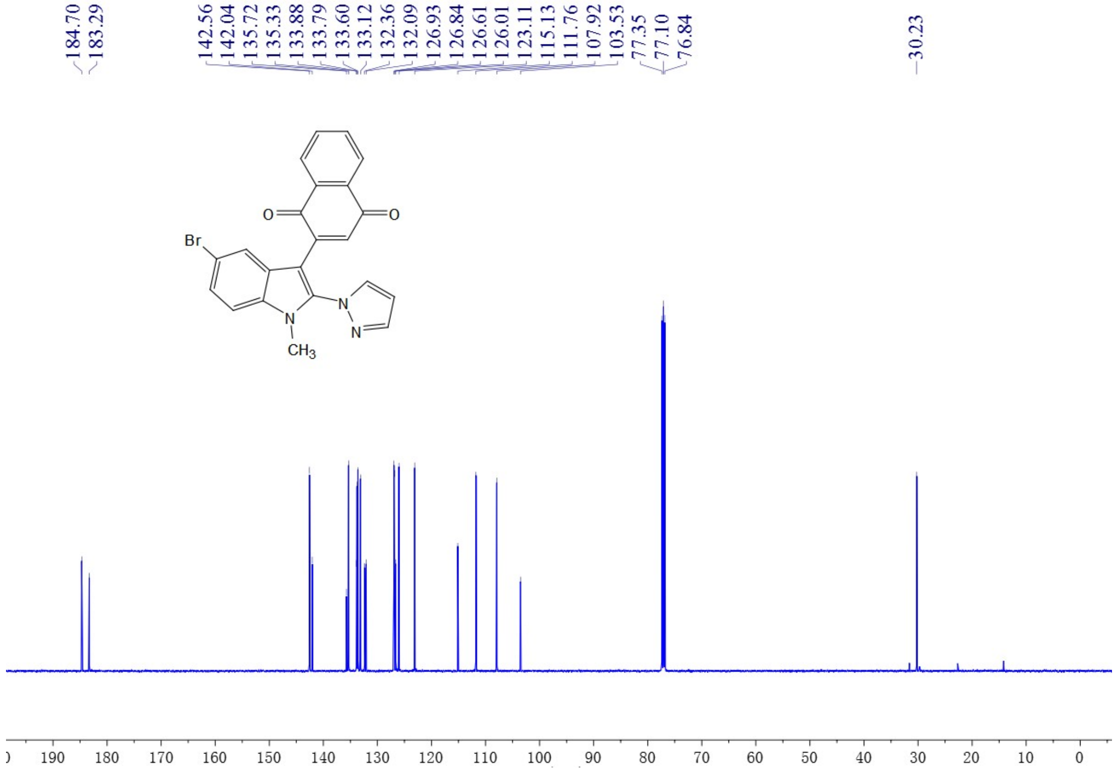
(25) ^{13}C-NMR (125 MHz, CDCl$_3$) spectrum of 4jaa



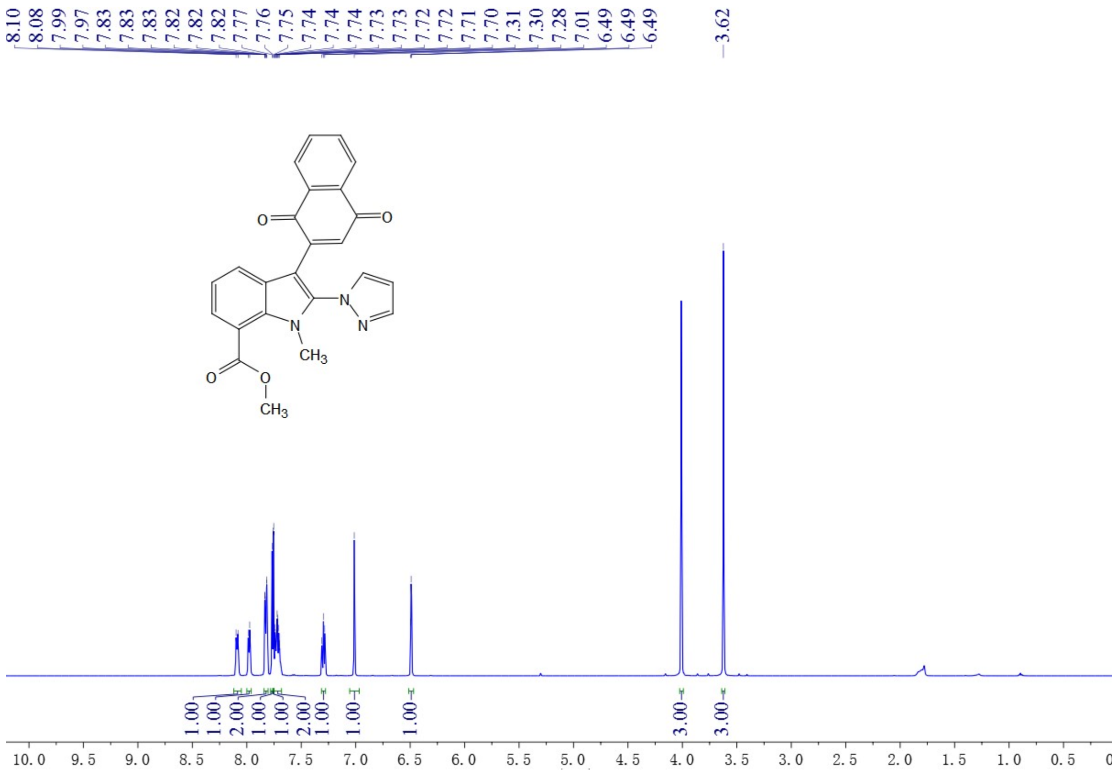
(26) ^{1}H-NMR (500 MHz, CDCl$_3$) spectrum of 4laa



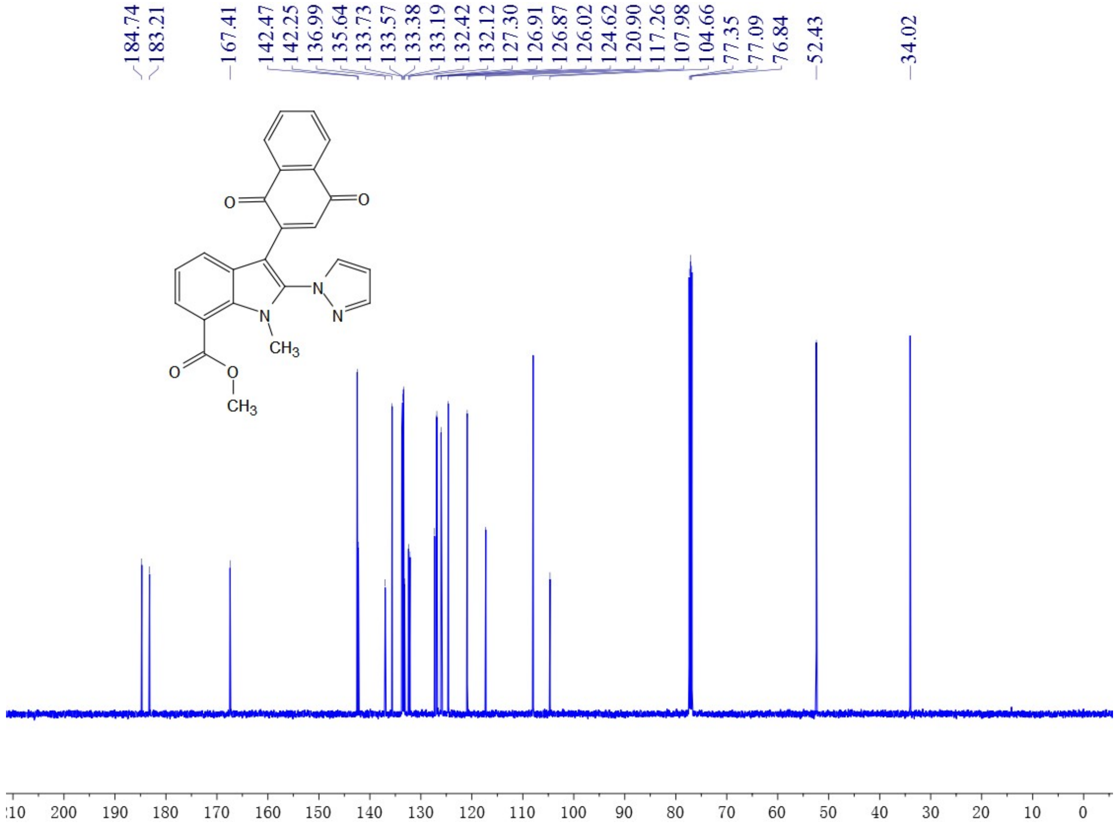
(27) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4laa



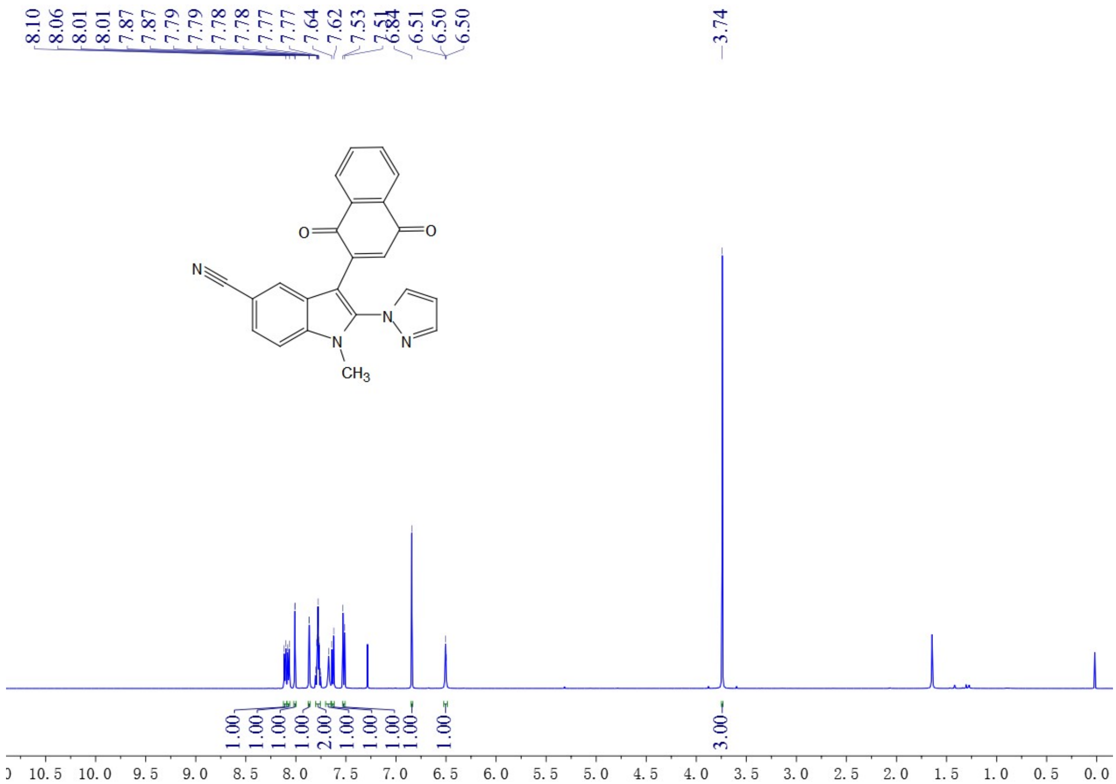
(28) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4maa



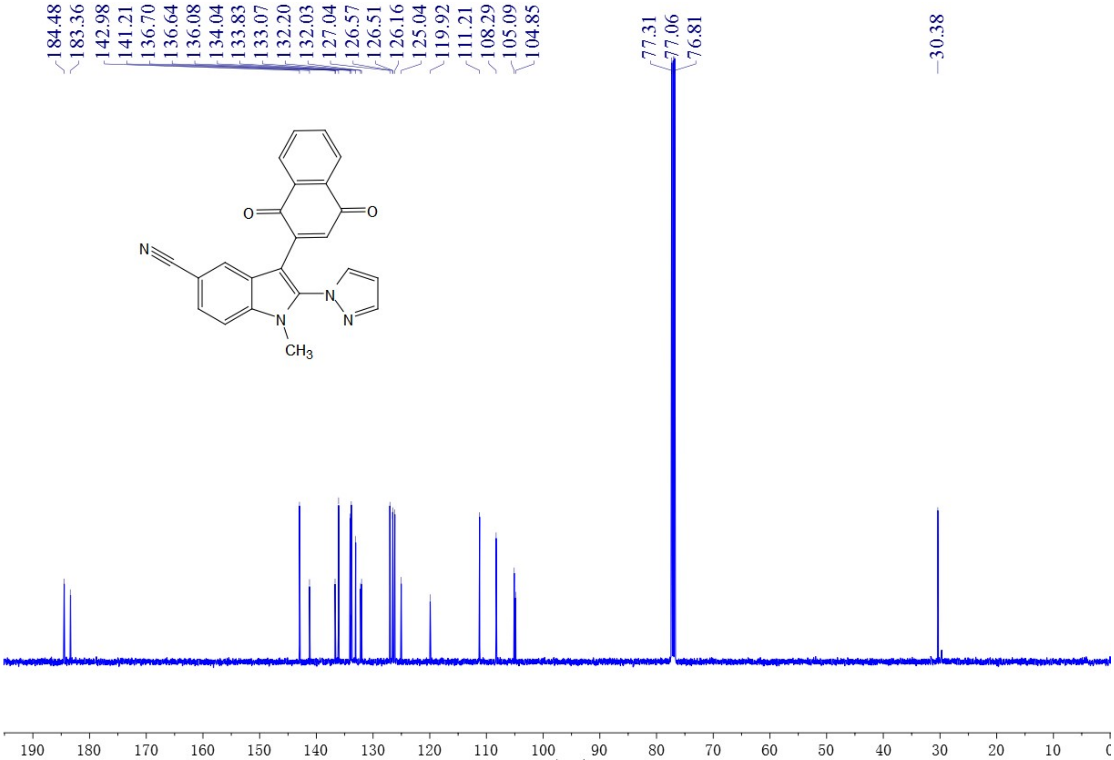
(29) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4maa



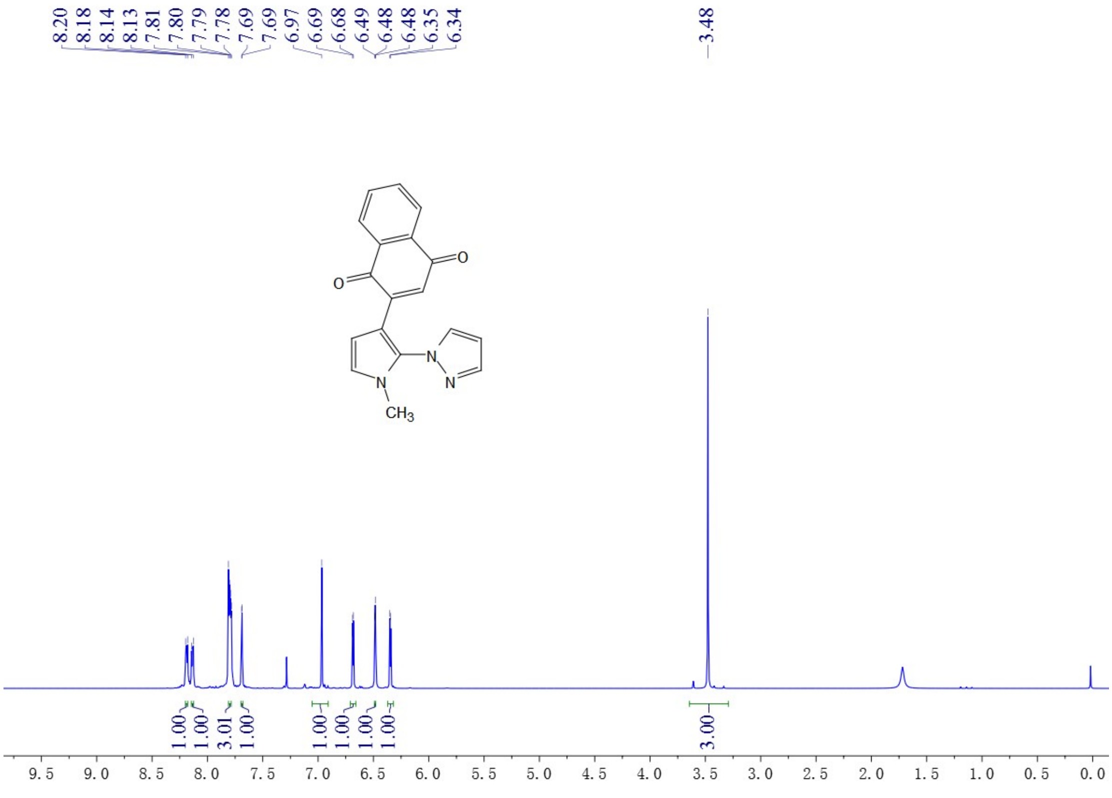
(30) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4naa



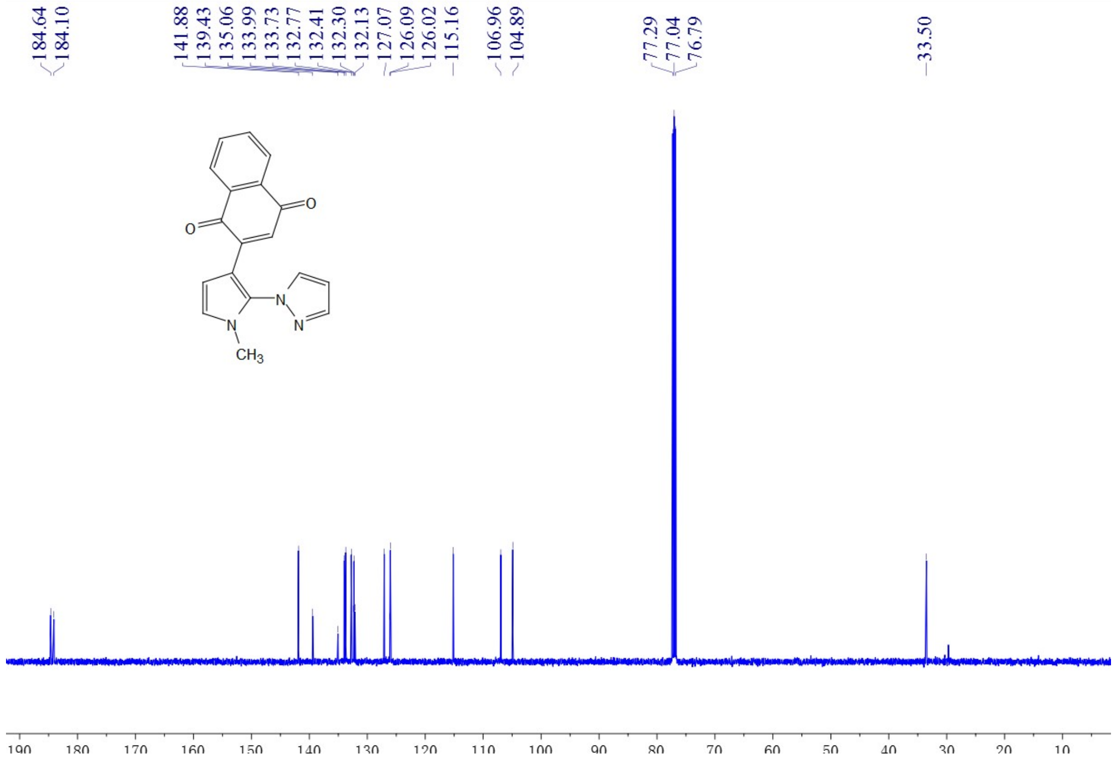
(31) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4naa



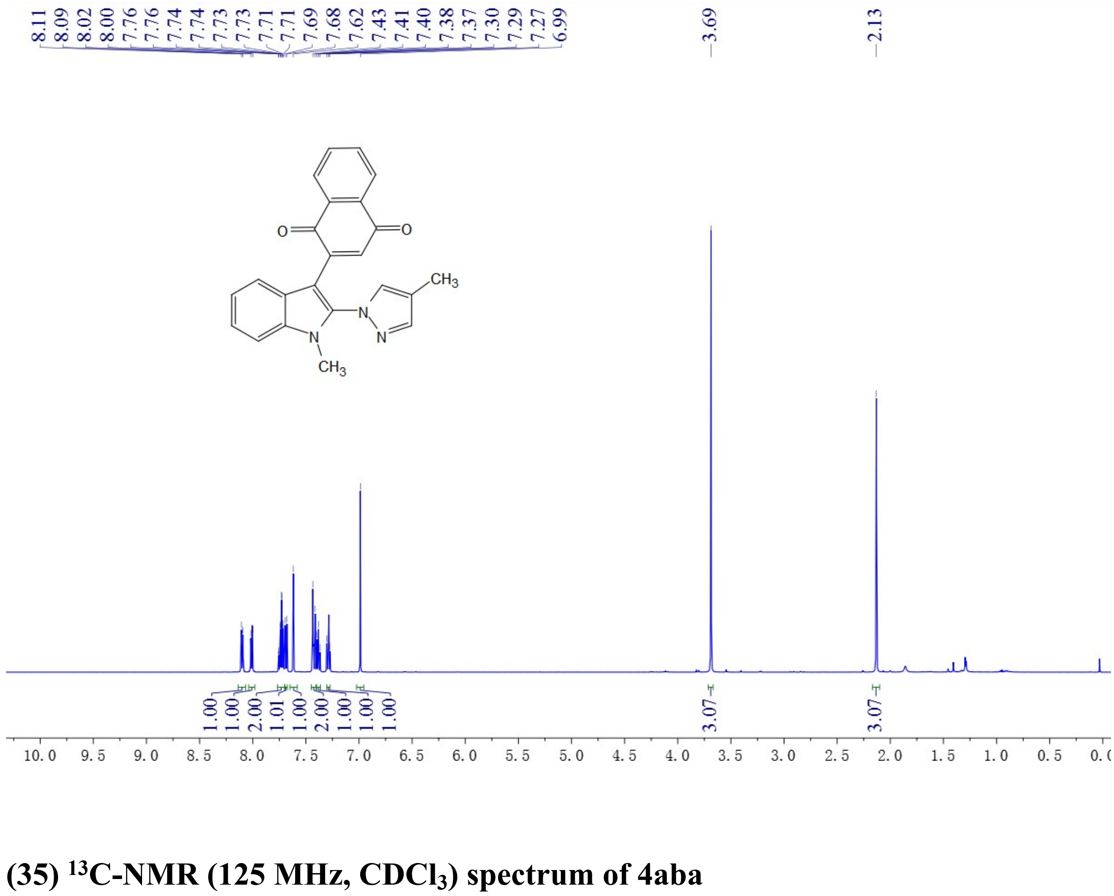
(32) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4oaa



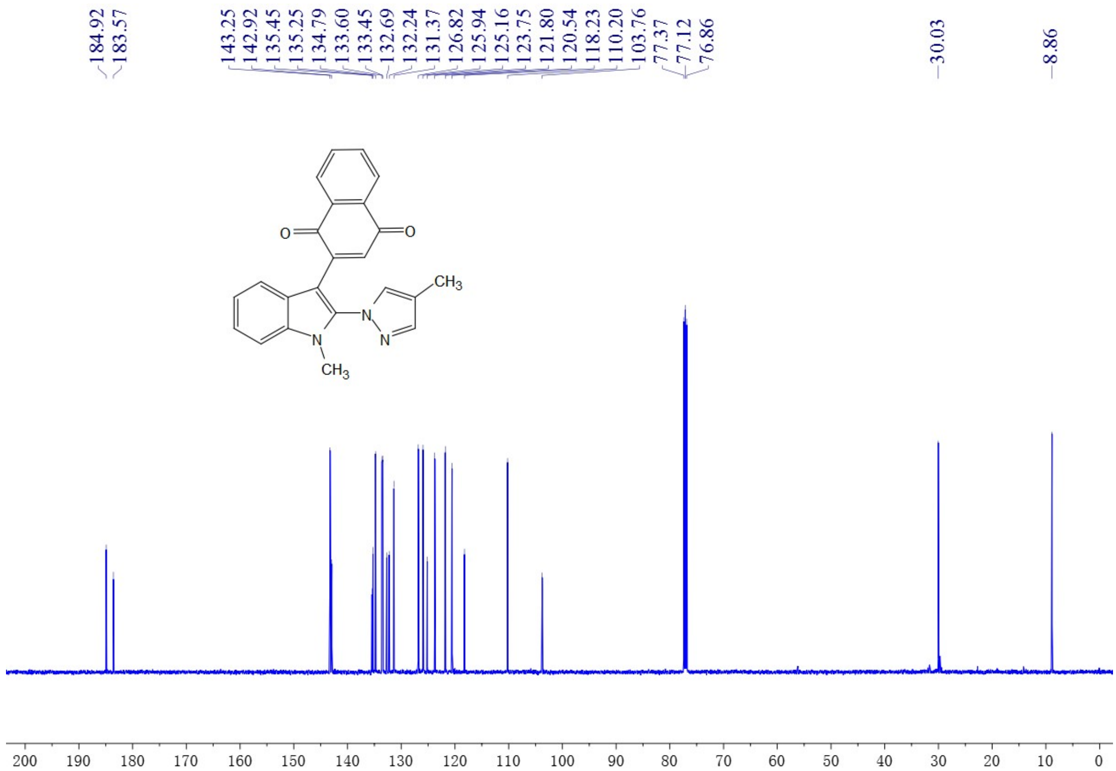
(33) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4oaa



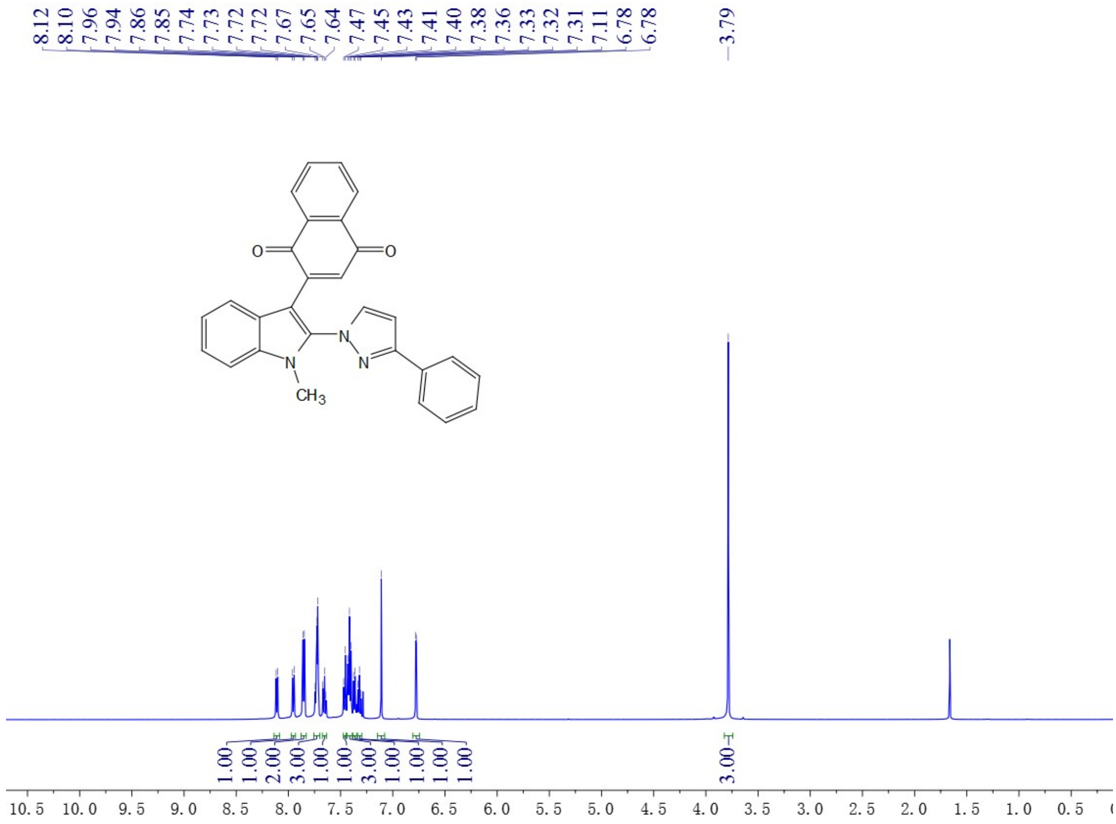
(34) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4aba



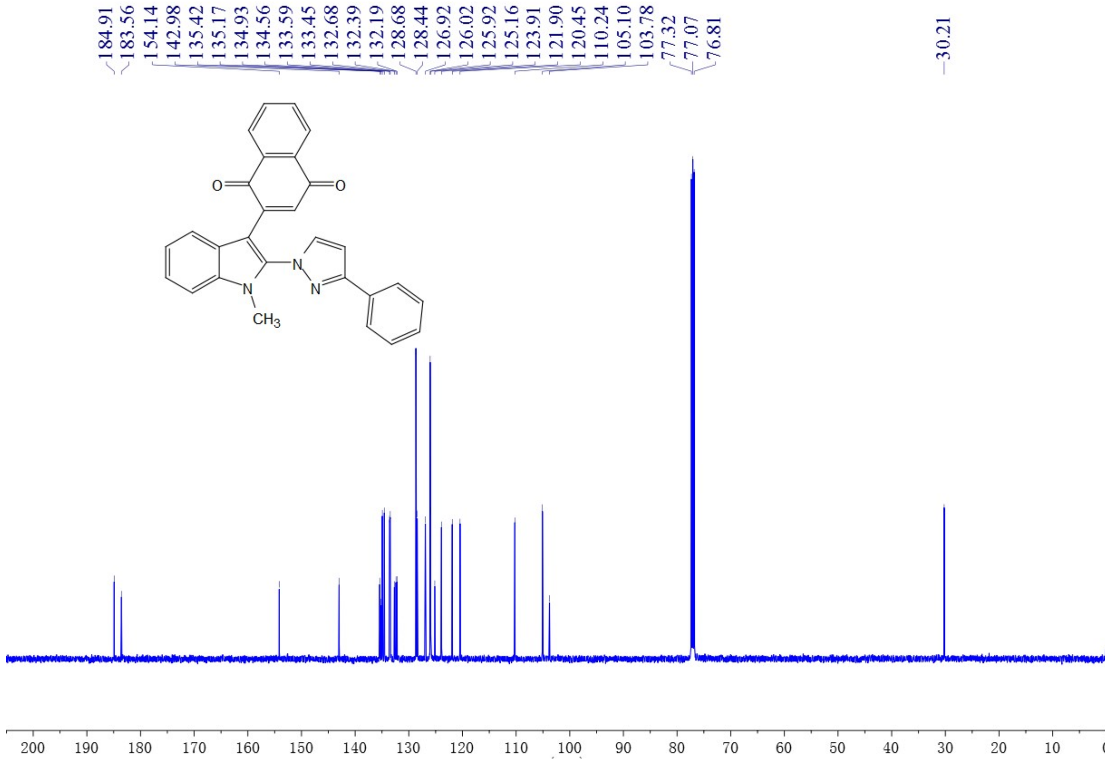
(35) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4aba



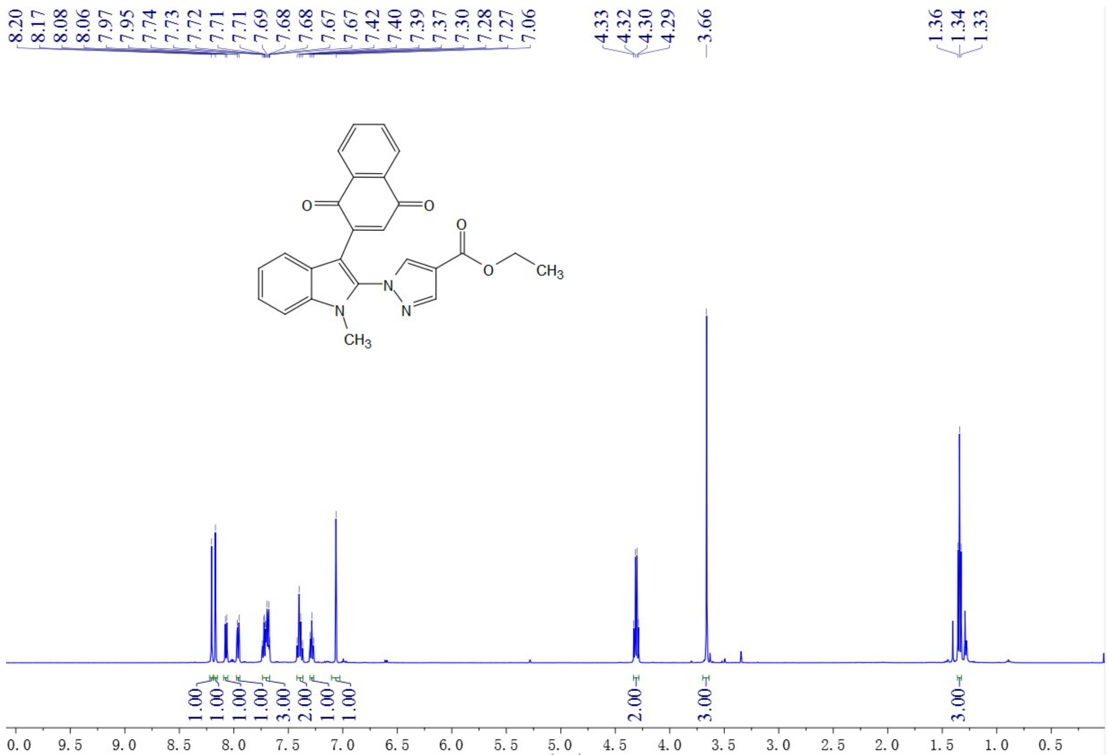
(36) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4aca



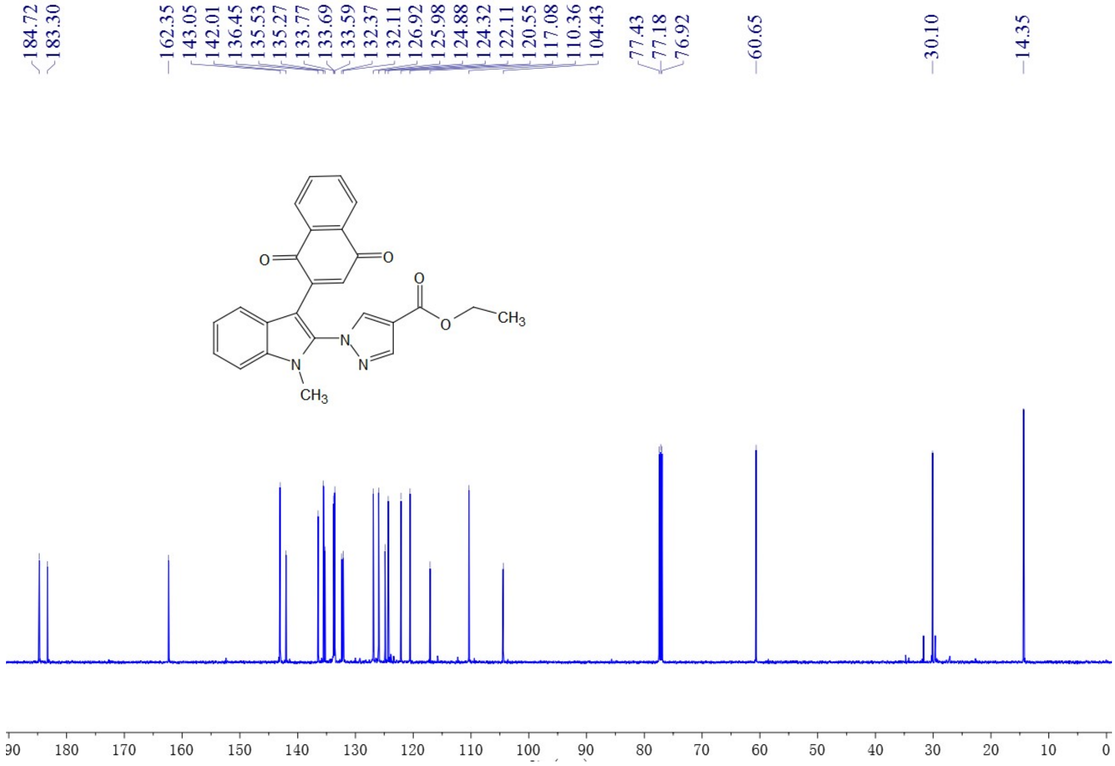
(37) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4aca



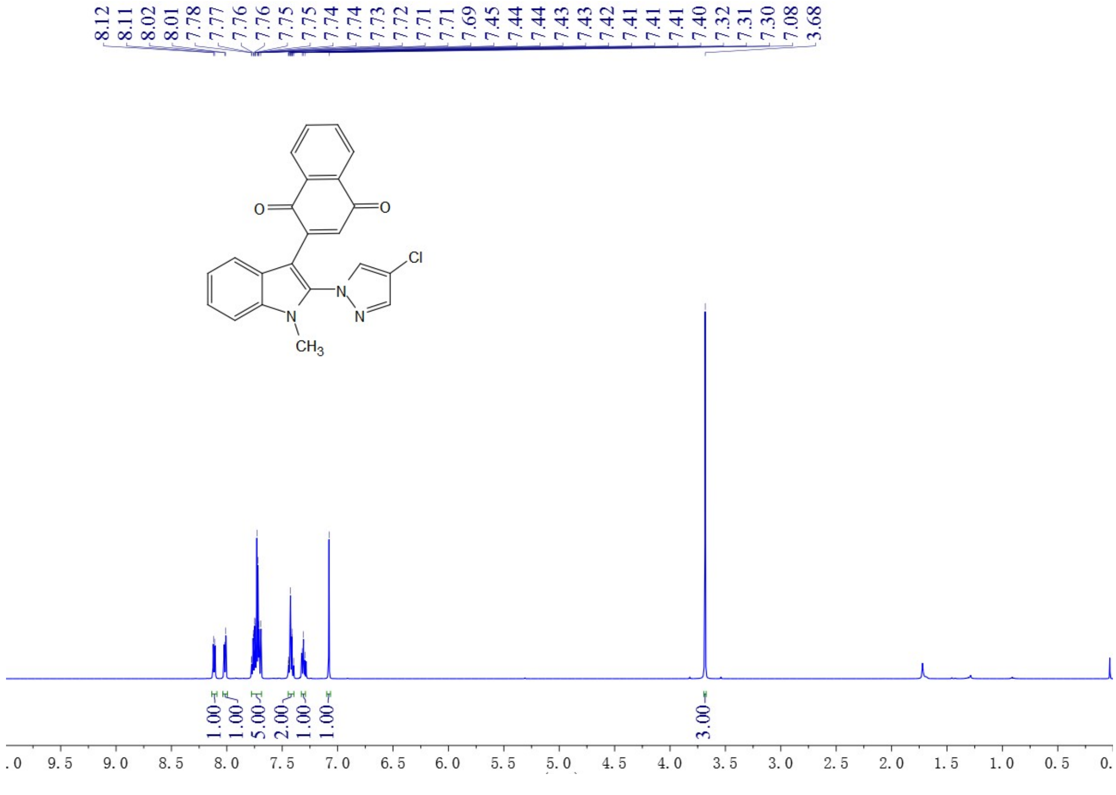
(38) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4ada



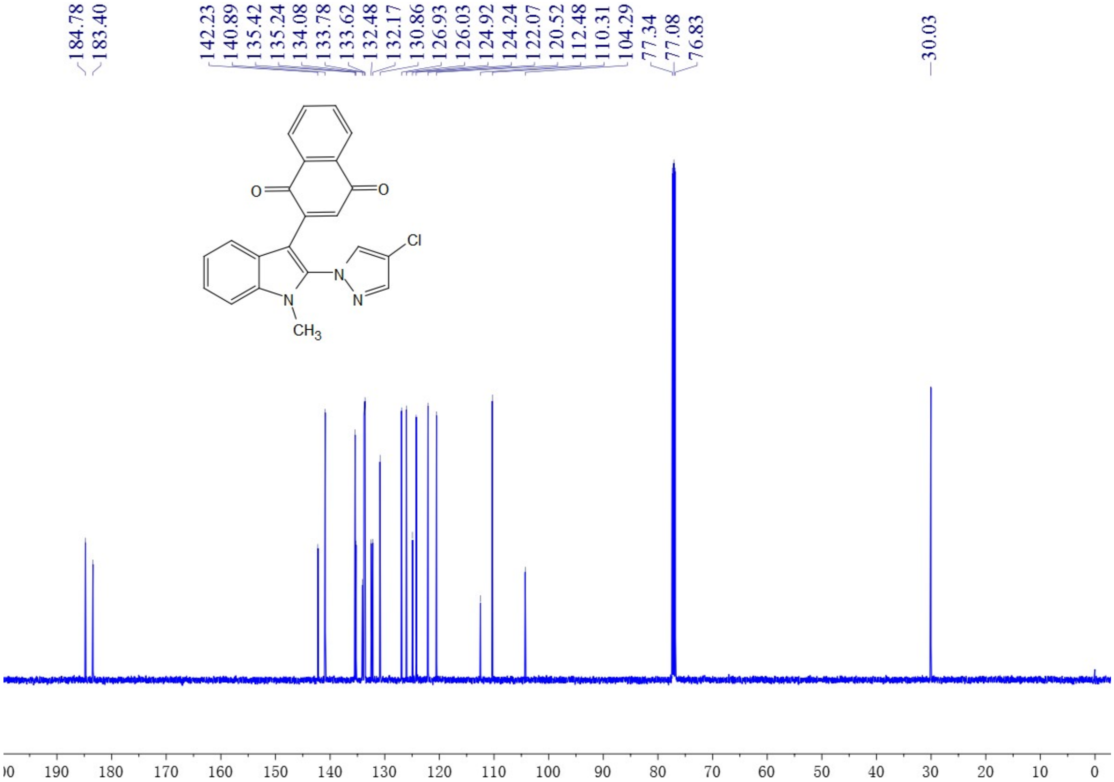
(39) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4ada



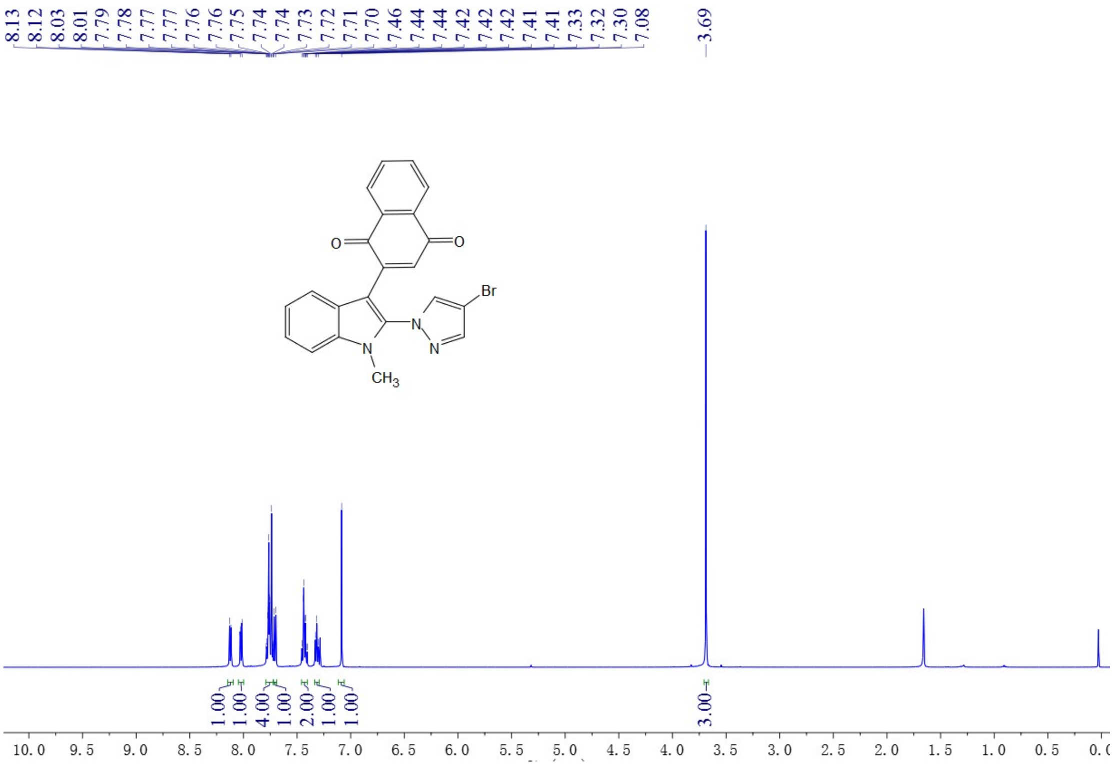
(40) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4afa



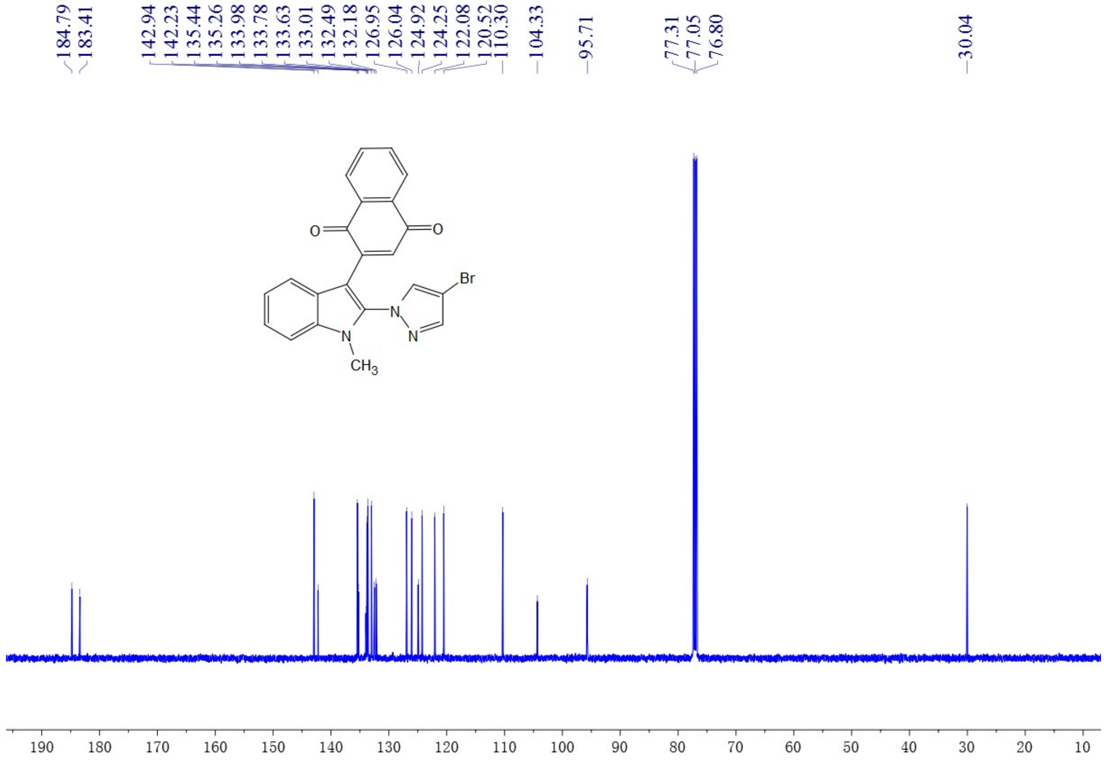
(41) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4afa



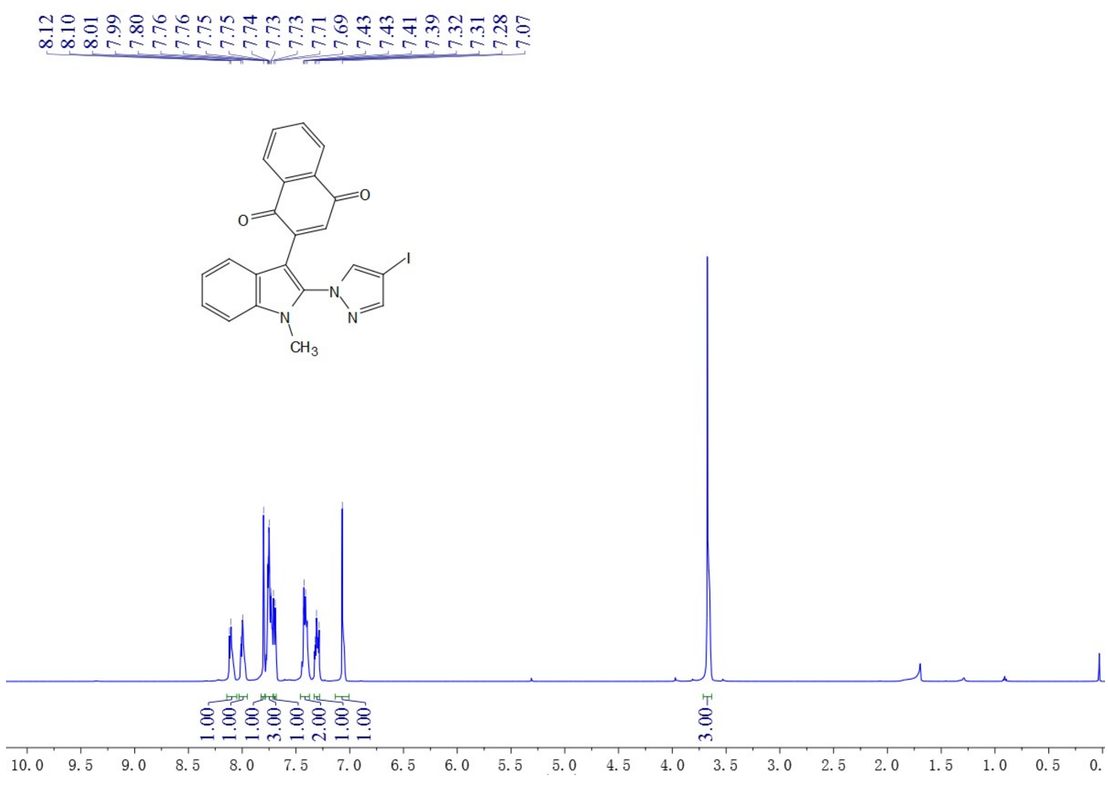
(42) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4aga



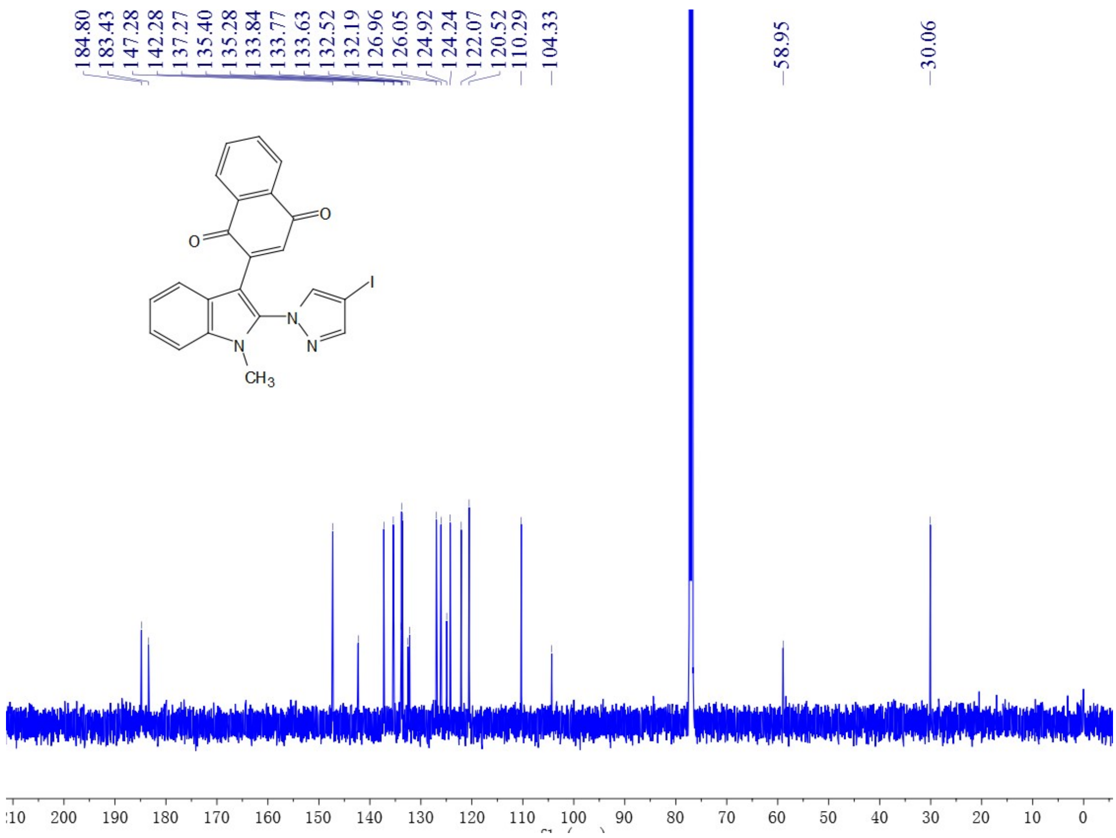
(43) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4aga



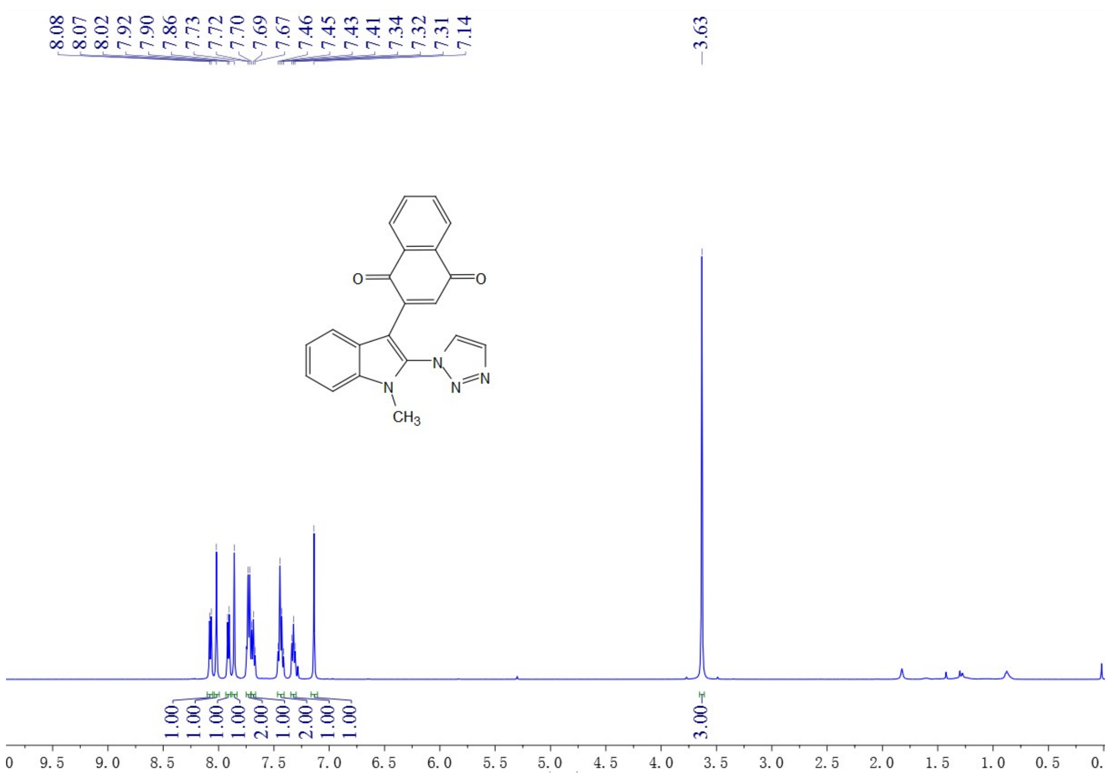
(44) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4aha



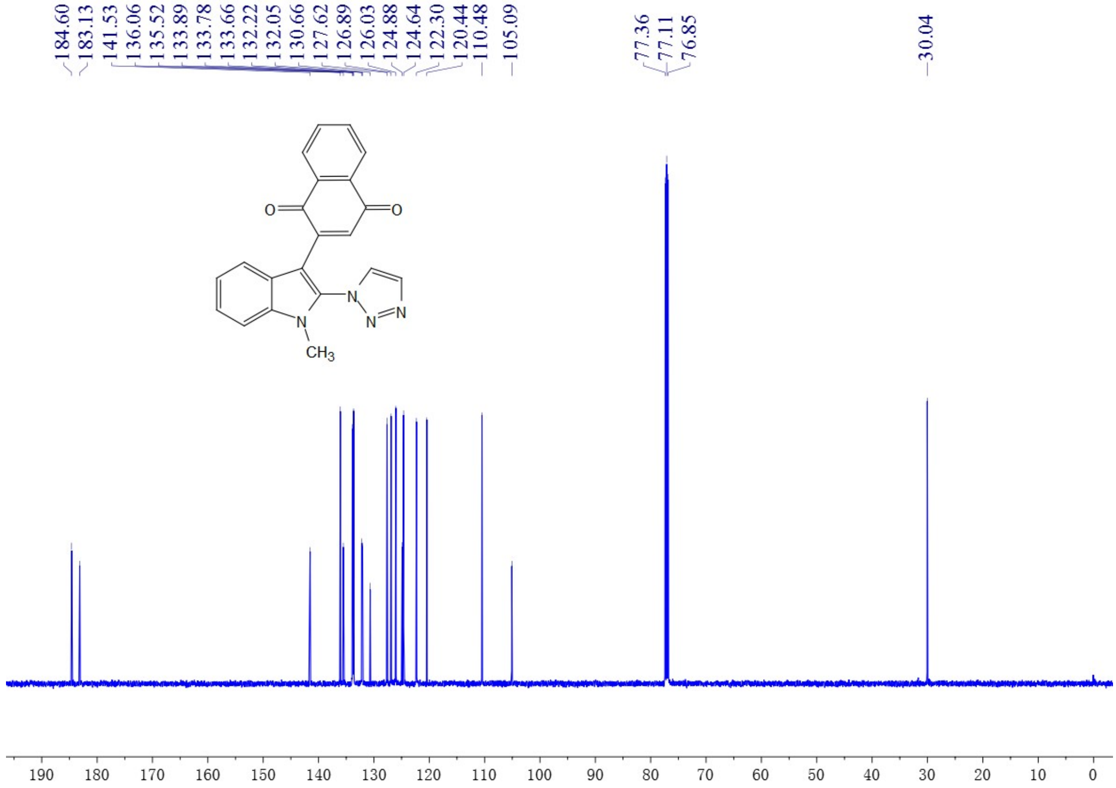
(45) ^{13}C-NMR (125 MHz, CDCl$_3$) spectrum of 4aha



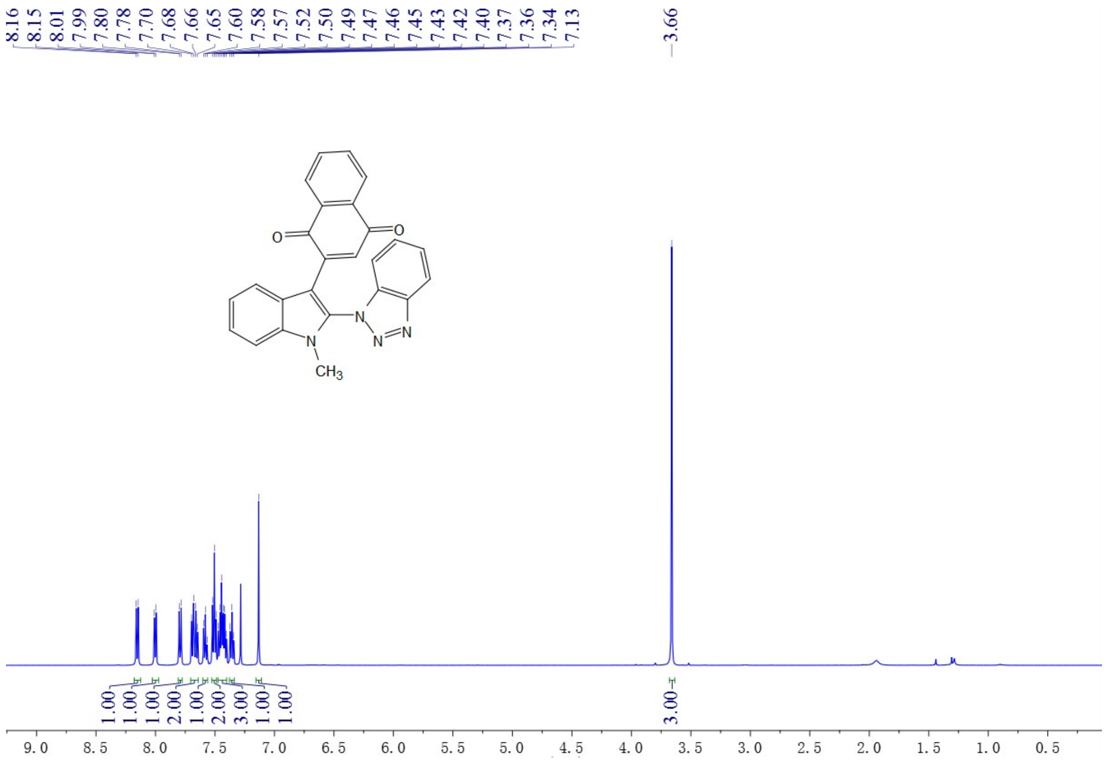
(46) ^{1}H-NMR (500 MHz, CDCl$_3$) spectrum of 4aia



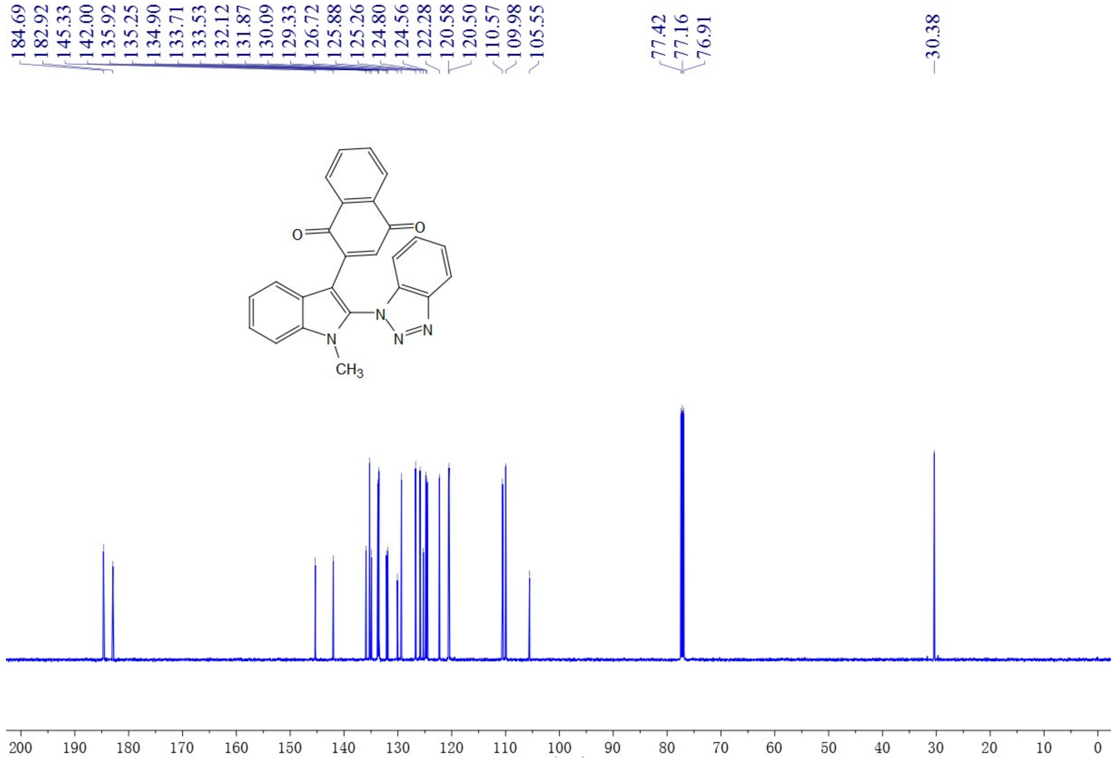
(47) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4aia



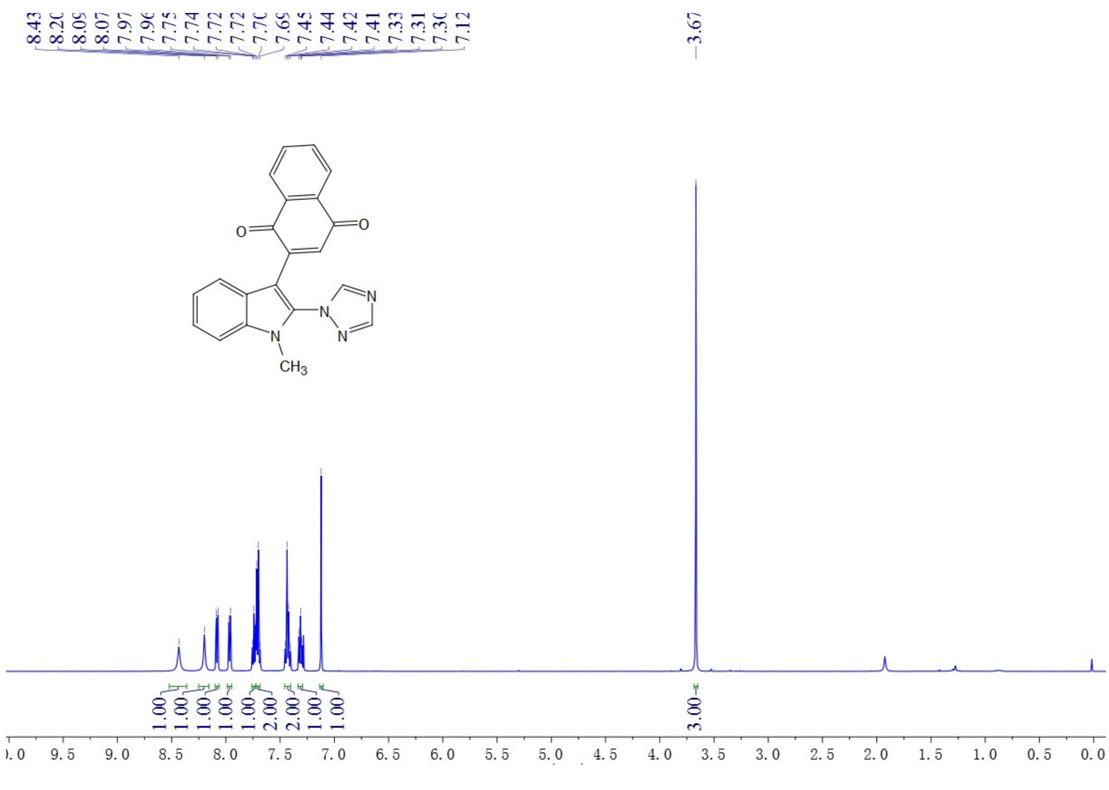
(48) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4aja



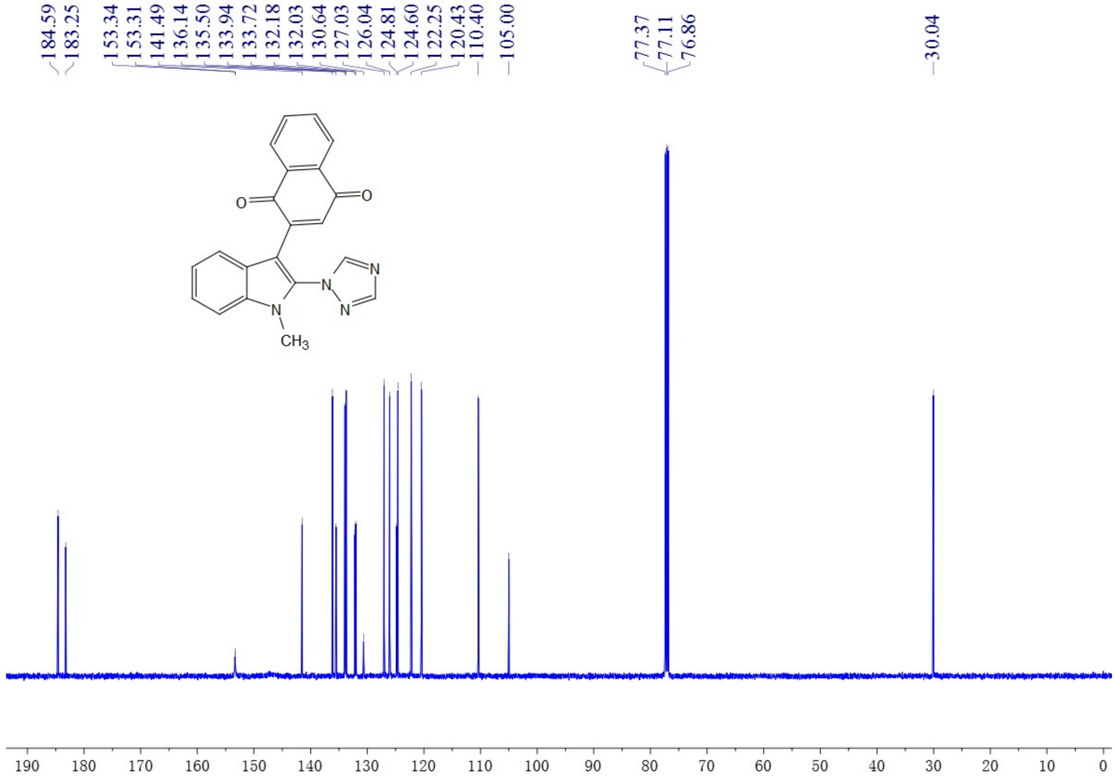
(49) ^{13}C-NMR (125 MHz, CDCl$_3$) spectrum of 4aja



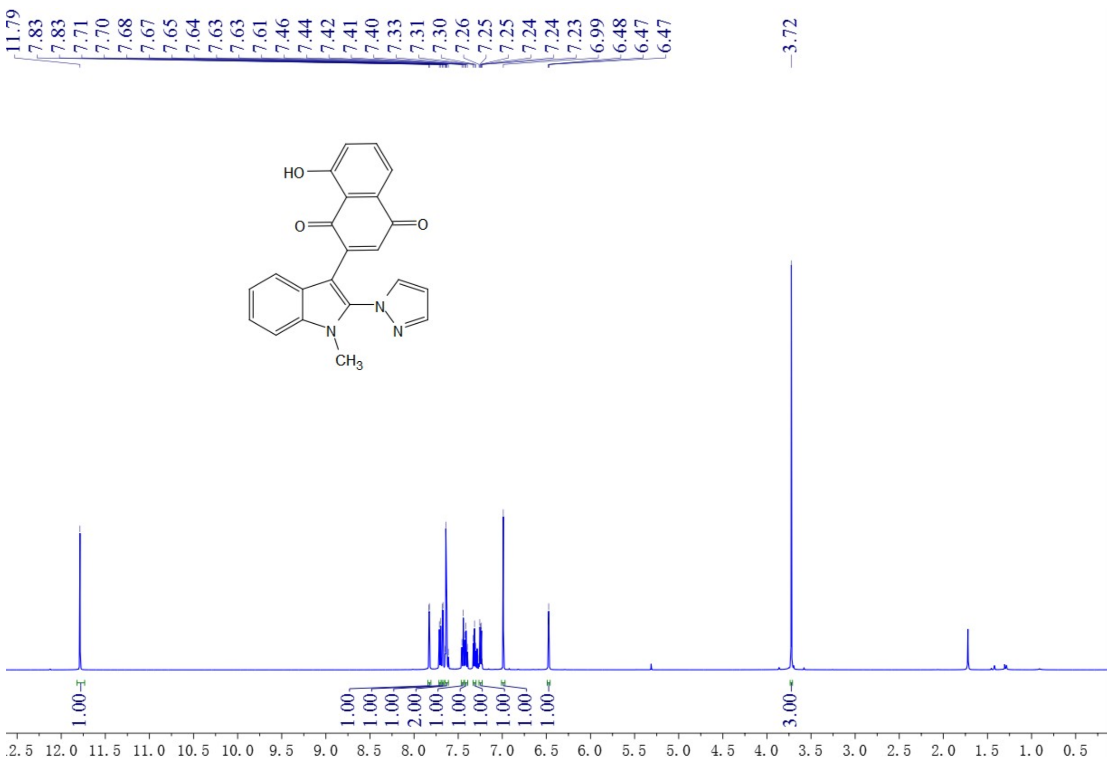
(50) ^{1}H-NMR (500 MHz, CDCl$_3$) spectrum of 4aka



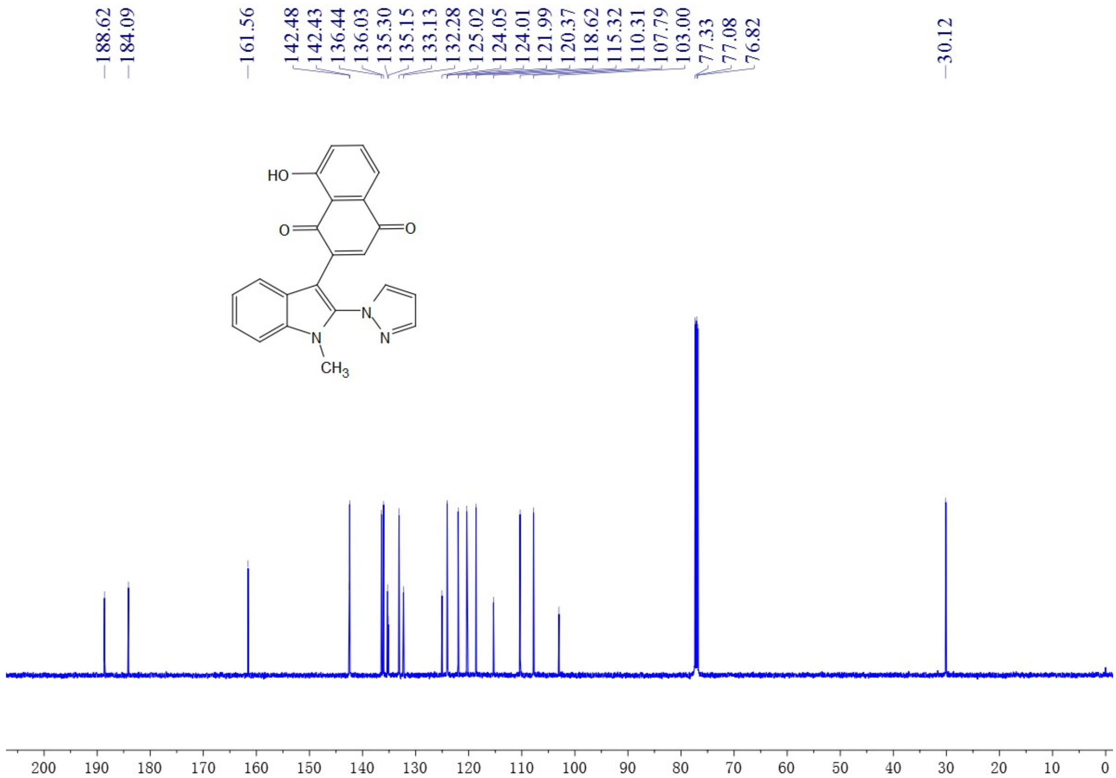
(51) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4aka



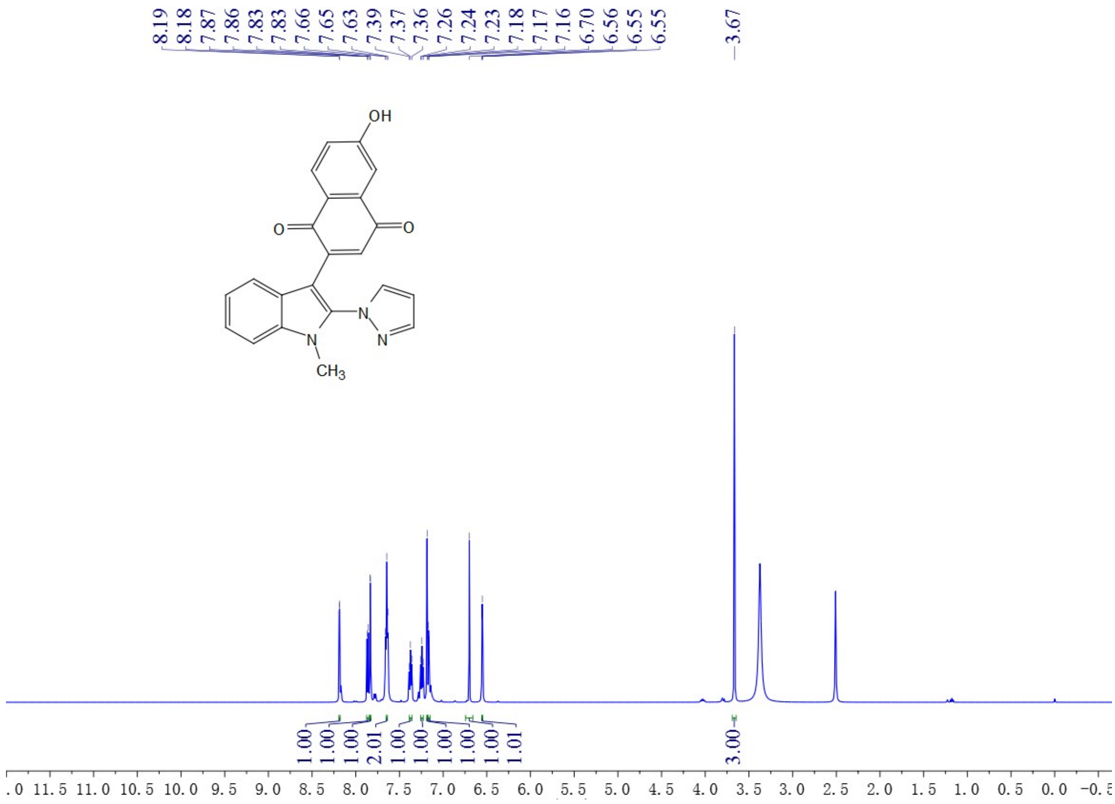
(52) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4aab



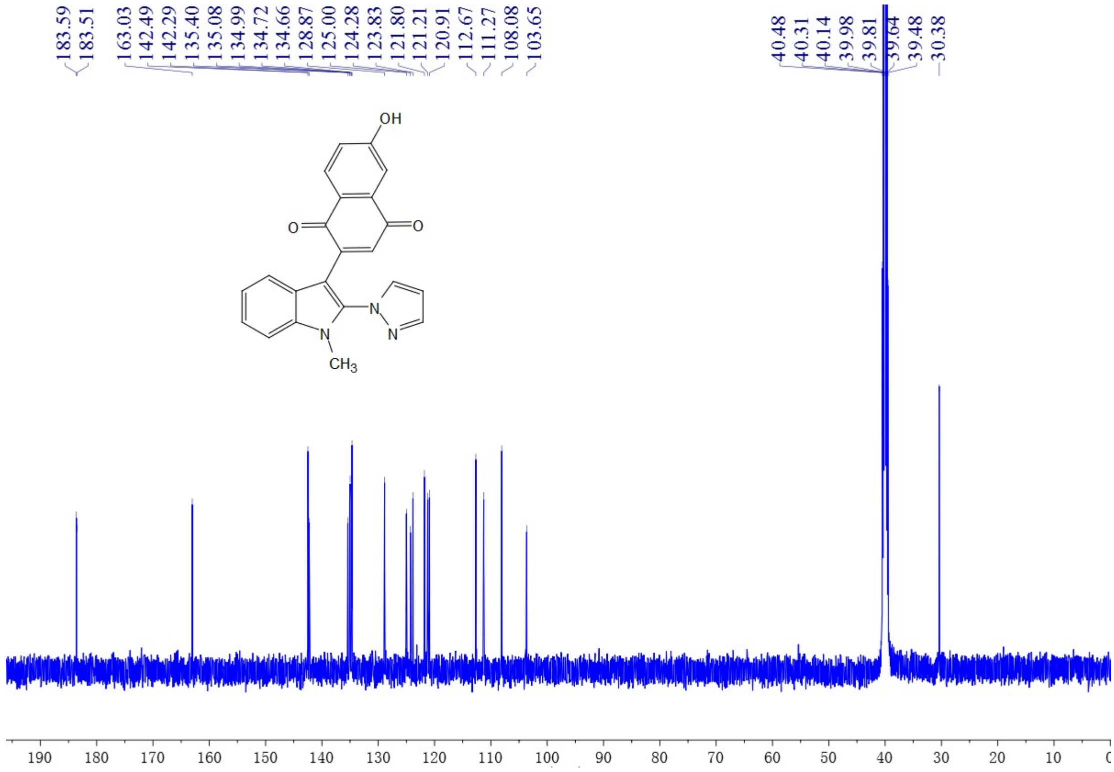
(53) **^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4aab**



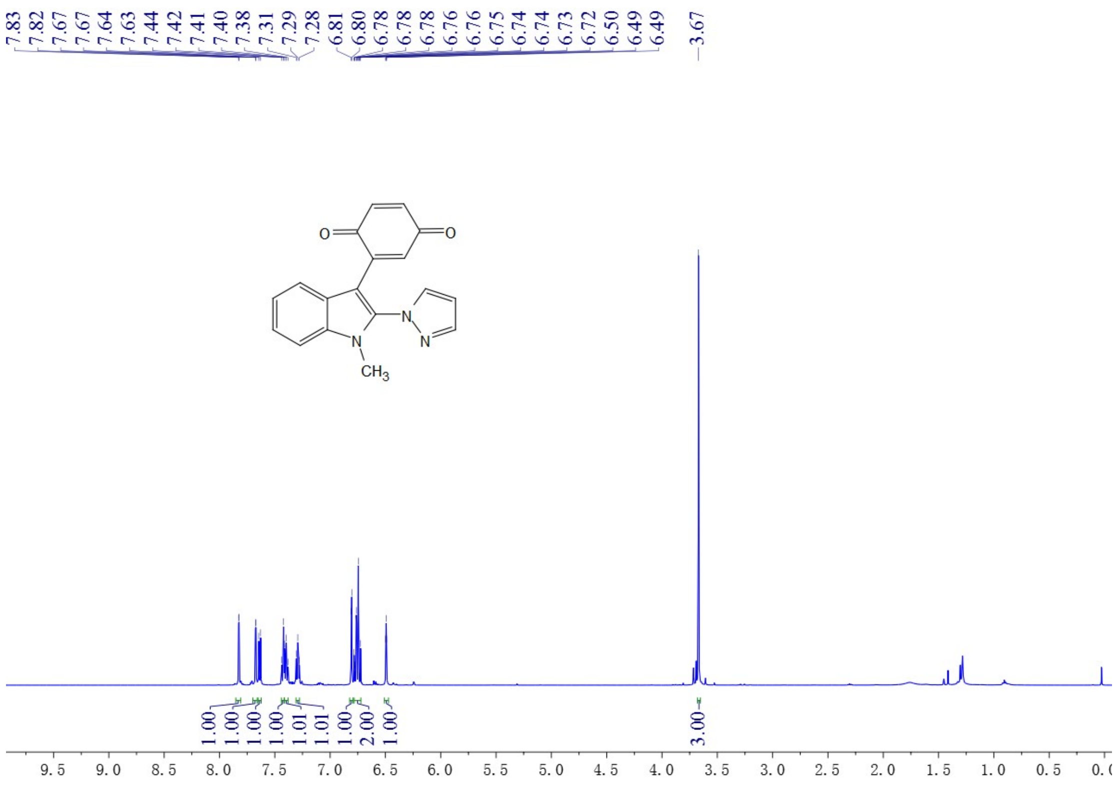
(54) **^{1}H-NMR (500 MHz, DMSO) spectrum of 4aac**



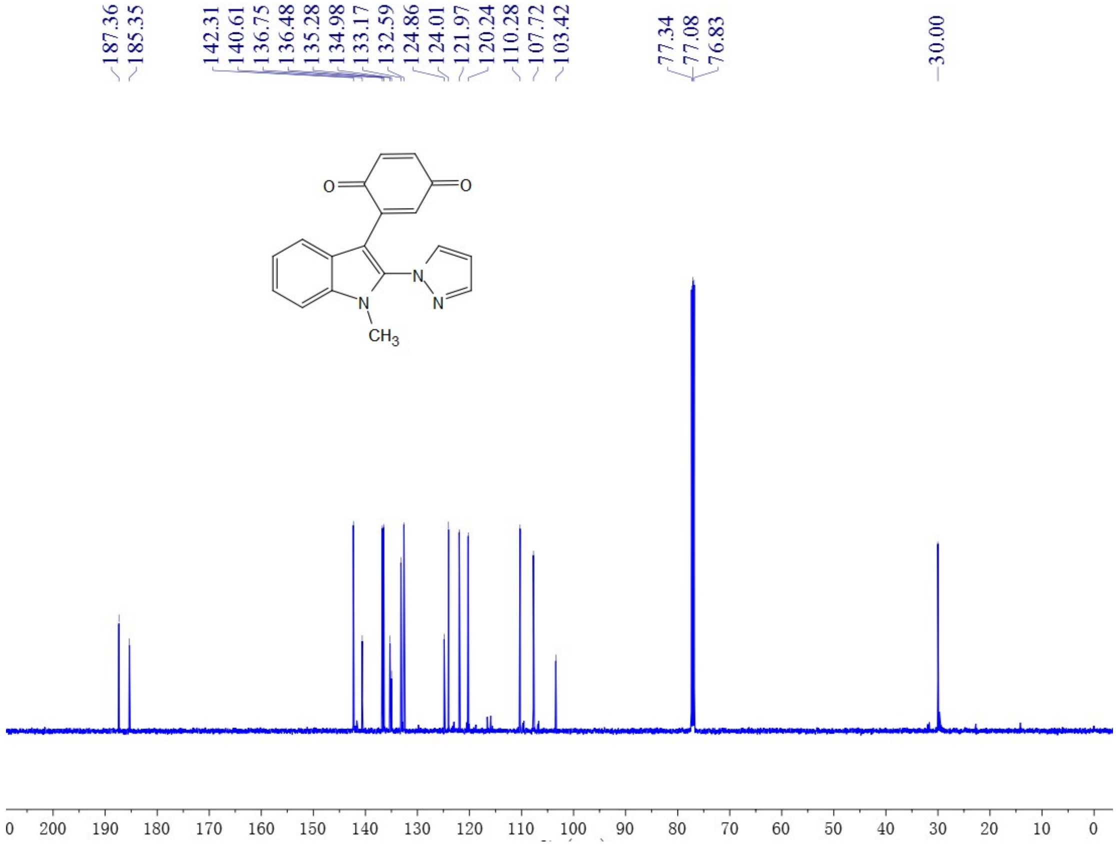
(55) ^{13}C-NMR (125 MHz, DMSO) spectrum of 4aac



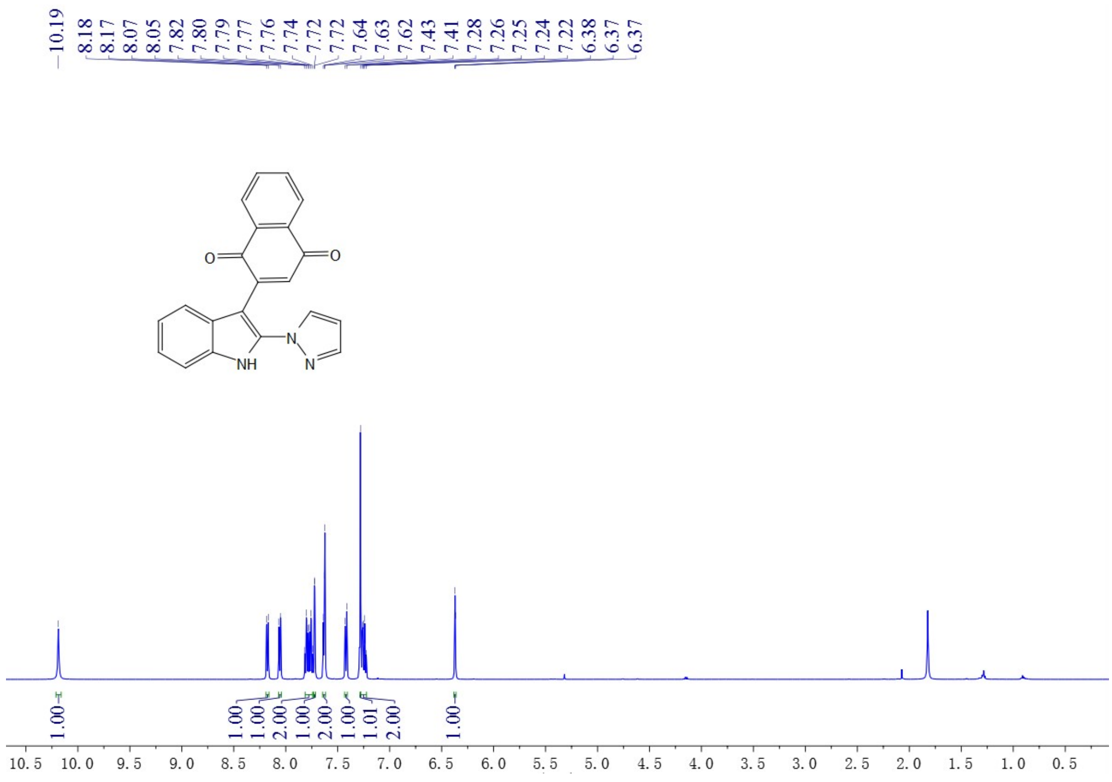
(56) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4aad



(57) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4aad



(58) ^{1}H-NMR (500 MHz, $CDCl_3$) spectrum of 4paa



(59) ^{13}C-NMR (125 MHz, $CDCl_3$) spectrum of 4paa