Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.

Electro-catalytic multicomponent reaction to asymmetrical biaryls from heteroarylation of in-situ generated fused polycyclic heteroaromatics

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1、General Information

Commercially available reagents and solvents are of reagent grade quality without further purification. Flash column chromatography is performed using silicycle silica gel (200-300 mesh). Analytical thin-layer chromatography (TLC) is performed on 0.2 mm coated silica gel plates (HSGF 254) and visualized using a UV lamp (254 nm). ^{1}H NMR and ^{13}C NMR are recorded on magnet system 400’54 ascend purchased from Bruker Biospin AG. HRMS (ESI) spectra are recorded on Agilent Q-TOF 6520.

Catalyst- and oxidant-free multicomponent reaction to asymmetrical biaryls from electrochemical arylation of in-situ generated fused polycyclic heteroaromatics was carried out in an undivided cell equipped with a carbon cloth anode and a platinum plate cathode under open air. The carbon cloth, graphite rod (⌀ 6 mm) and platinum plate were purchased from Shanghai Jing Chong Electronic Technology Development Co., Ltd. Reticulated vitreous carbon (RVC) was purchased from Gaoss Union (Tianjin) Photoelectric Technology Co., Ltd. And, electrolysis was conducted under an AXIOMET AX3003P potentiostat in constant current mode. Cyclic voltammogram experiments were investigated using a Metrohm Autolab PGSTAT204 workstation and Nova 2.0 software.

2、General Material Information for Electrolysis Setup

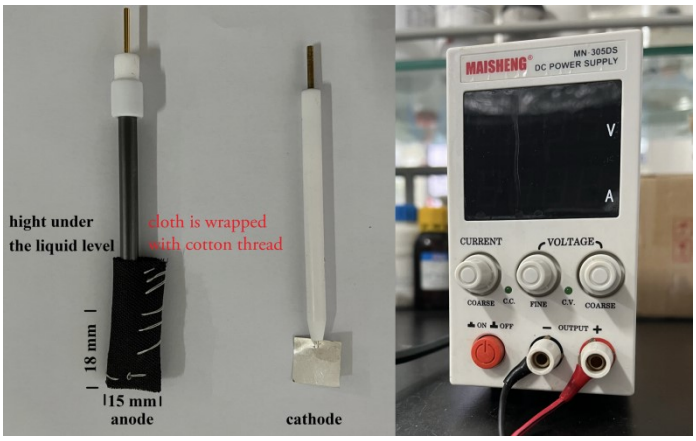
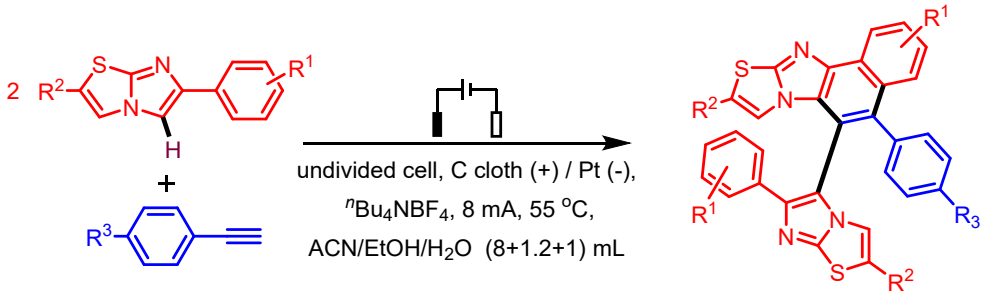


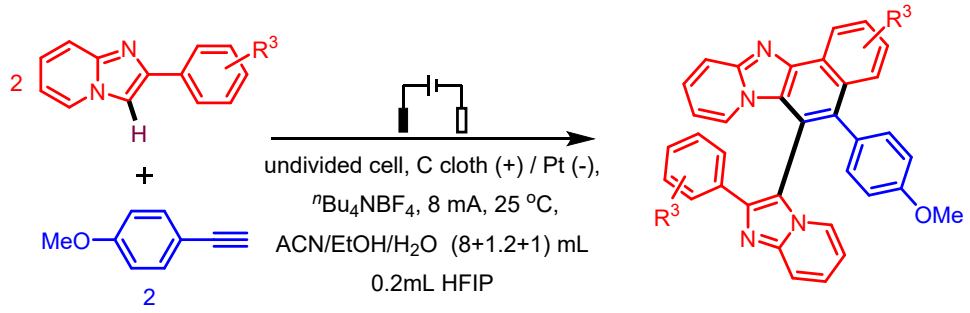
Figure S1 Picture of DIY electrolysis setup

The DIY electrolysis setup used is shown in Fig. **S1**.

3、General Procedure for Electrosynthesis of Asymmetrical Biaryls



In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, imidazo[2,1-*b*]thiazoles (0.2 mmol), alkyne (0.2 mmol), ^{n}Bu$_4$NBF$_4$ (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of ACN/EtOH/H$_2$O (8+1.2+1) mL. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 1.5 h under the open air. The reaction solution was diluted with ethyl acetate (50 mL) and washed with H$_2$O (50 mL). The separated organic layer was dried over anhydrous Na$_2$SO$_4$ and filtered. The filtrate was concentrated under reduced pressure to give the crude product, which was purified by column chromatographic separation (dichloromethane/ethyl acetate/petroleum ether: 10:2:30 to 10:22:30) to obtain the desired product.



In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 2-phenylimidazo[1,2-*a*]pyridines (0.2 mmol), 1-ethynyl-4-methoxybenzene **2** (0.2 mmol, 26.4 mg), ^{n}Bu$_4$NBF$_4$ (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of ACN/EtOH/H$_2$O (8+1.2+1) mL. Then, 0.2 mL HFIP was added. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 1.5-2 h under the open air. The reaction solution was diluted with ethyl acetate (50 mL) and washed with H$_2$O (50 mL). The separated organic layer was dried

over anhydrous Na_2SO_4 and filtered. The filtrate was concentrated under reduced pressure to give the crude product, which was purified by column chromatographic separation (dichloromethane/ethyl acetate/petroleum ether: 10:20:30) to obtain the desired product.

4、Optimization of the Reaction of 2-Phenylimidazo[1,2-*a*]pyridine with 2[a]

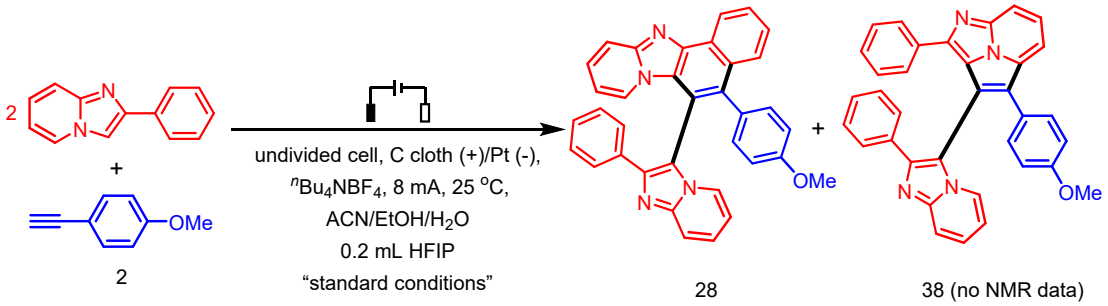
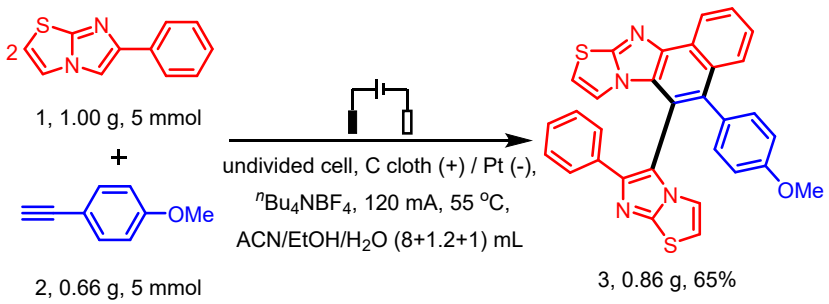


Table S1 Optimization of the reaction of 2-phenylimidazo[1,2-*a*]pyridine with **2**[a]

Entry	Variation from "standard conditions"	Yield[b] (%)	
		28	38
1	none	42[c]	18
2	no HFIP	32	15
3	no H_2O	trace	35
4	$LiClO_4$ instead of ^{n}Bu$_4$NBF$_4$	34	15
5	40 °C	20	8
6[d]	RVC as anode	32	12

[a]Reaction conditions: undivided cell, 2-phenylimidazo[1,2-*a*]pyridine (0.2 mmol, 38.8 mg), **2** (0.2 mmol, 26.4 mg), ^{n}Bu$_4$NBF$_4$ (0.2 mmol, 65.8 mg), ACN/EtOH/H$_2$O (8+1.2+1) mL, 0.2 mL HFIP, 8 mA, 25 °C, 1.5-2 h, under the open air, C cloth (35 × 15 mm), Pt (1.0 × 1.0 cm), C rod (⌀ 6 mm), RVC (reticulated vitreous carbon, 1.0 × 1.0 cm), ACN: acetonitrile, EtOH: ethyl alcohol, HFIP: 1,1,1,3,3,3-hexafluoro-2-propanol. [b]Yields were determined by ^{1}H NMR using dibromomethane as the internal standard. [c]Isolated yield. [d]Reaction at 75 °C.

5、Gram-Scale Synthesis of 3



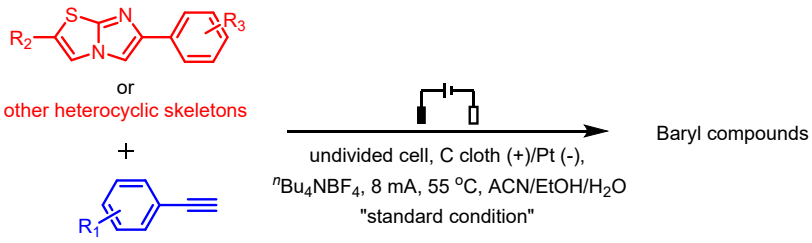
The 6-phenylimidazo[2,1-*b*]thiazole **1** (5 mmol, 1.00 g), $^{n}Bu_4NBF_4$ (3 mmol, 0.99 g), 1-ethynyl-4-methoxybenzene **2** (5 mmol, 0.66 g), ACN (120 mL) and EtOH (18 mL) and H_2O (15 mL) was added in a 250 mL beaker, which was equipped with a stir bar, a carbon cloth (100 mm × 50 mm) anode and a platinum plate (20 mm × 20 mm × 0.1 mm) cathode. The solution was stirred and electrolyzed at a constant current of 120 mA for 5 h under the open air. The reaction solution was diluted with ethyl acetate (200 mL) and washed with brine (200 mL) and H_2O (200 mL). The separated organic layer was dried over anhydrous Na_2SO_4 and filtered. The filtrate was concentrated under reduced pressure to give the crude product, which was purified by column chromatographic separation (dichloromethane/ethyl acetate/petroleum ether: 10:20:30, 0.86 g, 65%).

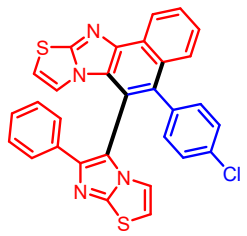
6、Unsuccessful substrates

6-1、Unsuccessful substrates

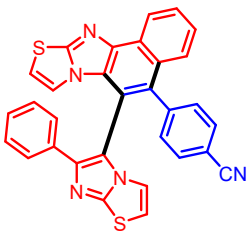
Some other electron-deficient substituted alkynes or R_2 were substituted by electron-withdrawing group could not work well in the system and poor yields were achieved due to the incomplete conversion of the substrates (**S-1**, **S-5**). No desired product was observed with much starting material unconverted due to the decrease of alkyne activity when R_1 was substituted by 4-CN (**S-2**).

Table **S2** The list of unsuccessful substrates.

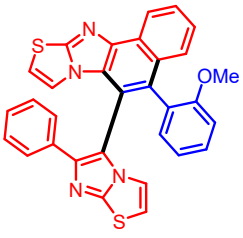




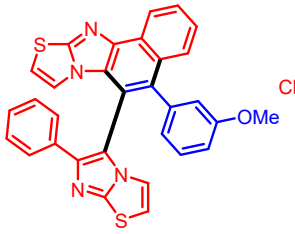
S-1, trace



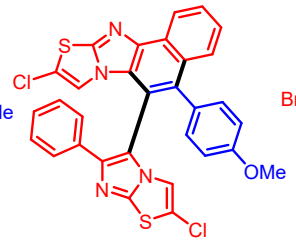
S-2, no



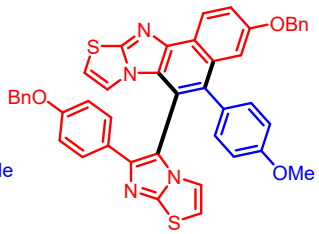
S-3, no



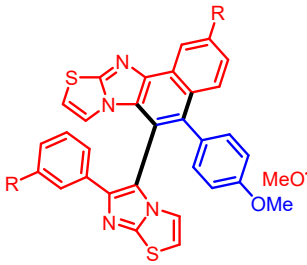
S-4, no



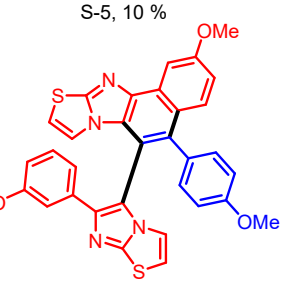
S-5, 10 %



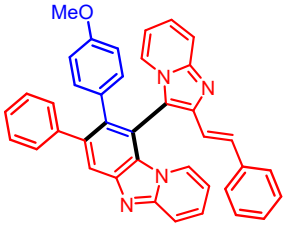
S-6, no



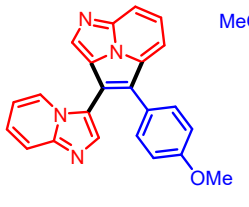
S-7, R=CH_3, S-8, R=F



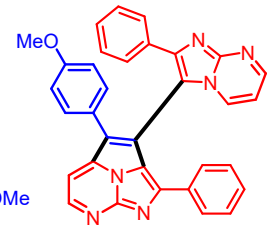
S-9



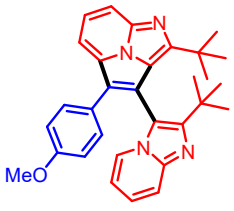
S-11, no



S-12, no



S-13, no



S-14, no

Reaction conditions: undivided cell, **1** (0.2 mmol), **2** (0.2 mmol), $^{n}Bu_4NBF_4$ (0.2 mmol, 65.8 mg), ACN/EtOH/H_2O (8+1.2+1) mL, 8 mA, 55 ℃, 1.5 h, under the open air, C cloth (35 × 15 mm), Pt (1.0 × 1.0 cm), isolated yield.

Due to the steric hindrance, only cyclization intermediates were produced when OMe replacing the *ortho* and *meta*-positions (**S-3, S-4**). When R_3 was replaced by 4-OBn, dimerization by-product was generated without any desired product detected (**S-6**). The reactions with both electron-donating groups (Me, OCH_3) and electron-withdrawing group (F) on the *inter*-position of the benzene ring of 6-phenylimidazo[2,1-*b*]thiazoles afforded the desired products, however, they were difficult to separate and purify the product due to the positional isomerization (**S-7, S-8, S-9**).

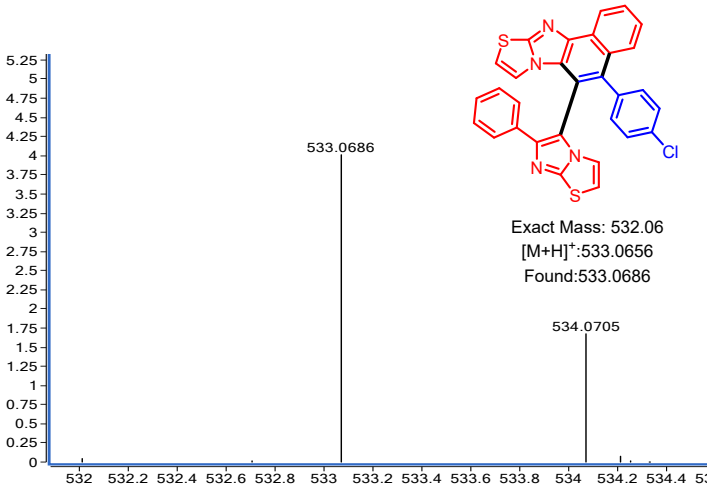
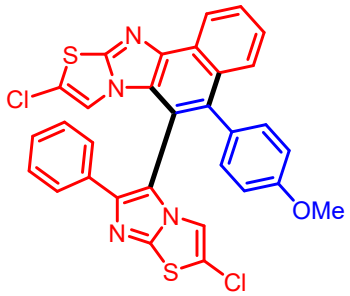


Figure **S2**: HRMS (ESI) analysis of **S-1**



S-5

9-chloro-6-(2-chloro-6-phenylimidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**S-5**):

Yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether: 10:20:30, 12.0 mg, 10 %;

^{1}H NMR (400 MHz, Chloroform-d) δ 8.78 (d, J = 8.2 Hz, 1H), 7.84 (d, J = 8.5 Hz, 1H), 7.77 (t, J = 7.6 Hz, 1H), 7.62 - 7.51 (m, 3H), 7.28 - 7.21 (m, 1H), 7.21 - 7.06 (m, 3H), 6.91 (ddd, J = 16.3, 8.5, 2.5 Hz, 2H), 6.79 (d, J = 10.6 Hz, 2H), 6.63 (dd, J = 8.5, 2.8 Hz, 1H), 3.77 (s, 3H); HRMS (ESI-TOF) Calcd for $C_{31}H_{19}Cl_2N_4OS_2$ $[M+H]^+$: 597.0372; found: 597.0364.

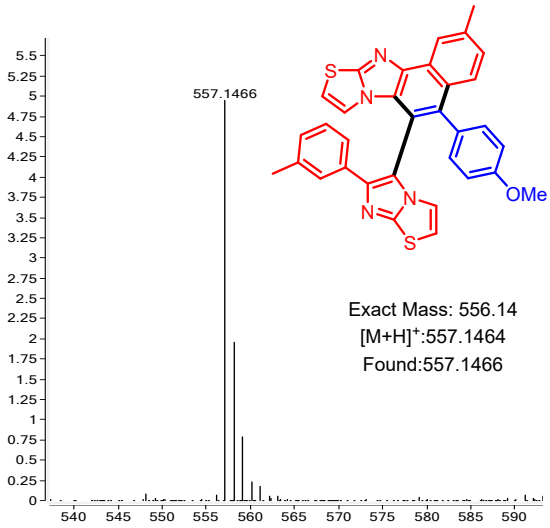


Figure **S3**: HRMS (ESI) analysis of **S-7**

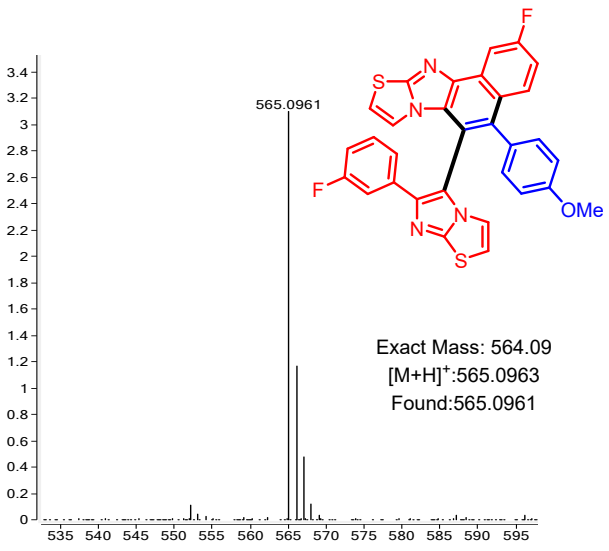


Figure **S4**: HRMS (ESI) analysis of **S-8**

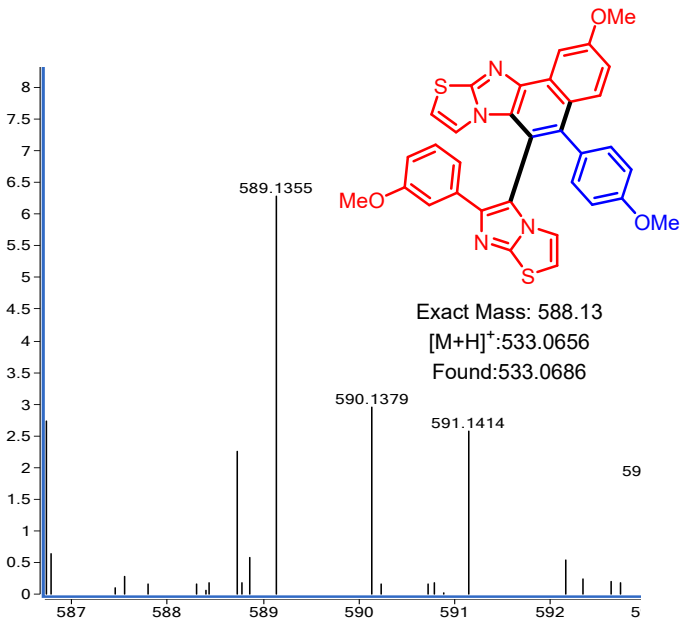
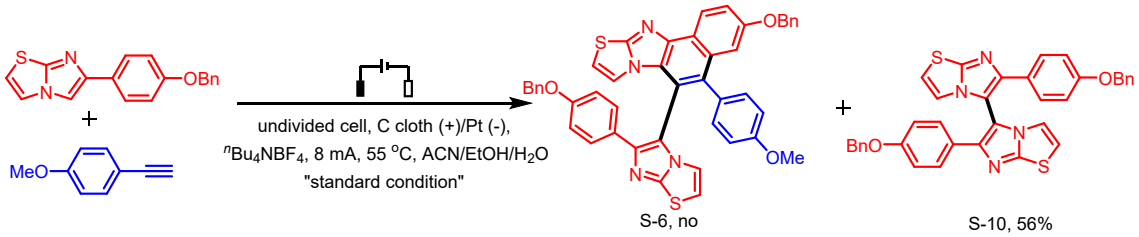
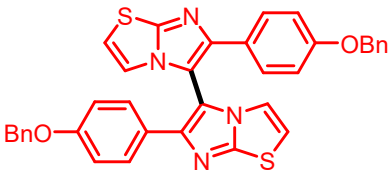


Figure **S5**: HRMS (ESI) analysis of **S-9**



In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 6-(4-(benzyloxy)phenyl)imidazo[2,1-*b*]thiazole (0.2 mmol, 61.3 mg), 1-ethynyl-4-methoxybenzene (0.2 mmol, 26.4 mg), $^{n}Bu_4NBF_4$ (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of ACN/EtOH/H_2O (8+1.2+1) mL. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 1.5 h under the open air. The reaction solution was diluted with ethyl acetate (50 mL) and washed with H_2O (50 mL). The separated organic layer was dried over anhydrous Na_2SO_4 and filtered. The filtrate was concentrated under reduced pressure to give the

crude product, which was purified by column chromatographic separation to obtain the **S-10** (ethyl acetate/petroleum ether: 1:15, 34.20 mg, 56 %).



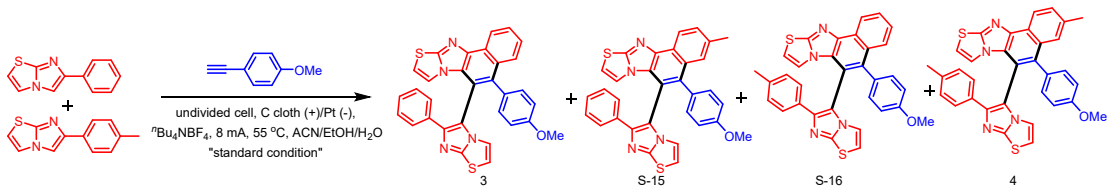
S-10

6,6'-bis(4-(benzyloxy)phenyl)-5,5'-biimidazo[2,1-*b*]thiazole (**S-10**):

Yellow solid; Eluent: ethyl acetate/petroleum ether: 1:15; 34.20 mg; 56 %;

^{1}H NMR (400 MHz, DMSO-*d6*) δ 7.90 - 7.84 (m, 6H), 7.49 - 7.44 (m, 4H), 7.44 - 7.37 (m, 6H), 7.37 - 7.30 (m, 2H), 7.16 - 7.08 (m, 4H), 5.15 (s, 4H).

6-2、 Different substituted 6-phenylimidazo[2,1-*b*]thiazoles were added at the same time



In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 6-phenylimidazo[2,1-*b*]thiazole (0.2 mmol, 40.0 mg), 6-(*p*-tolyl)imidazo[2,1-*b*]thiazole (0.2 mmol, 42.9 mg), 1-ethynyl-4-methoxybenzene **2** (0.4 mmol, 52.8 mg), ^{n}Bu$_4$NBF$_4$ (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of ACN/EtOH/H_2O (8+1.2+1) mL. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 1.5 h under the open air. The reaction solution was detected by HPLC-MS.

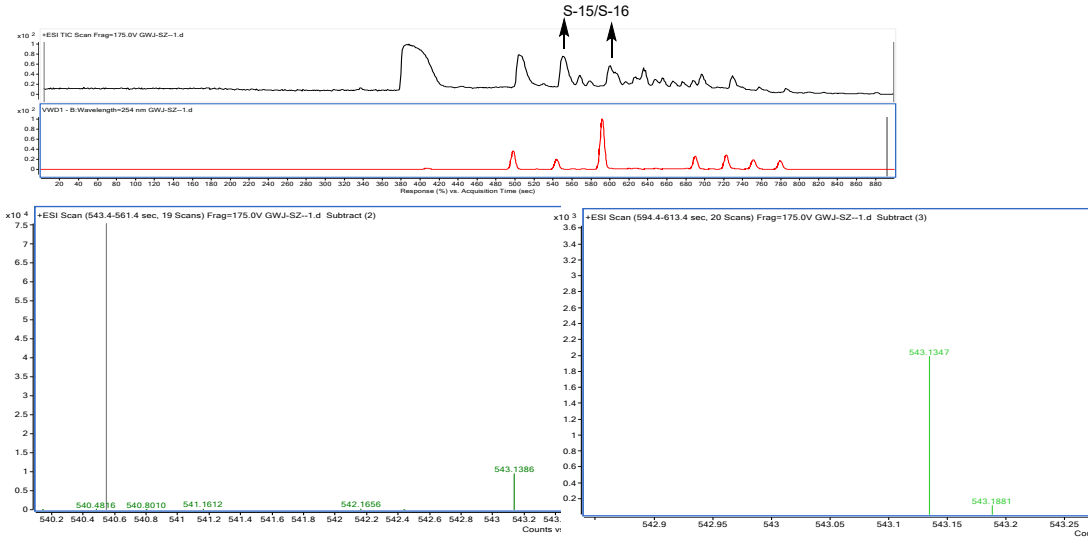
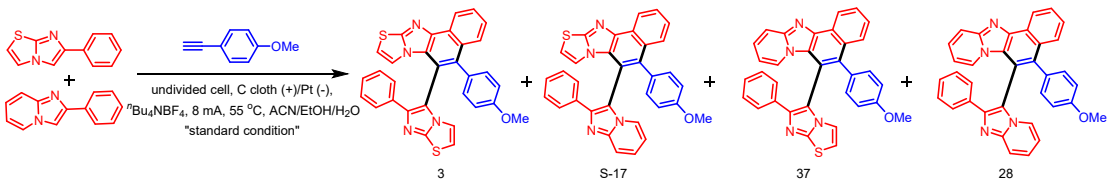


Figure **S6**. HPLC-MS of different substituted 6-phenylimidazo[2,1-*b*]thiazoles

6-3、6-Phenylimidazo[2,1-*b*]thiazole and 2-phenylimidazo[1,2-*a*]pyridine were added at the same time



In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 6-phenylimidazo[2,1-*b*]thiazole (0.2 mmol, 40.0 mg), 2-phenylimidazo[1,2-*a*]pyridine (0.2 mmol, 38.8 mg), 1-ethynyl-4-methoxybenzene **2** (0.4 mmol, 52.8 mg), $^{n}Bu_4NBF_4$ (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of ACN/EtOH/H_2O (8+1.2+1) mL. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 1.5 h under the open air. The reaction solution was detected by HPLC-MS. **37** was detected by NMR.

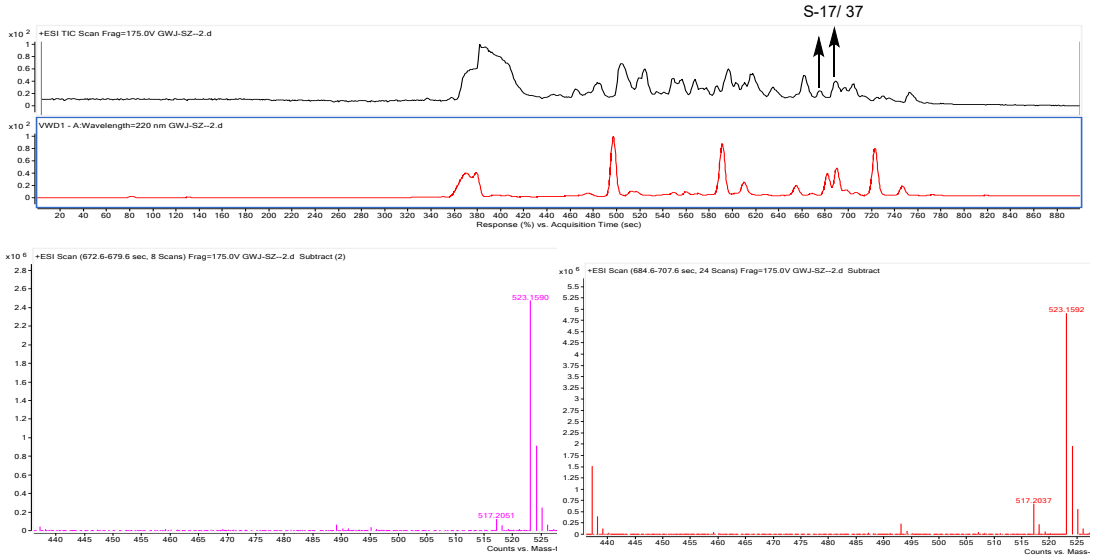
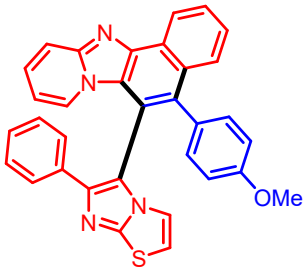


Figure **S7**. HPLC-MS of 6-phenylimidazo[2,1-*b*]thiazole and 2-phenylimidazo[1,2-*a*]pyridine



37

5-(5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridin-6-yl)-6-phenylimidazo[2,1-*b*]thiazole (**37**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; <10%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.03 (d, *J* = 6.9 Hz, 1H), 7.85 (d, *J* = 9.2 Hz, 1H), 7.79 (dd, *J* = 16.7, 8.5 Hz, 2H), 7.66 (d, *J* = 8.3 Hz, 1H), 7.60 - 7.50 (m, 3H), 7.34 - 7.28 (m, 1H), 7.14 - 7.08 (m, 3H), 7.06 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.86 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.83 (d, *J* = 4.5 Hz, 1H), 6.74 (dd, *J* = 8.4, 2.8 Hz, 1H), 6.65 (d, *J* = 4.5 Hz, 1H), 6.60 - 6.53 (m, 2H), 3.72 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.86, 149.32, 148.28, 145.55, 141.64, 136.95, 133.99, 131.86, 131.24, 130.79, 129.65, 128.67, 128.15, 127.95, 127.58, 127.50, 127.48, 126.61, 125.92, 125.66,

123.25, 122.59, 117.96, 117.24, 116.62, 114.20, 113.50, 113.22, 112.90, 112.12, 55.24; HRMS (ESI-TOF) Calcd for $C_{33}H_{23}N_4OS$ $[M+H]^+$: 523.1587; found: 523.1594.

6-4、HPLC spectrum of 4

In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 6-(p-tolyl)imidazo[2,1-b]thiazole (0.2 mmol), 1-ethynyl-4-methoxybenzene (0.2 mmol), $^nBu_4NBF_4$ (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of ACN/EtOH/H_2O (8+1.2+1) mL. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 1.5 h under the open air. The reaction solution was diluted with ethyl acetate (50 mL) and washed with H_2O (50 mL). The separated organic layer was dried over anhydrous Na_2SO_4 and filtered. The filtrate was concentrated under reduced pressure to give the crude product, which was purified by column chromatographic separation (dichloromethane/ethyl acetate/petroleum ether: 10:20:30) to obtain the desired product **4**.

HPLC analysis: Daicel Chiralpak AS-H, hexane/iso-propanol = 90:10, flow rate = 0.3 mL/min, λ = 245nm.

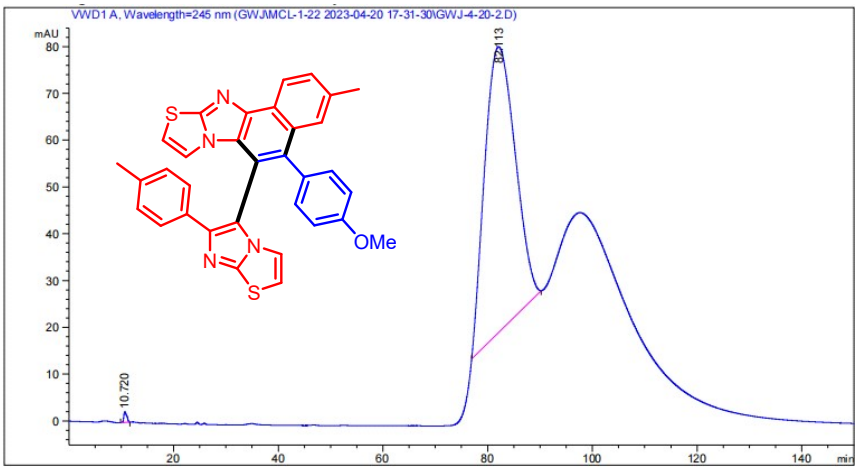
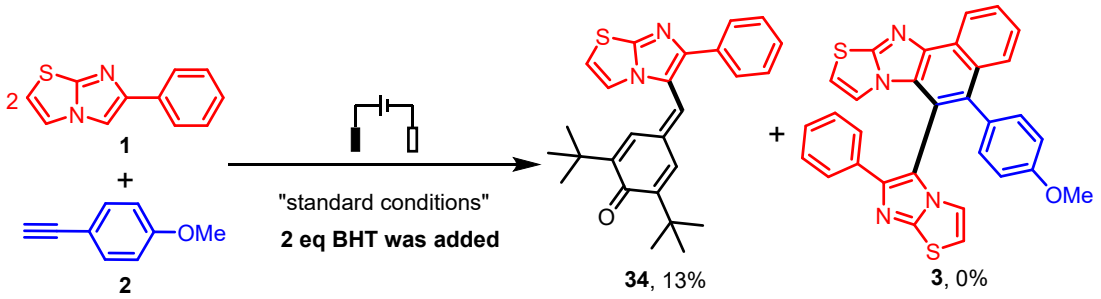


Figure **S8**. HPLC spectrum of **4**

7、Mechanistic Studies

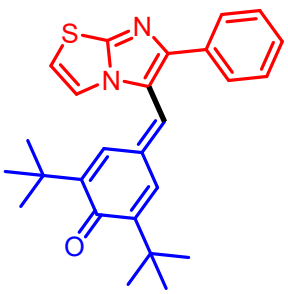
7-1、Radical-trapping experiments

7-1-1、 BHT was added



In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 6-phenylimidazo[2,1-*b*]thiazole **1** (0.2 mmol, 40.0 mg), 1-ethynyl-4-methoxybenzene **2** (0.2 mmol, 26.4 mg), $^{n}Bu_4NBF_4$ (0.2 mmol, 65.8 mg) and BHT (0.4 mmol, 88.2 mg) were dissolved in a mixed solvent of ACN/EtOH/H_2O (8+1.2+1) mL. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 3 h under the open air. No desired product was detected. Moreover, the corresponding radical trapping product **34** was generated.

The reaction solution was diluted with ethyl acetate (50 mL) and washed with brine (50 mL) and H_2O (50 mL). The separated organic layer was dried over anhydrous Na_2SO_4 and filtered. The filtrate was concentrated under reduced pressure to give the crude product, which was purified by column chromatographic separation (dichloromethane/ethyl acetate/petroleum ether: 10:2:30) to obtain the radical trapping product **34** (10.8 mg, 13%).



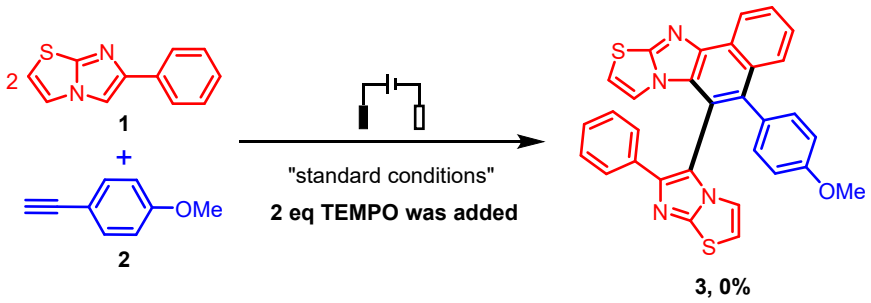
34

2,6-di-*tert*-butyl-4-((6-phenylimidazo[2,1-*b*]thiazol-5-yl)methylene)cyclohexa-2,5-dien-1-one (**34**):

Orange solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:2:30; 10.8 mg, 13%;

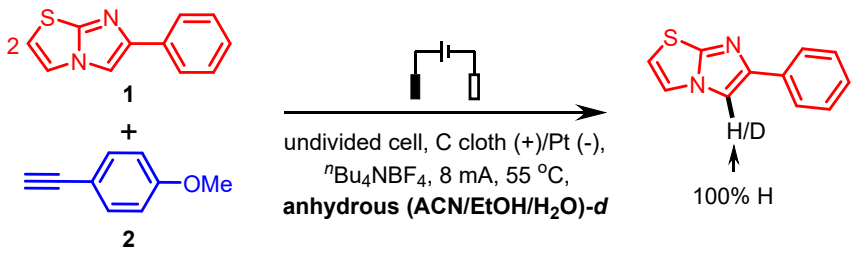
^{1}H NMR (400 MHz, DMSO-*d6*) δ 8.09 (d, *J* = 4.5 Hz, 1H), 7.68 - 7.61 (m, 3H), 7.47 (d, *J* = 4.4 Hz, 1H), 7.39 (t, *J* = 7.4 Hz, 2H), 7.31 (t, *J* = 7.3 Hz, 1H), 7.26 (d, *J* = 2.4 Hz, 1H), 6.92 (d, *J* = 2.3 Hz, 1H), 1.28 (s, 9H), 0.87 (s, 9H); HRMS (ESI-TOF) Calcd for $C_{26}H_{29}N_2OS$ $[M+H]^+$: 417.1995; found: 417.1991.

7-1-2、 TEMPO was added



In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 6-phenylimidazo[2,1-*b*]thiazole **1** (0.2 mmol, 40.0 mg), 1-ethynyl-4-methoxybenzene **2** (0.2 mmol, 26.4 mg), ^{n}Bu$_4$NBF$_4$ (0.2 mmol, 65.8 mg) and TEMPO (0.4 mmol, 62.5 mg) were dissolved in a mixed solvent of ACN/EtOH/H_2O (8+1.2+1) mL The mixture above was stirred and electrolyzed at a constant current of 8 mA for 1.5 h under the open air. No desired product was detected.

7-2、Isotope labeling experiments:



In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 6-phenylimidazo[2,1-*b*]thiazole **1** (0.2 mmol,

40.0 mg), 1-ethynyl-4-methoxybenzene **2** (0.2 mmol, 26.4 mg), $^{n}Bu_4NBF_4$ (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of (ACN/EtOH/H_2O)-*d6* (8+1.2+1) mL. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 40 min under the N_2 atmosphere. No H/D change was detected.

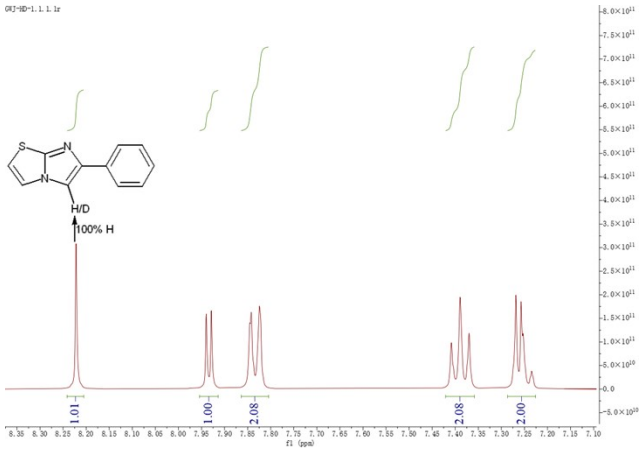
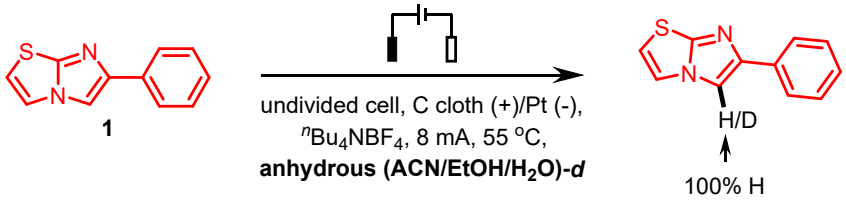


Figure S9 ^{1}H NMR data of H/D exchange experiments of substrates **1** and **2** under mixed solvents of (ACN/EtOH/H_2O)-*d6*



In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 6-phenylimidazo[2,1-*b*]thiazole **1** (0.2 mmol, 40.0 mg), $^{n}Bu_4NBF_4$ (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of (ACN/EtOH/H_2O)-*d6* (8+1.2+1) mL. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 40 min under the N_2 atmosphere. No H/D change was detected.

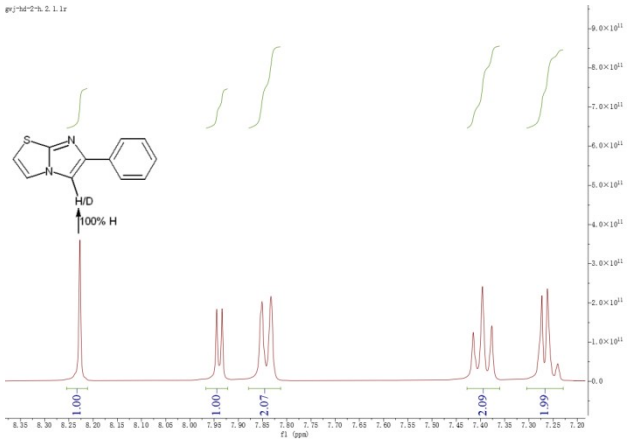
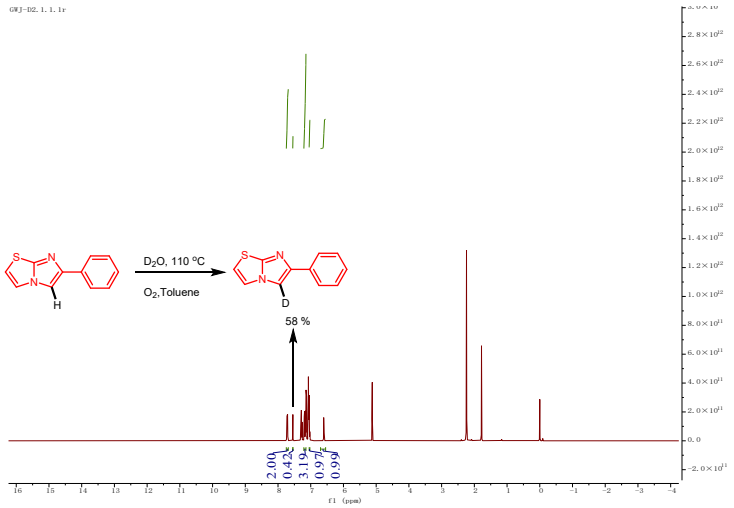
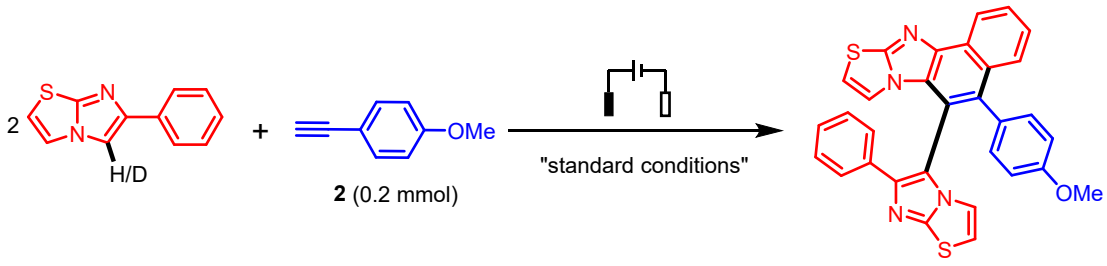


Figure S10 ^{1}H NMR data of H/D exchange experiments of substrate **1** under mixed solvents of (ACN/EtOH/H_2O)-*d6*

7-3、KIE experiments:



Under air, a sealed reaction tube was charged with **1** (0.2 mmol), D_2O (25.0 equiv.) and 3 mL of toluene. The reaction mixture was stirred at 110 °C for 12 h. After the completion of the reaction, the solvent was evaporated under reduced pressure and the residue was purified by flash column chromatography on a silica gel to give the product. The mixture was analyzed using ^{1}H NMR spectrometer.



In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 6-phenylimidazo[2,1-*b*]thiazole (0.2 mmol, 40.0 mg), 1-ethynyl-4-methoxybenzene **2** (0.2 mmol, 26.4 mg), ^{n}Bu_4NBF_4 (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of ACN/EtOH/H_2O (8+1.2+1) mL, 0.2 ml of

the reaction solution was taken every 15 minutes, and the yields were determined by ^{1}H NMR using dibromomethane as the internal standard. Taking time as abscissa and yield as ordinate, the image was drawn and K_H was obtained.

In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 6-phenylimidazo[2,1-*b*]thiazole-5-*d* (0.2 mmol), 1-ethynyl-4-methoxybenzene 2 (0.2 mmol, 26.4 mg), ^{n}Bu$_4$NBF$_4$ (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of ACN/EtOH/H_2O (8+1.2+1) mL. 0.2 ml of the reaction solution was taken every 15 minutes, and the yields were determined by ^{1}H NMR using dibromomethane as the internal standard. Taking time as abscissa and yield as ordinate, the image was drawn and K_D was obtained.

Time / min	15	30	45	60	90
Yield / % (H-substrate)	6	18	34	46	84
Yield / % (D-substrate)	3	12	22	24	40

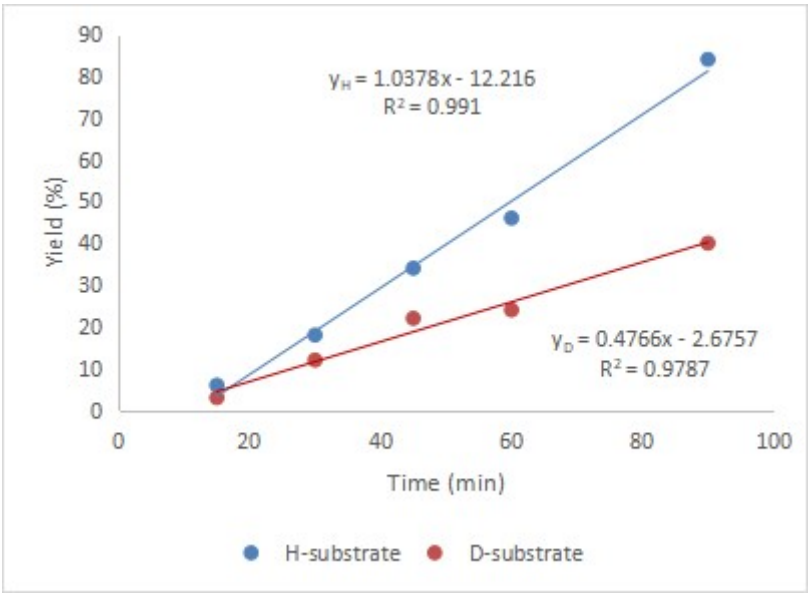
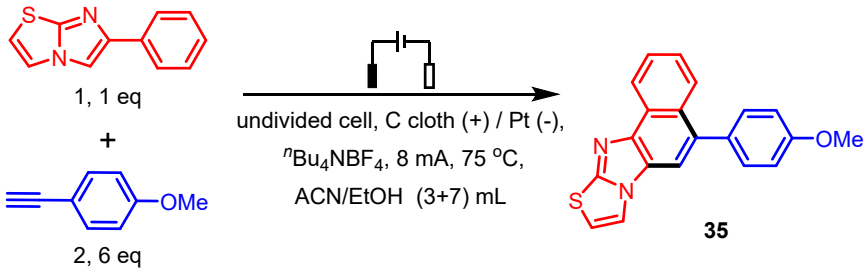


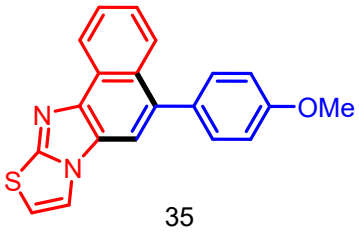
Figure S11. Kinetic isotope effect

7-4、Verification experiments of intermediates

7-4-1、Synthesis of substrates 35 and 36



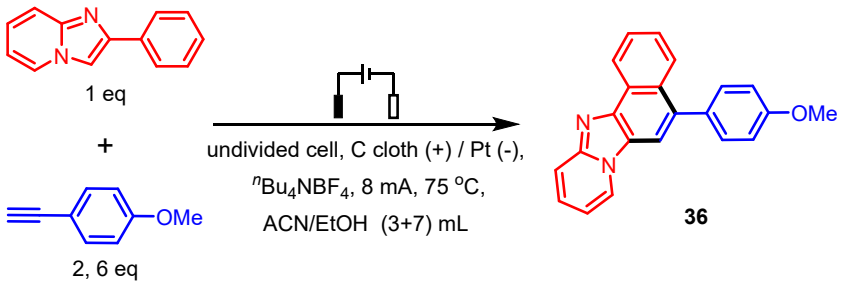
In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 6-phenylimidazo[2,1-*b*]thiazole (0.2 mmol, 40.0 mg), 1-ethynyl-4-methoxybenzene (1.2 mmol, 158.6 mg), $^{n}Bu_4NBF_4$ (0.2 mmol, 65.8 mg) were dissolved in 3 mL ACN and 7 mL EtOH. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 2 h under the open air. The reaction solution was diluted with ethyl acetate (50 mL) and washed with H_2O (50 mL). The separated organic layer was dried over anhydrous Na_2SO_4 and filtered. The filtrate was concentrated under reduced pressure to give the crude product, which was purified by column chromatographic separation (petroleum ether/ethyl acetate: 15:1) to obtain the desired product **35**.



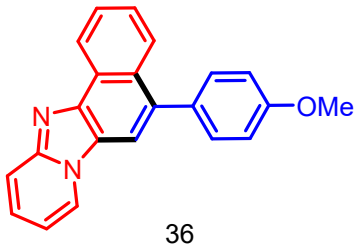
5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**35**):[1]

Pale yellow solid; Eluent: petroleum ether/ethyl acetate 15:1; 59.5 mg, 90%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.75 (d, *J* = 9.6 Hz, 1H), 7.97 (d, *J* = 8.4 Hz, 1H), 7.69 - 7.62 (m, 3H), 7.51 - 7.40 (m, 3H), 7.07 - 7.00 (m, 2H), 6.85 (d, *J* = 4.5 Hz, 1H), 3.90 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 159.07, 154.24, 143.39, 134.39, 133.38, 131.50, 129.65, 127.04, 126.85, 126.51, 125.16, 122.52, 117.44, 113.84, 111.74, 111.43, 55.48; HRMS (ESI-TOF) Calcd for $C_{20}H_{15}N_2OS$ $[M+H]^+$: 331.09; found: 331.0896.



In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 2-phenylimidazo[1,2-*a*]pyridine (0.2 mmol, 38.9 mg), 1-ethynyl-4-methoxybenzene **2** (1.2 mmol, 158.6 mg), ^{n}Bu$_4$NBF$_4$ (0.2 mmol, 65.8 mg) were dissolved in 3 mL ACN and 7 mL EtOH. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 1.5 h under the open air. The reaction solution was diluted with ethyl acetate (50 mL) and washed with H_2O (50 mL). The separated organic layer was dried over anhydrous Na_2SO_4 and filtered. The filtrate was concentrated under reduced pressure to give the crude product, which was purified by column chromatographic separation (petroleum ether/ethyl acetate: 20:1) to obtain the desired product **36**.



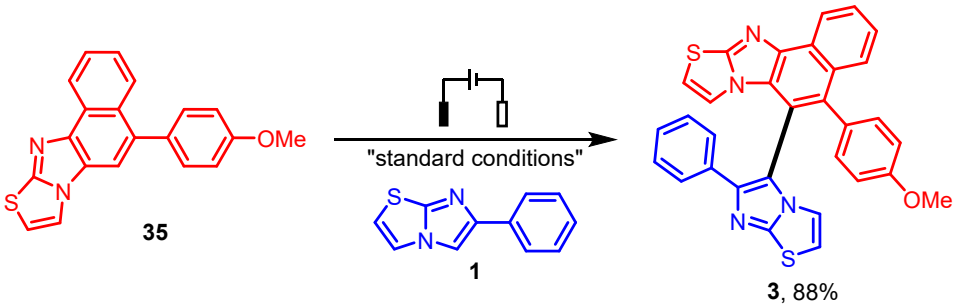
36

5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**36**):[1]

Pale yellow solid; Eluent: petroleum ether/ethyl acetate 20:1; 20.8 mg, 8%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.94 (d, *J* = 8.1 Hz, 1H), 8.51 (d, *J* = 6.8 Hz, 1H), 8.02 (d, *J* = 8.5 Hz, 1H), 7.93 (d, *J* = 9.2 Hz, 1H), 7.84 (s, 1H), 7.76 - 7.70 (m, 1H), 7.56 (t, *J* = 7.7 Hz, 1H), 7.48 (dd, *J* = 9.0, 2.8 Hz, 3H), 7.16 -7.03 (m, 2H), 6.98 (t, *J* = 6.7 Hz, 1H), 3.92 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 159.20, 146.86, 135.35, 133.30, 132.16, 131.53, 131.17, 130.11, 128.34, 127.97, 126.54, 126.32, 124.59, 123.86, 123.50, 117.75, 113.93, 111.95, 110.87, 55.54; HRMS (ESI-TOF) Calcd for $C_{22}H_{17}N_2O$ $[M+H]^+$: 325.1335; found: 325.1334.

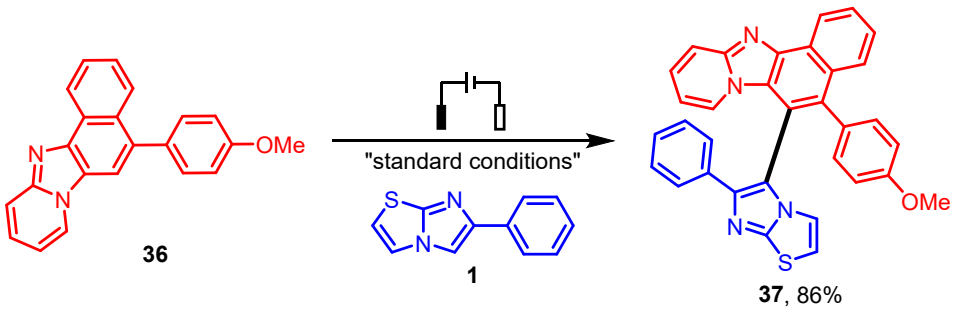
7-4-2、Verification experiments of intermediate 35



35 1 **3**, 88%

In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole **35** (0.2 mmol, 66.1 mg), 6-phenylimidazo[2,1-*b*]thiazole **1** (0.2 mmol, 40.0 mg), $^{n}Bu_4NBF_4$ (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of ACN/EtOH/H_2O (8+1.2+1) mL. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 1.5 h under the open air. The reaction solution was diluted with ethyl acetate (50 mL) and washed with H_2O (50 mL). The separated organic layer was dried over anhydrous Na_2SO_4 and filtered. The filtrate was concentrated under reduced pressure to give the crude product, which was purified by column chromatographic separation (dichloromethane/ethyl acetate/petroleum ether: 10:20:30) to obtain the desired product **3** (93.1 mg, 88%).

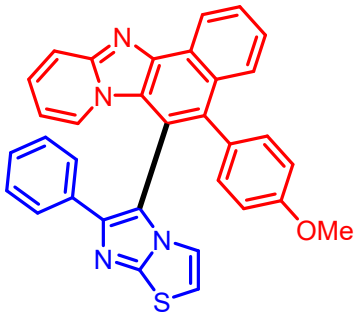
7-4-3、Verification experiments of intermediate 36



36 1 **37**, 86%

In an undivided cell equipped with a carbon cloth (35 mm x 15 mm) anode and a Pt (10 mm x 10 mm x 0.1 mm) cathode, 5-(4-

methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine **36** (0.2 mmol, 64.9 mg), 6-phenylimidazo[2,1-*b*]thiazole **1** (0.2 mmol, 40.0 mg), $^{n}Bu_4NBF_4$ (0.2 mmol, 65.8 mg) were dissolved in a mixed solvent of ACN/EtOH/H_2O (8+1.2+1) mL. The mixture above was stirred and electrolyzed at a constant current of 8 mA for 1.5 h under the open air. The reaction solution was diluted with ethyl acetate (50 mL) and washed with H_2O (50 mL). The separated organic layer was dried over anhydrous Na_2SO_4 and filtered. The filtrate was concentrated under reduced pressure to give the crude product, which was purified by column chromatographic separation (dichloromethane/ethyl acetate/petroleum ether: 10:20:30) to obtain the desired product **37** (89.9 mg, 86%).



37

5-(5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridin-6-yl)-6-phenylimidazo[2,1-*b*]thiazole (**37**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 89.9 mg, 86%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.03 (d, *J* = 6.9 Hz, 1H), 7.85 (d, *J* = 9.2 Hz, 1H), 7.79 (dd, *J* = 16.7, 8.5 Hz, 2H), 7.66 (d, *J* = 8.3 Hz, 1H), 7.60 - 7.50 (m, 3H), 7.34 - 7.28 (m, 1H), 7.14 - 7.08 (m, 3H), 7.06 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.86 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.83 (d, *J* = 4.5 Hz, 1H), 6.74 (dd, *J* = 8.4, 2.8 Hz, 1H), 6.65 (d, *J* = 4.5 Hz, 1H), 6.60 - 6.53 (m, 2H), 3.72 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.86, 149.32, 148.28, 145.55, 141.64, 136.95, 133.99, 131.86, 131.24, 130.79, 129.65, 128.67, 128.15, 127.95, 127.58, 127.50, 127.48, 126.61, 125.92, 125.66, 123.25, 122.59, 117.96, 117.24, 116.62, 114.20, 113.50, 113.22, 112.90, 112.12, 55.24; HRMS (ESI-TOF) Calcd for $C_{33}H_{23}N_4OS$ $[M+H]^+$: 523.1587; found: 523.1594.

7-5、Cyclic voltammetry experiments

The undivided cell was equipped with glassy-carbon disk working electrode (diameter, 3.0 mm) and Pt wire auxiliary electrode. The Ag/AgCl was used as reference electrode. The scan range was 0.0 V to 3 V. The scan rate was 100 mVs^{-1}. ACN (8 mL) and EtOH (1.2 mL) and H_2O (1 mL) containing $^{n}Bu_4NBF_4$ (0.2 mmol, 65.8 mg) was poured into the electrochemical cell in all experiments.

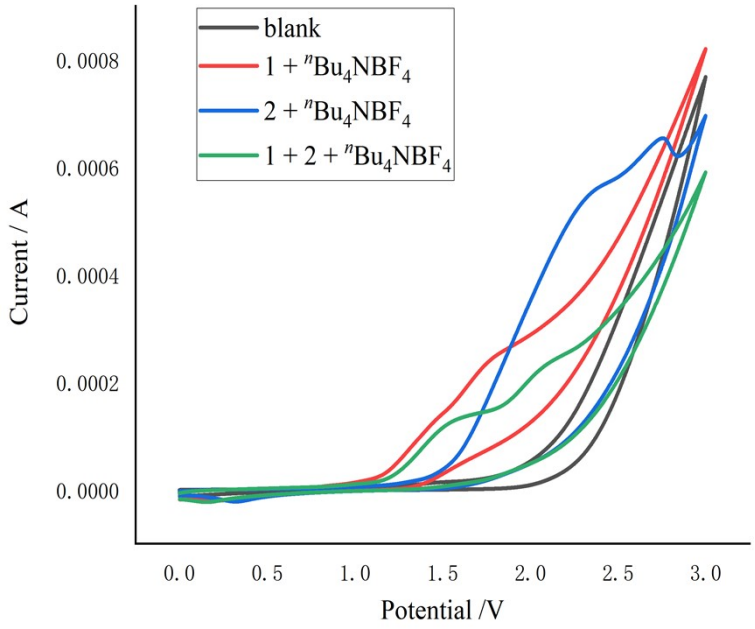


Figure S12 Cyclic voltammetry experiments of substrates

100 mVs-1: (black) blank; (red) 6-phenylimidazo[2,1-*b*]thiazole **1** (0.2 mmol, 40.0 mg); (blue) 1-ethynyl-4-methoxybenzene **2** (0.2 mmol, 26.4 mg); (green) 6-phenylimidazo[2,1-*b*]thiazole **1** (0.2 mmol, 40.0 mg) and 1-ethynyl-4-methoxybenzene **2** (0.2 mmol, 26.4 mg).

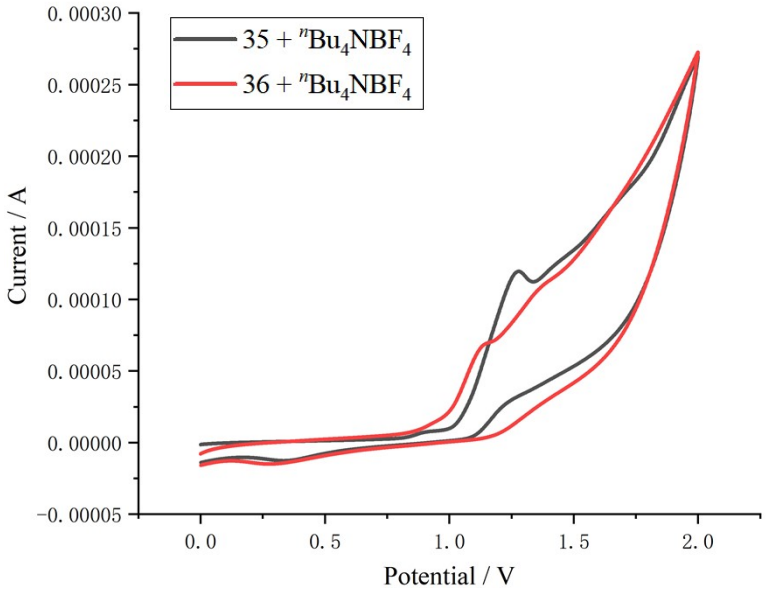
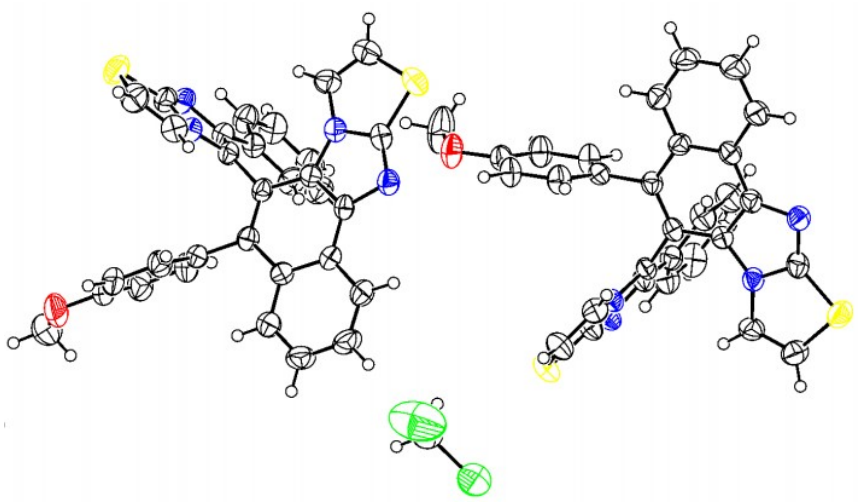


Figure S13 Cyclic voltammetry experiments of intermediates **35** and **36**

100 mVs-1: (black) 5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole **35** (0.2 mmol, 66.1 mg); (red) 5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine **36** (0.2 mmol, 64.9 mg).

8、X-ray Crystallography Studies of Product 3



Single crystal suitable for X-ray diffraction was obtained by slow evaporation of a saturated solution of compound **3** (cyclohexane/CH_2Cl_2) in a loosely capped vial.

Figure S14 Structure of **3** by X-Ray crystallographic (CCDC = 2182532)

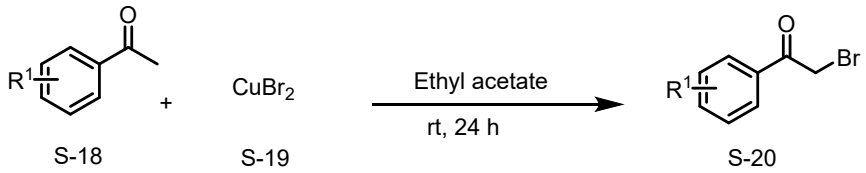
Table S3 Crystal data and structure refinement for **3**

Empirical formula	$C_{63}H_{42}Cl_2N_8O_2S_4$
Formula weight	1142.18

Temperature/K	296.15
Crystal system	triclinic
Space group	P-1
a/Å	12.1415(7)
b/Å	12.5166(6)
c/Å	19.4380(12)
α/°	96.476(2)
β/°	91.801(2)
γ/°	112.426(2)
Volume/Å^3	2704.2(3)
Z	2
ρcalcg/cm^3	1.403
μ/mm^{-1}	0.330
F(000)	1180.0
Crystal size/mm^3	0.15 × 0.12 × 0.1
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection/°	3.552 to 54.98
Index ranges	$-15 \leq h \leq 12$, $-16 \leq k \leq 16$, $-25 \leq l \leq 24$
Reflections collected	25923
Independent reflections	12341 [Rint = 0.0481, Rsigma = 0.0879]
Data/restraints/parameters	12341/0/714
Largest diff. peak/hole / e Å^{-3}	0.63/-0.69

9、Synthesis of Substrates

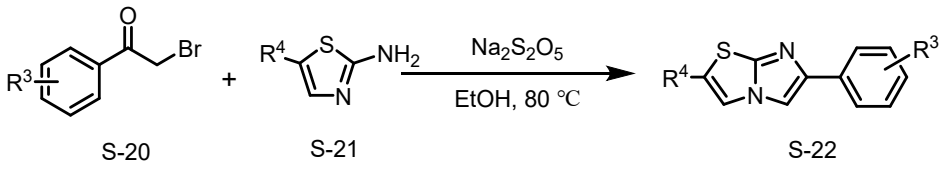
Step 1: General procedure for the synthesis of 2-bromo-1-phenylethan-1-one (**S-20**):[2]



S-18 S-19 S-20

To the solution of substituted acetophenone **S-18** (5 mmol) in ethyl acetate (30 mL), copper(II)bromide (6 mmol) was added. The reaction mixture was allowed to stir at room temperature for about 24 hours. After **S-18** was consumed, the reaction solution was diluted with ethyl acetate (150 mL) and washed with saturated NH_4Cl aqueous solution (150 mL) and H_2O (150 mL). The separated organic layer was dried with anhydrous Na_2SO_4 and filtered. The filtrate was concentrated under reduced pressure to get the desired product **S-20** in 95-99% yield.

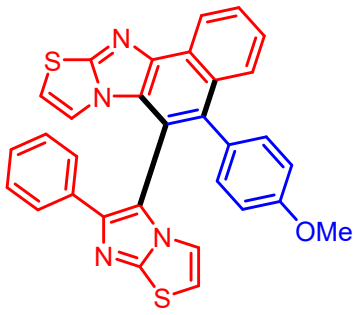
Step 2: General procedure for the synthesis of substrates **S-22**:[3]



S-20 S-21 S-22

Substituted 2-bromo-1-phenylethan-1-ones (5 mmol, 1 eq), 2-aminothiazoles (5 mmol, 1 eq), $Na_2S_2O_5$ (5 mmol, 1 eq, 950 mg) and EtOH (30 mL) was placed in an oven-dried round bottom flask equipped with a magnetic stir bar. The reaction mixture was stirred at 80 °C for 24 hours. After **S-20** was consumed, the reaction solution was diluted with ethyl acetate (150 mL) and washed with brine (150 mL) and H_2O (150 mL). The separated organic layer was dried with anhydrous Na_2SO_4 and filtered. The filtrate was concentrated under reduced pressure to obtain a crude product, which was separated by column chromatography (eluent: petroleum ether = 3 : 1) to give the desired product **S-22**.

10、Characterization Data for Electrolysis Products

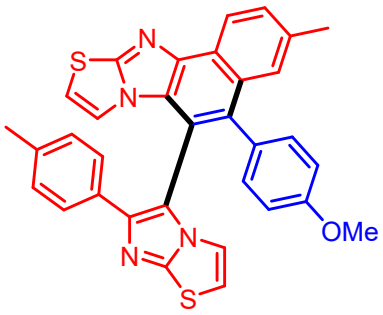


3

5-(4-methoxyphenyl)-6-(6-phenylimidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**3**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 44.4 mg, 84%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.84 (d, *J* = 8.2 Hz, 1H), 7.84 (d, *J* = 8.5 Hz, 1H), 7.76 (t, *J* = 7.6 Hz, 1H), 7.62 - 7.56 (m, 2H), 7.52 (t, *J* = 7.0 Hz, 1H), 7.20 (dd, *J* = 8.4, 2.2 Hz, 1H), 7.14 (t, *J* = 2.8 Hz, 3H), 6.96 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.84 - 6.76 (m, 3H), 6.65 - 6.55 (m, 3H), 3.73 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.92, 155.47, 149.34, 145.18, 144.12, 135.45, 133.80, 131.35, 131.32, 130.66, 129.64, 128.80, 127.96, 127.61, 127.55, 127.48, 125.84, 125.79, 124.63, 122.53, 118.43, 117.44, 116.59, 113.70, 113.62, 113.47, 112.65, 111.97, 55.27; HRMS (ESI-TOF) Calcd for $C_{31}H_{21}N_4OS_2$ $[M+H]^+$: 529.1151; found: 529.1151.



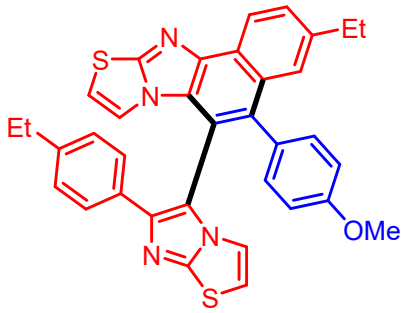
4

5-(4-methoxyphenyl)-3-methyl-6-(6-(*p*-tolyl)imidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**4**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:15:30;

47.3 mg, 85%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.73 (d, *J* = 7.3 Hz, 1H), 7.61 (d, *J* = 9.1 Hz, 2H), 7.49 (d, *J* = 8.2 Hz, 2H), 7.21 (dd, *J* = 8.4, 2.3 Hz, 1H), 7.01 - 6.91 (m, 3H), 6.82 (dd, *J* = 8.4, 2.8 Hz, 1H), 6.80 - 6.74 (m, 2H), 6.65 - 6.58 (m, 2H), 6.56 (d, *J* = 4.6 Hz, 1H), 3.75 (s, 3H), 2.49 (s, 3H), 2.23 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.84, 155.37, 149.20, 145.34, 144.21, 137.33, 135.51, 134.87, 131.41, 131.35, 131.11, 130.87, 129.86, 129.56, 129.52, 127.01, 125.70, 125.52, 124.32, 122.45, 118.55, 117.46, 116.24, 113.82, 113.68, 113.46, 112.27, 111.68, 55.27, 22.16, 21.28; HRMS (ESI-TOF) Calcd for $C_{33}H_{25}N_4OS_2$ $[M+H]^+$: 557.1464; found: 557.1467.

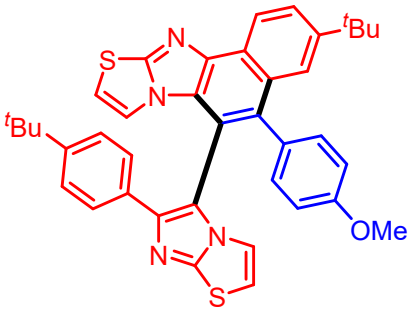


5

3-ethyl-6-(6-(4-ethylphenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**5**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:15:30; 51.4 mg, 88%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.76 (d, *J* = 8.4 Hz, 1H), 7.69 - 7.60 (m, 2H), 7.51 (d, *J* = 8.3 Hz, 2H), 7.21 (dd, *J* = 8.4, 2.3 Hz, 1H), 7.02 - 6.92 (m, 3H), 6.82 (dd, *J* = 8.4, 2.8 Hz, 1H), 6.78 (d, *J* = 4.5 Hz, 2H), 6.67 - 6.51 (m, 3H), 3.75 (s, 3H), 2.78 (q, *J* = 7.5 Hz, 2H), 2.53 (q, *J* = 7.6 Hz, 2H), 1.27 (t, *J* = 7.6 Hz, 3H), 1.13 (d, *J* = 7.6 Hz, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.83, 155.35, 149.16, 145.32, 144.18, 143.68, 141.87, 135.06, 131.40, 131.24, 130.88, 129.85, 128.42, 128.30, 125.96, 125.80, 125.77, 124.37, 122.60, 118.57, 117.46, 116.30, 113.74, 113.65, 113.45, 112.32, 111.68, 55.27, 29.49, 28.59, 15.96, 15.37; HRMS (ESI-TOF) Calcd for $C_{35}H_{29}N_4OS_2$ $[M+H]^+$: 585.1777; found: 585.1779.

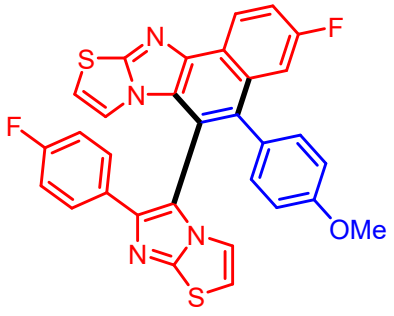


6

3-(*tert*-butyl)-6-(6-(4-(*tert*-butyl)phenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**6**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 41.6 mg, 65%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.77 (d, *J* = 8.6 Hz, 1H), 7.90 - 7.81 (m, 2H), 7.57 - 7.49 (m, 2H), 7.26 - 7.19 (m, 1H), 7.16 (d, *J* = 8.6 Hz, 2H), 7.00 (dd, *J* = 8.5, 2.2 Hz, 1H), 6.82 (dd, *J* = 8.4, 2.7 Hz, 1H), 6.77 (dd, *J* = 7.6, 4.4 Hz, 2H), 6.65 - 6.54 (m, 3H), 3.75 (s, 3H), 1.34 (s, 9H), 1.22 (s, 9H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.82, 155.33, 150.56, 149.13, 148.50, 145.27, 144.00, 135.51, 131.48, 131.38, 130.90, 130.51, 129.81, 126.19, 125.72, 125.55, 125.51, 124.55, 123.33, 122.30, 118.65, 117.45, 116.42, 113.60, 113.56, 113.42, 112.37, 111.67, 55.27, 35.24, 34.62, 31.44, 31.30; HRMS (ESI-TOF) Calcd for $C_{39}H_{37}N_4OS_2$ $[M+H]^+$: 641.2403; found: 641.2404.

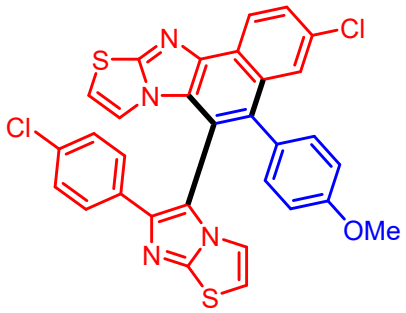


7

3-fluoro-6-(6-(4-fluorophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**7**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:22:30; 28.2 mg, 50%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.81 (dd, *J* = 9.0, 5.8 Hz, 1H), 7.56 - 7.48 (m, 3H), 7.44 (dd, *J* = 11.4, 2.5 Hz, 1H), 7.15 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.92 - 6.79 (m, 5H), 6.76 (d, J = 4.0 Hz, 1H), 6.68 (d, *J* = 3.9 Hz, 1H), 6.63 (dd, *J* = 8.5, 2.8 Hz, 2H), 3.75 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 163.63, 162.15, 161.17, 159.72, 159.15, 155.83, 149.49, 144.15 (d, *J* = 7.1 Hz), 134.76, 134.72, 131.94 (d, *J* = 9.1 Hz), 131.14 (d, *J* = 4.0 Hz), 129.73, 129.07, 127.67 (d, *J* = 8.1 Hz), 124.99 (d, *J* = 9.1 Hz), 124.17 (d, *J* = 10.1 Hz), 118.31, 117.56, 117.45, 117.31, 116.04, 115.84 (d, *J* = 21.2 Hz), 114.34, 113.86 (d, *J* = 29.3 Hz), 113.13, 112.37, 112.03, 111.80, 55.33; ^{19}F NMR (376 MHz, Chloroform-*d*) δ -113.68, -113.76; HRMS (ESI-TOF) Calcd for $C_{31}H_{19}F_2N_4OS_2$ $[M+H]^+$: 565.0963; found: 565.0962.

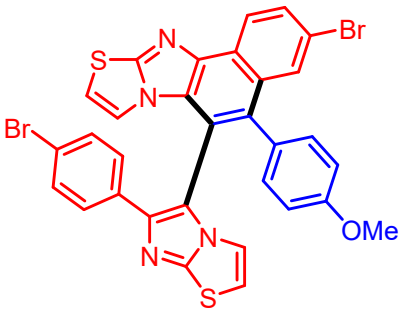


8

3-chloro-6-(6-(4-chlorophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**8**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 36.4 mg, 61%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.75 (d, *J* = 8.8 Hz, 1H), 7.78 (d, *J* = 2.0 Hz, 1H), 7.70 (dd, *J* = 8.7, 2.0 Hz, 1H), 7.47 (d, *J* = 8.6 Hz, 2H), 7.18 - 7.08 (m, 3H), 6.89 (dd, *J* = 8.5, 2.2 Hz, 1H), 6.83 (dd, *J* = 11.3, 3.2 Hz, 2H), 6.71 (dd, *J* = 17.9, 4.3 Hz, 2H), 6.63 (dd, *J* = 8.5, 2.7 Hz, 2H), 3.75 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 159.18, 156.00, 149.64, 144.14, 144.01, 134.65, 133.54, 132.17, 132.02, 131.57, 131.18, 129.04, 128.82, 128.38, 127.10, 126.86, 125.70, 124.59, 124.28, 118.20, 117.38, 116.40, 114.36, 114.03, 113.72, 113.22, 112.58, 55.33; HRMS (ESI-TOF) Calcd for $C_{31}H_{19}Cl_2N_4OS_2$ $[M+H]^+$: 597.0372; found: 597.0362.

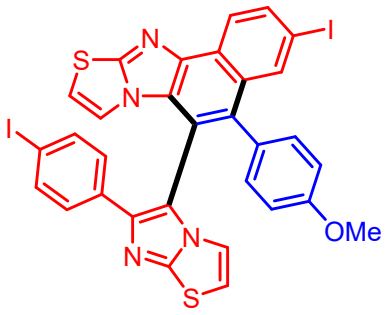


9

3-bromo-6-(6-(4-bromophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**9**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 52.1 mg, 76%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.65 (d, *J* = 8.7 Hz, 1H), 7.93 (d, *J* = 1.9 Hz, 1H), 7.80 (dd, *J* = 8.7, 1.9 Hz, 1H), 7.40 (d, *J* = 6.7 Hz, 2H), 7.25 (d, *J* = 5.2 Hz, 2H), 7.13 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.88 (dd, *J* = 8.5, 2.3 Hz, 1H), 6.84 (d, *J* = 4.5 Hz, 1H), 6.81 (dd, *J* = 8.5, 2.7 Hz, 1H), 6.69 (dd, *J* = 13.7, 4.5 Hz, 2H), 6.62 (q, *J* = 3.2 Hz, 2H), 3.74 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 159.16, 155.97, 149.64, 144.17, 144.01, 134.55, 132.64, 131.94, 131.91, 131.16, 130.87, 130.01, 128.74, 127.35, 125.90, 124.58, 124.34, 121.74, 120.21, 118.14, 117.36, 116.41, 114.33, 114.01, 113.71, 113.21, 112.60, 55.31; HRMS (ESI-TOF) Calcd for $C_{31}H_{19}Br_2N_4OS_2$ $[M+H]^+$: 684.9362; found: 684.9360.

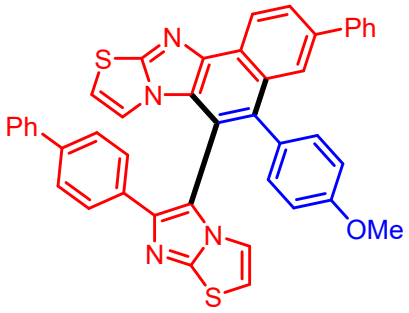


10

3-iodo-6-(6-(4-iodophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**10**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:15:30; 55.4 mg, 71%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.54 (d, *J* = 8.6 Hz, 1H), 8.16 (d, *J* = 1.7 Hz, 1H), 8.01 (dd, *J* = 8.7, 1.7 Hz, 1H), 7.50 - 7.43 (m, 2H), 7.30 - 7.26 (m, 2H), 7.13 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.89 (dd, *J* = 8.5, 2.3 Hz, 1H), 6.82 (q, *J* = 3.2 Hz, 2H), 6.69 (dd, *J* = 13.9, 4.5 Hz, 2H), 6.66 - 6.60 (m, 2H), 3.76 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 159.17, 155.99, 149.63, 144.28, 144.06, 137.91, 136.51, 136.19, 134.41, 133.24, 132.27, 131.21, 131.17, 128.72, 127.53, 126.29, 124.69, 124.28, 118.16, 117.33, 116.52, 114.14, 114.03, 113.71, 113.22, 112.64, 93.41, 91.84, 55.33; HRMS (ESI-TOF) Calcd for $C_{31}H_{19}I_2N_4OS_2$ [M+H]$^+$: 780.9084; found: 780.9075.

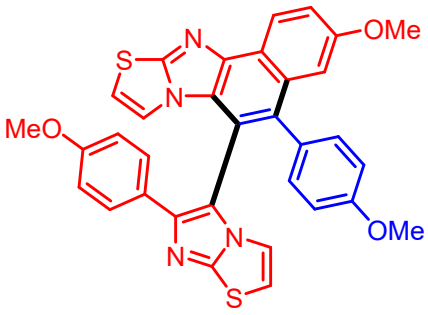


11

6-(6-([1,1'-biphenyl]-4-yl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)-3-phenylnaphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**11**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 51.1 mg, 75%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.92 (d, *J* = 8.4 Hz, 1H), 8.10 - 8.01 (m, 2H), 7.70 (d, *J* = 8.2 Hz, 2H), 7.62 (d, *J* = 7.1 Hz, 2H), 7.52 (d, *J* = 7.2 Hz, 2H), 7.47 - 7.40 (m, 4H), 7.37 (t, *J* = 7.6 Hz, 3H), 7.28 (dd, *J* = 7.9, 5.0 Hz, 2H), 7.06 (dd, *J* = 8.5, 2.2 Hz, 1H), 6.88 - 6.82 (m, 3H), 6.68 - 6.58 (m, 3H), 3.75 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.97, 155.72, 149.47, 144.88, 144.09, 141.28, 140.45, 140.09, 138.64, 135.74, 132.78, 131.40, 130.99, 129.53, 129.01, 128.86, 127.59, 127.48, 127.42, 127.21, 126.89, 126.58, 126.20, 126.02, 124.76, 123.19, 118.49, 117.49, 116.68, 114.06, 113.86, 113.61, 112.74, 112.14, 55.29; HRMS (ESI-TOF) Calcd for $C_{43}H_{29}N_4OS_2$ [M+H]$^+$: 681.1777; found: 681.1775.

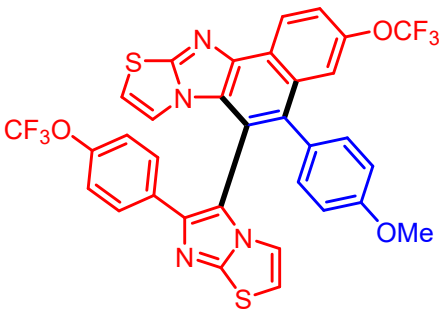


12

3-methoxy-5-(4-methoxyphenyl)-6-(6-(4-methoxyphenyl)imidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (Only ^{1}H NMR spectra data was obtained because of the poor solubility of product **12**) (**12**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:15:30; 29.4 mg, 50%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.75 (d, *J* = 9.0 Hz, 1H), 7.53 (d, *J* = 8.5 Hz, 2H), 7.44 (dd, *J* = 8.9, 2.4 Hz, 1H), 7.23 (dd, *J* = 8.5, 2.3 Hz, 1H), 7.18 (d, *J* = 2.5 Hz, 1H), 6.96 (dd, *J* = 8.5, 2.3 Hz, 1H), 6.84 - 6.78 (m, 3H), 6.70 (d, *J* = 8.6 Hz, 2H), 6.68 - 6.57 (m, 3H), 3.77 (s, 3H), 3.75 (s, 3H), 3.72 (s, 3H); HRMS (ESI-TOF) Calcd for $C_{33}H_{25}N_4O_3S_2$ $[M+H]^+$: 589.1363; found: 589.1357.



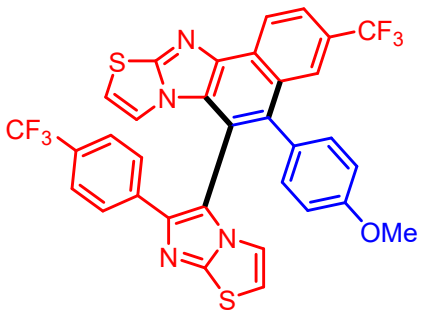
13

5-(4-methoxyphenyl)-3-(trifluoromethoxy)-6-(6-(4-(trifluoromethoxy)phenyl)imidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**13**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:8:30; 28.6 mg, 41%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.85 (d, *J* = 8.0 Hz, 1H), 7.63 (d, *J* = 12.4 Hz, 2H), 7.59 - 7.51 (m, 2H), 7.12 (dd, *J* = 8.4, 2.3 Hz, 1H), 7.00 (d, *J* = 8.0 Hz, 2H), 6.90

- 6.80 (m, 3H), 6.73 (d, J = 4.6 Hz, 1H), 6.69 (d, J = 4.5 Hz, 1H), 6.65 (d, J = 4.6 Hz, 1H), 6.62 (dd, J = 8.5, 2.7 Hz, 1H), 3.75 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 159.21, 156.09, 149.74, 148.62, 147.10, 144.14, 143.99, 135.07, 132.50, 131.27, 131.23, 131.06, 128.70, 127.29, 125.86, 124.75, 124.71, 121.99, 121.77, 121.46, 121.19, 119.43, 119.27, 119.21, 118.16, 117.31, 116.46, 114.57, 114.04, 113.66, 113.29, 112.64, 55.30; ^{19}F NMR (376 MHz, Chloroform-*d*) δ -57.63, -57.81; HRMS (ESI-TOF) Calcd for $C_{33}H_{19}F_6N_4O_3S_2$ $[M+H]^+$: 697.0797; found: 697.0796.

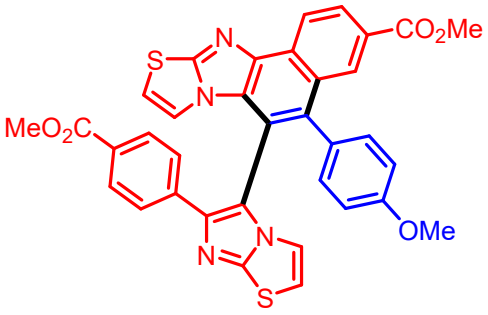


14

5-(4-methoxyphenyl)-3-(trifluoromethyl)-6-(6-(4-(trifluoromethyl)phenyl)imidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**14**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 27.9 mg, 42%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.94 (d, J = 8.6 Hz, 1H), 8.12 (s, 1H), 7.94 (dd, J = 8.6, 1.8 Hz, 1H), 7.65 (d, J = 8.2 Hz, 2H), 7.40 (d, J = 8.3 Hz, 2H), 7.13 (dd, J = 8.4, 2.3 Hz, 1H), 6.93 - 6.81 (m, 3H), 6.75 - 6.71 (m, 2H), 6.69 (d, J = 4.6 Hz, 1H), 6.63 (dd, J = 8.5, 2.7 Hz, 1H), 3.76 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 159.34, 156.35, 150.00, 143.99, 143.84, 137.17, 136.17, 131.17, 131.10, 129.67, 129.57, 129.25, 129.01, 128.42, 127.97, 127.65, 125.95, 125.80 (q, J = 4.0 Hz), 125.61 (q, J = 5.1 Hz), 123.77, 123.47, 123.44, 123.14, 122.81, 118.01, 117.25, 117.19, 114.48, 114.11, 113.76, 113.74, 113.13, 55.33; ^{19}F NMR (376 MHz, Chloroform-*d*) δ -61.89, -62.60; HRMS (ESI-TOF) Calcd for $C_{33}H_{19}F_6N_4OS_2$ $[M+H]^+$: 665.0899; found: 665.0895.

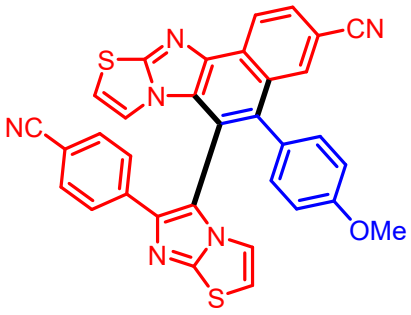


15

methyl6-(6-(4-(methoxycarbonyl)phenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole-3-carboxylate (**15**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 29.6 mg, 46%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.88 (d, *J* = 8.6 Hz, 1H), 8.60 (d, *J* = 1.6 Hz, 1H), 8.37 (dd, *J* = 8.6, 1.6 Hz, 1H), 7.85 - 7.79 (m, 2H), 7.67 - 7.60 (m, 2H), 7.17 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.92 (dd, *J* = 8.5, 2.3 Hz, 1H), 6.87 - 6.80 (m, 2H), 6.73 (dd, *J* = 14.8, 4.6 Hz, 2H), 6.65 (d, *J* = 4.6 Hz, 1H), 6.62 (dd, *J* = 8.5, 2.7 Hz, 1H), 3.92 (s, 3H), 3.83 (s, 3H), 3.76 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 167.34, 166.88, 159.28, 156.21, 149.92, 143.83, 136.74, 131.32, 131.22, 130.89, 130.21, 129.90, 129.09, 128.76, 127.55, 127.42, 125.73, 125.65, 122.98, 118.15, 117.56, 117.38, 114.10, 114.01, 113.73, 113.63, 112.94, 55.35, 52.44, 52.19; HRMS (ESI-TOF) Calcd for $C_{35}H_{25}N_4O_5S_2$ $[M+H]^+$: 645.1261; found: 645.1268.

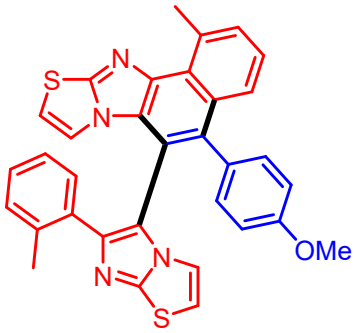


16

6-(6-(4-cyanophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole-3-carbonitrile (**16**):

Orange solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:15:30; 11.6 mg, 20%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.90 (d, *J* = 8.5 Hz, 1H), 8.18 (d, *J* = 1.5 Hz, 1H), 7.90 (dd, *J* = 8.5, 1.6 Hz, 1H), 7.64 - 7.54 (m, 2H), 7.43 (d, *J* = 8.5 Hz, 2H), 7.09 (dd, *J* = 8.4, 2.2 Hz, 1H), 6.90 - 6.80 (m, 3H), 6.77 (d, *J* = 4.5 Hz, 1H), 6.73 (d, *J* = 4.6 Hz, 1H), 6.69 (d, *J* = 4.6 Hz, 1H), 6.64 (dd, *J* = 8.5, 2.7 Hz, 1H), 3.77 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 159.56, 156.76, 150.35, 143.76, 143.52, 138.06, 135.75, 134.10, 132.68, 131.07, 130.99, 129.82, 129.15, 128.50, 127.87, 126.14, 125.89, 124.00, 119.43, 118.86, 117.83, 117.40, 117.15, 114.71, 114.29, 114.26, 113.99, 113.65, 110.96, 109.45, 55.41; HRMS (ESI-TOF) Calcd for $C_{33}H_{19}N_6OS_2$ $[M+H]^+$: 579.1056; found: 579.1044.

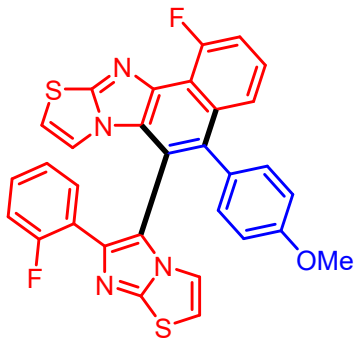


17

5-(4-methoxyphenyl)-1-methyl-6-(6-(*o*-tolyl)imidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**17**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:15:30; 41.7 mg, 75%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.47 (dd, *J* = 18.0, 7.7 Hz, 2H), 7.34 - 7.27 (m, 1H), 7.12 - 7.02 (m, 2H), 6.90 (d, *J* = 3.7 Hz, 1H), 6.83 (s, 1H), 6.78 (dd, *J* = 8.1, 2.0 Hz, 3H), 6.72 - 6.64 (m, 2H), 6.56 (dd, *J* = 8.4, 2.6 Hz, 1H), 6.45 (s, 1H), 6.33 (dd, *J* = 8.4, 2.1 Hz, 1H), 3.79 (s, 3H), 3.34 (s, 3H), 2.03 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.67, 153.71, 148.23, 146.89, 144.50, 137.57, 136.73, 135.38, 133.00, 131.93, 131.68, 130.78, 130.67, 130.23, 129.91, 129.34, 127.75, 126.51, 126.16, 126.11, 125.28, 124.87, 118.17, 117.71, 117.01, 113.37, 113.34, 113.27, 112.88, 112.59, 55.35, 24.33, 20.70; HRMS (ESI-TOF) Calcd for $C_{33}H_{25}N_4OS_2$ $[M+H]^+$: 557.1464; found: 557.1478.

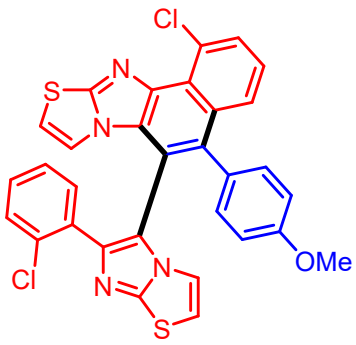


18

1-fluoro-6-(6-(2-fluorophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**18**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:22:30; 32.2 mg, 57%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.46 (dd, *J* = 8.3, 1.3 Hz, 1H), 7.42 - 7.38 (m, 1H), 7.36 - 7.28 (m, 2H), 7.19 - 7.11 (m, 1H), 7.02 - 6.96 (m, 1H), 6.92 (d, *J* = 4.4 Hz, 1H), 6.85 (dd, *J* = 8.4, 2.2 Hz, 1H), 6.82 - 6.75 (m, 2H), 6.74 - 6.69 (m, 2H), 6.68 (d, *J* = 4.6 Hz, 1H), 6.61 (dd, *J* = 8.5, 2.2 Hz, 1H), 6.54 (dd, *J* = 8.5, 2.7 Hz, 1H), 3.76 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 160.54 (d, *J* = 48.5 Hz), 158.88, 158.04 (d, *J* = 53.5 Hz), 155.54 (d, *J* = 4.0 Hz), 149.49, 141.27, 140.70, 135.54, 133.01 (d, *J* = 5.1 Hz), 131.53, 130.95 (d, *J* = 3.0 Hz), 130.44, 129.68, 129.60 (d, *J* = 8.1 Hz), 125.79, 125.11 (d, *J* = 8.1 Hz), 124.14 (d, *J* = 4.0 Hz), 123.88 (d, *J* = 4.0 Hz), 121.76 (d, *J* = 13.1 Hz), 118.50, 118.09, 117.24, 117.09, 115.90, 115.67, 114.57, 113.78, 113.42 (d, *J* = 22.2 Hz), 112.77, 112.53 (d, *J* = 19.2 Hz), 55.35; ^{19}F NMR (376 MHz, Chloroform-*d*) δ -114.33, -115.40; HRMS (ESI-TOF) Calcd for $C_{31}H_{19}F_2N_4OS_2$ $[M+H]^+$: 565.0963; found: 565.0962.

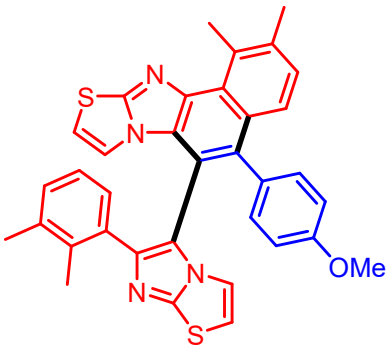


19

1-chloro-6-(6-(2-chlorophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**19**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:15:30; 12.5 mg, 21%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.74 (d, *J* = 7.5 Hz, 1H), 7.60 (s, 1H), 7.29 (d, *J* = 7.5 Hz, 1H), 7.17 (dd, *J* = 8.0, 1.5 Hz, 1H), 7.15 - 7.08 (m, 1H), 7.04 - 6.97 (m, 1H), 6.92 - 6.83 (m, 3H), 6.77 (d, *J* = 4.6 Hz, 1H), 6.74 - 6.60 (m, 2H), 6.57 - 6.50 (m, 2H), 6.43 (dd, *J* = 8.4, 2.0 Hz, 1H), 3.80 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.87, 154.61, 148.96, 143.98, 142.41, 136.37, 133.03, 132.92, 132.63, 132.33, 131.83, 130.15, 130.07, 129.80, 129.76, 129.72, 129.17, 127.35, 126.45, 126.30, 124.95, 124.65, 118.71, 118.10, 116.94, 114.35, 114.26, 113.65, 113.54, 113.13, 55.41; HRMS (ESI-TOF) Calcd for $C_{31}H_{19}Cl_2N_4OS_2$ $[M+H]^+$: 597.0372; found: 597.0369.

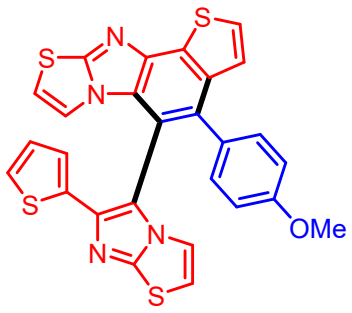


20

6-(6-(2,3-dimethylphenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)-1,2-dimethylnaphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**20**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:7:30; 39.7 mg, 68%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.33 (d, *J* = 8.6 Hz, 1H), 7.21 (d, *J* = 8.6 Hz, 1H), 6.95 (d, *J* = 7.4 Hz, 1H), 6.90 (d, *J* = 4.5 Hz, 1H), 6.81 - 6.76 (m, 2H), 6.73 - 6.61 (m, 3H), 6.58 - 6.44 (m, 2H), 6.36 (d, *J* = 4.6 Hz, 1H), 6.30 (dd, *J* = 8.4, 2.2 Hz, 1H), 3.80 (s, 3H), 3.32 (s, 3H), 2.53 (s, 3H), 2.22 (s, 3H), 1.88 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.60, 153.06, 148.04, 147.17, 144.23, 137.09, 136.77, 136.13, 136.10, 133.03, 132.91, 131.87, 130.82, 130.66, 130.21, 129.39, 128.05, 127.99, 126.77, 126.42, 125.53, 124.85, 118.41, 117.74, 117.08, 113.39, 113.16, 112.55, 111.86, 55.32, 21.22, 20.73, 18.52, 16.81; HRMS (ESI-TOF) Calcd for $C_{35}H_{29}N_4OS_2$ $[M+H]^+$: 585.1777; found: 585.1778.

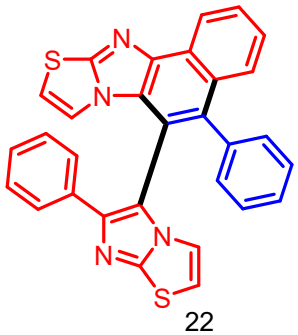


21

4-(4-methoxyphenyl)-5-(6-(thiophen-2-yl)imidazo[2,1-*b*]thiazol-5-yl)thieno[3",2":5',6']benzo[1',2':4,5]imidazo[2,1-*b*]thiazole (**21**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:15:30; 24.3 mg, 45%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.48 (d, *J* = 5.4 Hz, 1H), 7.37 (d, *J* = 5.4 Hz, 2H), 7.12 (dd, *J* = 5.0, 1.2 Hz, 1H), 7.02 (d, *J* = 9.5 Hz, 1H), 6.91 (d, *J* = 4.6 Hz, 1H), 6.85 (dd, *J* = 3.7, 1.2 Hz, 2H), 6.80 (dd, *J* = 5.0, 3.6 Hz, 1H), 6.76 (d, *J* = 4.5 Hz, 1H), 6.63 (d, *J* = 9.4 Hz, 1H), 6.58 (d, *J* = 4.5 Hz, 2H), 3.74 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 159.04, 156.86, 149.69, 142.59, 141.64, 137.35, 137.09, 132.47, 131.52, 130.87, 130.35, 127.98, 125.42, 125.34, 125.03, 125.00, 123.10, 118.93, 117.44, 115.33, 113.99, 113.66, 112.60, 111.53, 109.57, 55.32; HRMS (ESI-TOF) Calcd for $C_{27}H_{17}N_4OS_4$ $[M+H]^+$: 541.028; found: 541.0281.

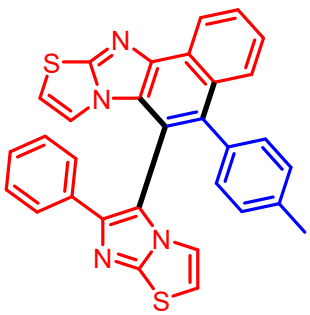


22

5-phenyl-6-(6-phenylimidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**22**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:7:30; 18.9 mg, 38%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.85 (d, *J* = 8.3 Hz, 1H), 7.85 - 7.73 (m, 2H), 7.57 (dd, *J* = 6.7, 2.9 Hz, 2H), 7.54 - 7.48 (m, 1H), 7.27 (d, *J* = 5.1 Hz, 2H), 7.23 - 7.11 (m, 4H), 7.10 - 7.00 (m, 2H), 6.81 (dd, *J* = 13.7, 4.5 Hz, 2H), 6.61 (dd, *J* = 10.3, 4.5 Hz, 2H);^{13}C NMR (101 MHz, Chloroform-*d*) δ 155.59, 149.39, 145.37, 144.28, 137.56, 135.74, 133.82, 130.40, 130.24, 130.19, 128.78, 128.48, 127.92, 127.69, 127.61, 127.48, 125.94, 125.85, 124.64, 122.56, 118.43, 117.45, 116.43, 113.52, 112.59, 112.04; HRMS (ESI-TOF) Calcd for $C_{30}H_{19}N_4S_2$ $[M+H]^+$: 499.1046; found: 499.1045.

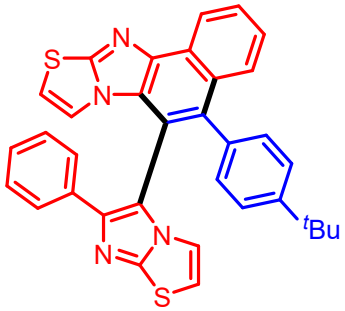


23

6-(6-phenylimidazo[2,1-*b*]thiazol-5-yl)-5-(*p*-tolyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**23**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 15.4 mg, 30%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.84 (d, *J* = 8.1 Hz, 1H), 7.82 (d, *J* = 8.4 Hz, 1H), 7.79 - 7.73 (m, 1H), 7.58 (dd, *J* = 6.8, 3.0 Hz, 2H), 7.54 - 7.48 (m, 1H), 7.19 - 7.11 (m, 4H), 7.07 (d, *J* = 7.9 Hz, 1H), 6.95 - 6.87 (m, 2H), 6.84 (d, *J* = 4.4 Hz, 1H), 6.77 (d, *J* = 4.6 Hz, 1H), 6.65 (d, *J* = 4.5 Hz, 1H), 6.60 (d, *J* = 4.6 Hz, 1H), 2.27 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 155.47, 149.27, 144.95, 144.08, 137.31, 136.00, 134.42, 133.58, 130.57, 130.04, 129.98, 129.28, 128.82, 128.43, 128.04, 127.70, 127.62, 127.45, 125.91, 125.81, 124.61, 122.54, 118.39, 117.54, 116.58, 113.33, 112.77, 112.10, 21.30; HRMS (ESI-TOF) Calcd for $C_{31}H_{21}N_4S_2$ $[M+H]^+$: 513.1202; found: 513.1202.

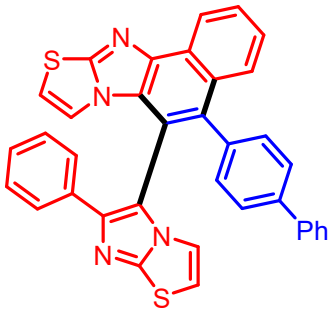


24

5-(4-(*tert*-butyl)phenyl)-6-(6-phenylimidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**24**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:11:30; 24.9 mg, 45%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.84 (d, *J* = 9.7 Hz, 1H), 7.92 (d, *J* = 8.5 Hz, 1H), 7.80 - 7.73 (m, 1H), 7.59 - 7.48 (m, 3H), 7.28 (dd, *J* = 8.0, 2.1 Hz, 1H), 7.21 (dd, *J* = 8.0, 2.0 Hz, 1H), 7.16 - 7.10 (m, 3H), 7.05 (dd, *J* = 8.1, 2.1 Hz, 1H), 6.94 (dd, *J* = 8.1, 2.0 Hz, 1H), 6.84 (d, *J* = 4.6 Hz, 1H), 6.77 (d, *J* = 4.5 Hz, 1H), 6.57 (d, *J* = 4.5 Hz, 2H), 1.24 (s, 9H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 155.46, 150.43, 149.40, 145.37, 144.17, 135.63, 134.37, 133.87, 130.39, 130.12, 129.84, 128.75, 128.07, 127.58, 127.52, 127.47, 125.95, 125.76, 125.30, 124.63, 124.50, 122.52, 118.55, 117.69, 116.70, 113.56, 112.14, 111.81, 34.62, 31.35; HRMS (ESI-TOF) Calcd for $C_{34}H_{27}N_4S_2$ $[M+H]^+$: 555.1672; found: 555.1673.

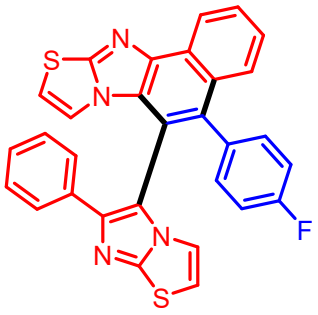


25

5-([1,1'-biphenyl]-4-yl)-6-(6-phenylimidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**25**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 27.6 mg, 48%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.87 (d, *J* = 8.2 Hz, 1H), 7.90 (d, *J* = 8.5 Hz, 1H), 7.80 (d, *J* = 7.5 Hz, 1H), 7.62 - 7.48 (m, 6H), 7.42 (t, *J* = 7.5 Hz, 2H), 7.37 - 7.28 (m, 3H), 7.19 - 7.13 (m, 3H), 7.09 (dd, *J* = 8.0, 1.8 Hz, 1H), 6.83 (dd, *J* = 19.2, 4.3 Hz, 2H), 6.63 (dd, *J* = 8.1, 4.1 Hz, 2H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 155.65, 149.48, 145.39, 144.33, 140.57, 140.21, 136.56, 135.37, 133.77, 130.71, 130.69, 130.42, 128.92, 128.83, 127.95, 127.71, 127.68, 127.55, 127.52, 127.12, 127.09, 126.36, 126.01, 125.95, 124.68, 122.63, 118.49, 117.45, 116.44, 113.54, 112.80, 112.14; HRMS (ESI-TOF) Calcd for $C_{36}H_{23}N_4S_2$ $[M+H]^+$: 575.1359; found: 575.1358.

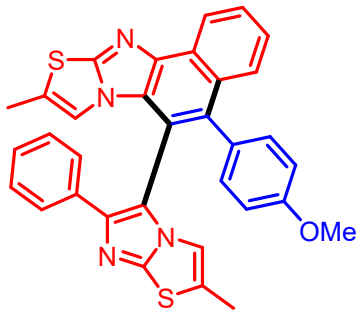


26

5-(4-fluorophenyl)-6-(6-phenylimidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**26**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 23.7 mg, 46%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.85 (d, *J* = 7.5 Hz, 1H), 7.77 (dd, *J* = 14.0, 7.2 Hz, 2H), 7.57 - 7.49 (m, 3H), 7.24 - 7-18 (m, 1H), 7.17 - 7.11 (m, 3H), 7.01 - 6.91 (m, 2H), 6.82 (d, *J* = 4.4 Hz, 1H), 6.80 - 6.71 (m, 2H), 6.67 (d, *J* = 4.4 Hz, 1H), 6.62 (d, *J* = 4.4 Hz, 1H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 163.41, 160.96, 155.73, 149.50, 145.50, 144.40, 134.54, 133.75, 133.44 (d, *J* = 3.0 Hz), 131.98 (d, *J* = 8.1 Hz), 131.68 (d, *J* = 8.1 Hz), 130.39, 128.81, 127.71, 127.66 (d, *J* = 4.0 Hz), 127.47, 125.99, 125.94, 124.61, 122.65, 118.35, 117.19, 116.17, 115.49 (d, *J* = 21.2 Hz), 114.93, 114.71, 113.75, 113.00, 112.24; ^{19}F NMR (376 MHz, Chloroform-*d*) δ -114.27; HRMS (ESI-TOF) Calcd for $C_{30}H_{18}FN_4S_2$ $[M+H]^+$: 517.0951; found: 517.0954.

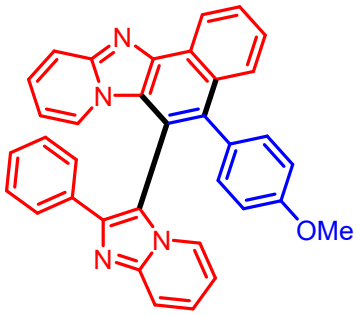


27

5-(4-methoxyphenyl)-9-methyl-6-(2-methyl-6-phenylimidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**27**):

Pale yellow solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:10:30; 28.9 mg, 52%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.80 (d, *J* = 8.3 Hz, 1H), 7.78 (d, *J* = 8.5 Hz, 1H), 7.75 - 7.69 (m, 1H), 7.54 (dd, *J* = 7.5, 2.2 Hz, 2H), 7.5 - 7.43 (m, 1H), 7.18 - 7.10 (m, 4H), 6.91 (dd, *J* = 8.5, 2.3 Hz, 1H), 6.83 - 6.76 (m, 1H), 6.59 (dd, *J* = 8.5, 2.8 Hz, 1H), 6.54 (d, *J* = 1.5 Hz, 1H), 6.43 (d, *J* = 1.6 Hz, 1H), 3.73 (s, 3H), 2.26 (d, *J* = 1.4 Hz, 3H), 2.23 (d, *J* = 1.5 Hz, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.82, 154.98, 148.62, 143.95, 143.08, 135.35, 134.09, 131.37, 131.12, 130.50, 129.81, 128.65, 127.94, 127.39, 127.34, 127.23, 126.66, 125.96, 125.73, 125.44, 124.74, 122.32, 116.44, 114.54, 113.91, 113.71, 113.57, 113.21, 55.23, 14.24, 14.16; HRMS (ESI-TOF) Calcd for $C_{33}H_{25}N_4OS_2$ $[M+H]^+$: 557.1464; found: 557.1466.

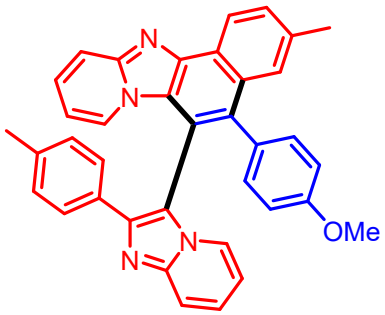


28

5-(4-methoxyphenyl)-6-(2-phenylimidazo[1,2-*a*]pyridin-3-yl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**28**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 21.7 mg, 42%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.05 (d, *J* = 9.6 Hz, 1H), 7.89 (d, *J* = 9.2 Hz, 1H), 7.82 (t, *J* = 7.0 Hz, 1H), 7.73 (dd, *J* = 14.7, 8.7 Hz, 2H), 7.67 - 7.61 (m, 2H), 7.60 - 7.54 (m, 1H), 7.47 (d, *J* = 6.8 Hz, 1H), 7.36 - 7.27 (m, 2H), 7.25 - 7.13 (m, 4H), 6.96 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.77 (dd, *J* = 8.5, 2.3 Hz, 1H), 6.72 - 6.64 (m, 1H), 6.61 (dd, *J* = 8.4, 2.7 Hz, 1H), 6.51 (dd, *J* = 8.3, 2.7 Hz, 2H), 3.68 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.92, 148.14, 144.65, 143.49, 141.49, 137.75, 132.99, 132.10, 131.08, 130.02, 129.40, 128.82, 128.36, 128.27, 127.86, 127.47, 126.99, 126.78, 126.07, 125.28, 123.47, 123.33, 122.66, 117.97, 117.27, 115.27, 113.39, 113.35, 113.09, 113.01, 112.55, 55.20; HRMS (ESI-TOF) Calcd for $C_{35}H_{25}N_4O$ $[M+H]^+$: 517.2023; found: 517.2022.

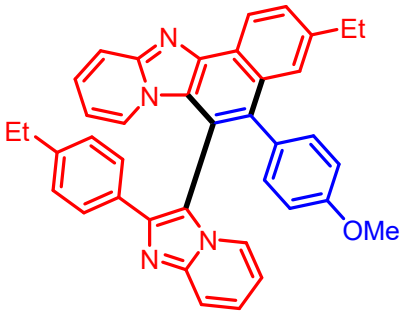


29

5-(4-methoxyphenyl)-3-methyl-6-(2-(*p*-tolyl)imidazo[1,2-*a*]pyridin-3-yl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**29**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 21.7 mg, 40%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.94 (d, *J* = 8.3 Hz, 1H), 7.85 (d, *J* = 9.2 Hz, 1H), 7.70 - 7.60 (m, 2H), 7.54 (d, *J* = 8.3 Hz, 2H), 7.51 (s, 1H), 7.43 (d, *J* = 6.9 Hz, 1H), 7.32 - 7.26 (m, 1H), 7.26 - 7.23 (m, 1H), 7.21 - 7.15 (m, 1H), 7.02 - 6.94 (m, 3H), 6.82 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.66 - 6.58 (m, 2H), 6.53 (dd, *J* = 8.4, 2.7 Hz, 1H), 6.49 - 6.44 (m, 1H), 3.68 (s, 3H), 2.48 (s, 3H), 2.23 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.79, 148.25, 144.81, 143.96, 141.82, 138.03, 137.07, 136.56, 132.31, 131.14, 130.62, 130.13, 129.70, 129.50, 127.91, 127.42, 126.79, 125.57, 125.47, 125.39, 123.41, 123.21, 122.41, 117.89, 117.25, 114.95, 113.45, 113.30, 113.06, 112.85, 112.21, 55.17, 22.18, 21.33; HRMS (ESI-TOF) Calcd for $C_{37}H_{29}N_4O$ $[M+H]^+$: 545.2336; found: 545.2332.



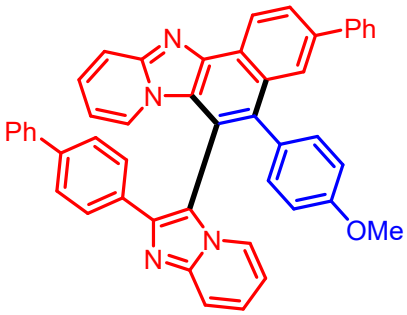
30

3-ethyl-6-(2-(4-ethylphenyl)imidazo[1,2-*a*]pyridin-3-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**30**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 25.7 mg, 45%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.97 (d, *J* = 8.4 Hz, 1H), 7.85 (d, *J* = 9.1 Hz, 1H), 7.71 (dd, *J* = 8.4, 1.7 Hz, 1H), 7.63 (d, *J* = 9.0 Hz, 1H), 7.59 - 7.51 (m, 3H), 7.41 (d, *J* = 6.8 Hz, 1H), 7.32 - 7.27 (m, 1H), 7.24 (d, *J* = 7.0 Hz, 1H), 7.16 (dd, *J* = 8.7, 7.1 Hz, 1H), 6.99 (dd, *J* = 8.3, 4.1 Hz, 3H), 6.82 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.65 - 6.56 (m, 2H), 6.52 (dd, *J* = 8.4, 2.8 Hz, 1H), 6.46 (t, *J* = 6.9 Hz, 1H), 3.69 (s, 3H), 2.77 (q, *J* = 7.6 Hz, 2H), 2.54 (q, *J* = 7.6 Hz, 2H), 1.26 (t, *J* = 7.6 Hz, 3H), 1.14 (t, *J* = 7.6 Hz, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.75, 148.33, 145.01, 144.35, 144.18, 142.82, 141.96, 137.14, 132.33, 131.24, 131.14, 130.16, 129.79, 128.45, 128.22, 127.76, 126.90, 126.37, 125.88, 125.44, 125.15, 123.36, 122.53, 117.95, 117.38,

114.99, 113.62, 113.25, 113.03, 112.65, 112.09, 55.18, 29.52, 28.64, 15.96, 15.37; HRMS (ESI-TOF) Calcd for $C_{39}H_{33}N_4O$ $[M+H]^+$: 573.2649; found: 573.2646.

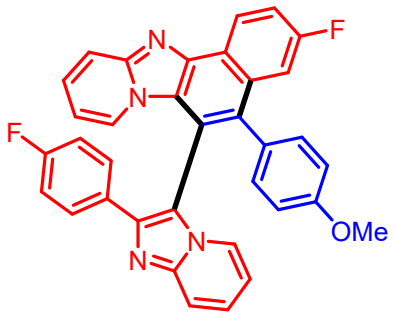


31

6-(2-([1,1'-biphenyl]-4-yl)imidazo[1,2-*a*]pyridin-3-yl)-5-(4-methoxyphenyl)-3-phenylnaphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**31**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 27.4 mg, 41%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.14 (d, *J* = 8.5 Hz, 1H), 8.11 (dd, *J* = 8.5, 1.8 Hz, 1H), 7.98 (d, *J* = 1.8 Hz, 1H), 7.90 (d, *J* = 9.3 Hz, 1H), 7.76 (d, *J* = 8.3 Hz, 2H), 7.70 (d, *J* = 9.1 Hz, 1H), 7.60 (d, *J* = 7.1 Hz, 2H), 7.51 (dd, *J* = 11.3, 7.6 Hz, 3H), 7.43 (t, *J* = 7.5 Hz, 4H), 7.40 - 7.32 (m, 4H), 7.29 (dd, *J* = 7.1, 2.9 Hz, 2H), 7.25 - 7.20 (m, 1H), 7.04 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.91 (dd, *J* = 8.5, 2.3 Hz, 1H), 6.70 - 6.59 (m, 2H), 6.58 - 6.48 (m, 2H), 3.69 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 158.91, 148.52, 145.01, 141.80, 141.19, 140.65, 140.44, 139.55, 137.80, 132.44, 131.20, 130.14, 129.42, 129.01, 128.86, 128.12, 127.66, 127.58, 127.49, 127.36, 127.31, 126.94, 126.70, 126.32, 125.67, 125.38, 123.95, 123.43, 122.88, 118.09, 117.43, 115.34, 113.82, 113.50, 113.21, 113.07, 112.42, 55.20; HRMS (ESI-TOF) Calcd for $C_{47}H_{33}N_4O$ $[M+H]^+$: 669.2649; found: 669.2640.

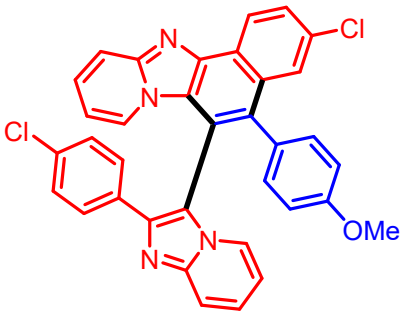


32

3-fluoro-6-(2-(4-fluorophenyl)imidazo[1,2-*a*]pyridin-3-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**32**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 24.3 mg, 44%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.04 (dd, *J* = 9.0, 5.8 Hz, 1H), 7.88 (d, *J* = 9.3 Hz, 1H), 7.65 (d, *J* = 9.3 Hz, 1H), 7.60 - 7.53 (m, 3H), 7.44 (d, *J* = 6.9 Hz, 1H), 7.40 - 7.29 (m, 2H), 7.26 - 7.17 (m, 2H), 6.96 - 6.81 (m, 3H), 6.73 (dd, *J* = 8.5, 2.3 Hz, 1H), 6.66 (t, *J* = 6.8 Hz, 1H), 6.61 (dd, *J* = 8.4, 2.7 Hz, 1H), 6.56 - 6.48 (m, 2H), 3.69 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 163.95, 162.70, 161.49, 160.26, 159.06, 146.87 (d, *J* = 345.4 Hz), 143.35, 141.93, 136.68, 133.51 (d, *J* = 9.1 Hz), 130.99, 129.88, 129.84, 129.09, 128.74 (d, *J* = 8.1 Hz), 128.29, 125.74 (d, *J* = 9.1 Hz), 125.59, 125.24, 124.38, 123.30, 122.39, 118.10, 117.57, 117.33 (d, *J* = 24.2 Hz), 115.72 (d, *J* = 22.2 Hz), 114.70, 114.45, 113.21 (d, *J* = 11.1 Hz), 113.15, 112.47, 112.26, 55.24; ^{19}F NMR (376 MHz, Chloroform-*d*) δ -112.68, -113.45; HRMS (ESI-TOF) Calcd for $C_{35}H_{23}F_2N_4O$ $[M+H]^+$: 553.1834; found: 553.1833.



33

3-chloro-6-(2-(4-chlorophenyl)imidazo[1,2-*a*]pyridin-3-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**33**):

White solid; Eluent: dichloromethane/ethyl acetate/petroleum ether 10:20:30; 23.4 mg, 40%;

^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.98 (d, *J* = 8.7 Hz, 1H), 7.87 (d, *J* = 9.4 Hz, 1H), 7.77 (dd, *J* = 8.7, 2.1 Hz, 1H), 7.70 (d, *J* = 2.0 Hz, 1H), 7.67 (d, *J* = 9.1 Hz, 1H), 7.53 (d, *J* = 8.6 Hz, 2H), 7.45 (d, *J* = 6.7 Hz, 1H), 7.39 - 7.30 (m, 1H), 7.23 (d, *J* = 7.9 Hz, 1H), 7.19 (d, *J* = 7.1 Hz, 1H), 7.16 - 7.10 (m, 2H), 6.91 (dd, *J* = 8.4, 2.3 Hz, 1H), 6.75 (dd, *J* = 8.3, 2.2 Hz, 1H), 6.68 (t, *J* = 6.8 Hz, 1H), 6.61 (dd, *J* = 8.4, 2.7 Hz, 1H), 6.58 - 6.48 (m, 2H), 3.70 (s, 3H); ^{13}C NMR (101 MHz, Chloroform-*d*) δ 159.13, 148.62, 145.05, 141.70, 136.61, 134.19, 133.03, 132.97, 131.94, 131.00, 129.93, 129.00, 128.75, 128.52, 128.47, 128.17, 127.19, 125.88, 125.18, 125.02, 123.35, 122.76, 118.20, 117.57, 115.05, 114.27, 113.69, 113.38, 113.31, 112.65, 55.25; HRMS (ESI-TOF) Calcd for $C_{35}H_{23}Cl_2N_4O$ $[M+H]^+$: 585.1243; found: 585.1243.

11、Reference

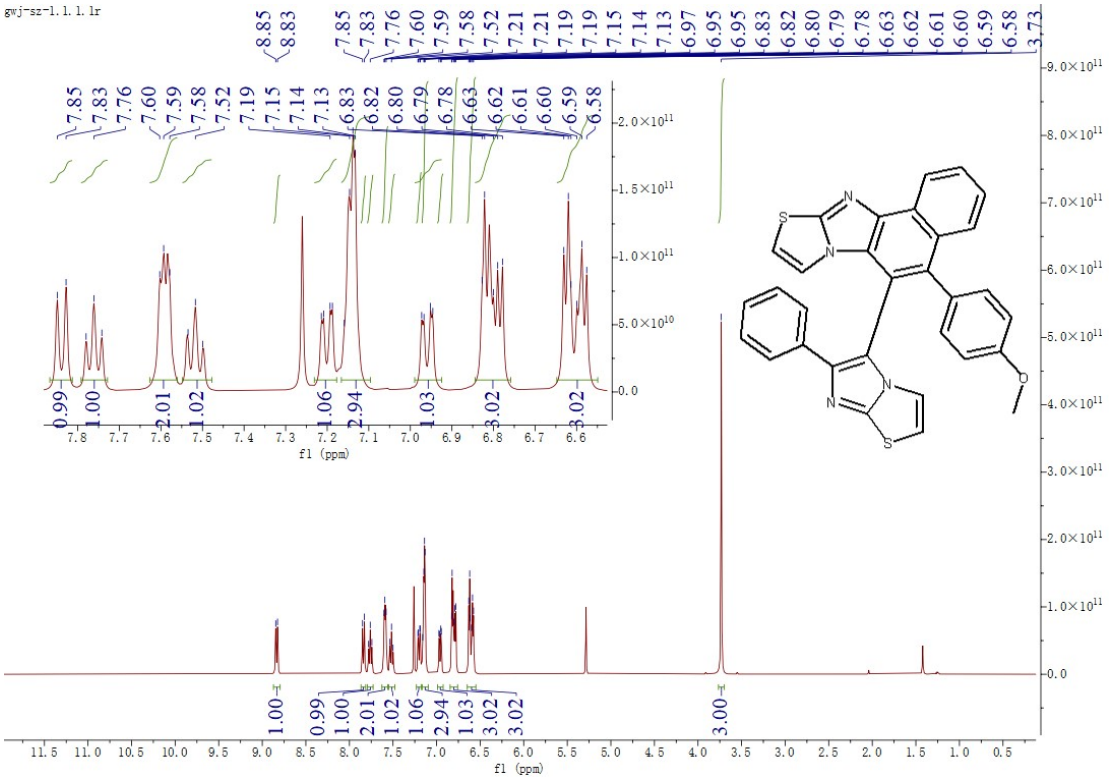
[1] W.-J. Guan, K. Ying, C.-C. Yuan, J.-L. Hang, C.-K. Liu, X.-X. Huang, Z. Fang and K. Guo, Catalyst- and oxidant-free electrooxidative site-selective 3/4+2 annulation to fused polycyclic heteroaromatics, *Green Chem*., 2022, **24**, 5191-5196.

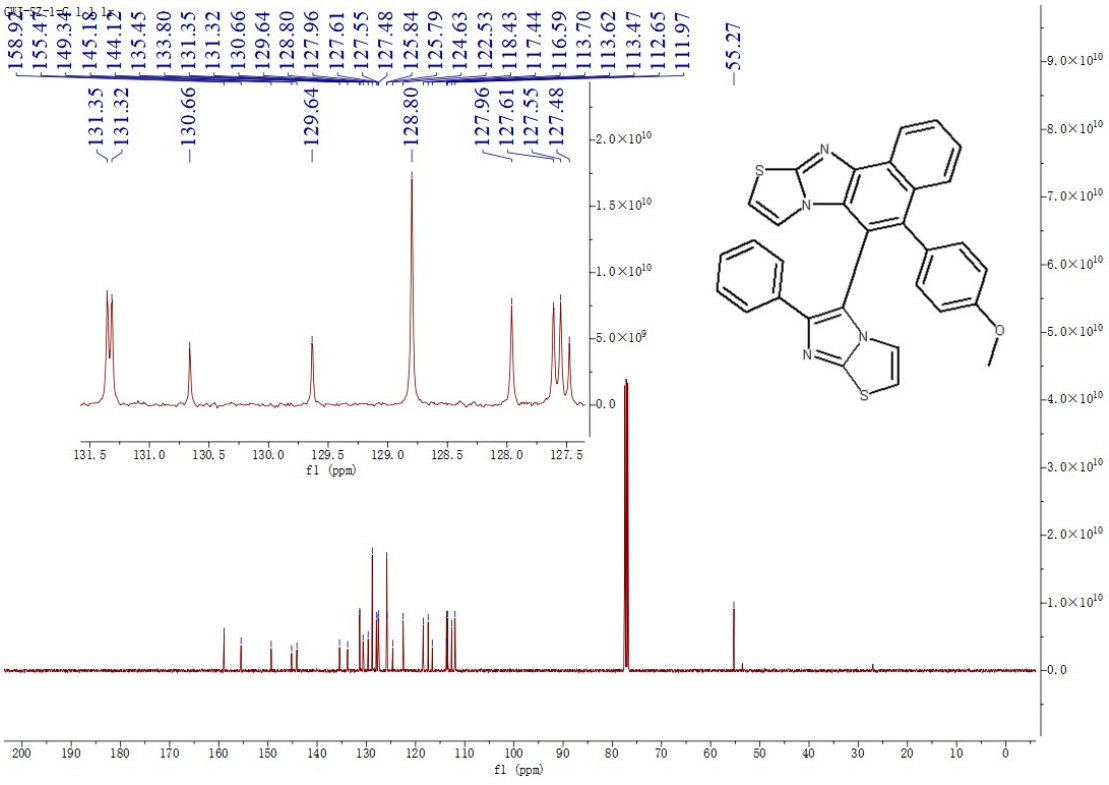
[2] R. Aeluri, M. Alla, S. Polepalli and N. Jain, Eur. Synthesis and antiproliferative activity of imidazo[1,2-*a*]pyrimidine Mannich bases, *J. Med. Chem*., 2015, **100**, 18-23.

[3] M. F. Baig, V. L. Nayak, P. Budaganaboyina, K. Mullagiri, S. Sunkari, J. Gour and A. Kamal, Synthesis and biological evaluation of imidazo[2,1-*b*]thiazole-benzimidazole conjugates as microtubule-targeting agents, *Bioorg. Chem*., 2018, **77**, 515-526.

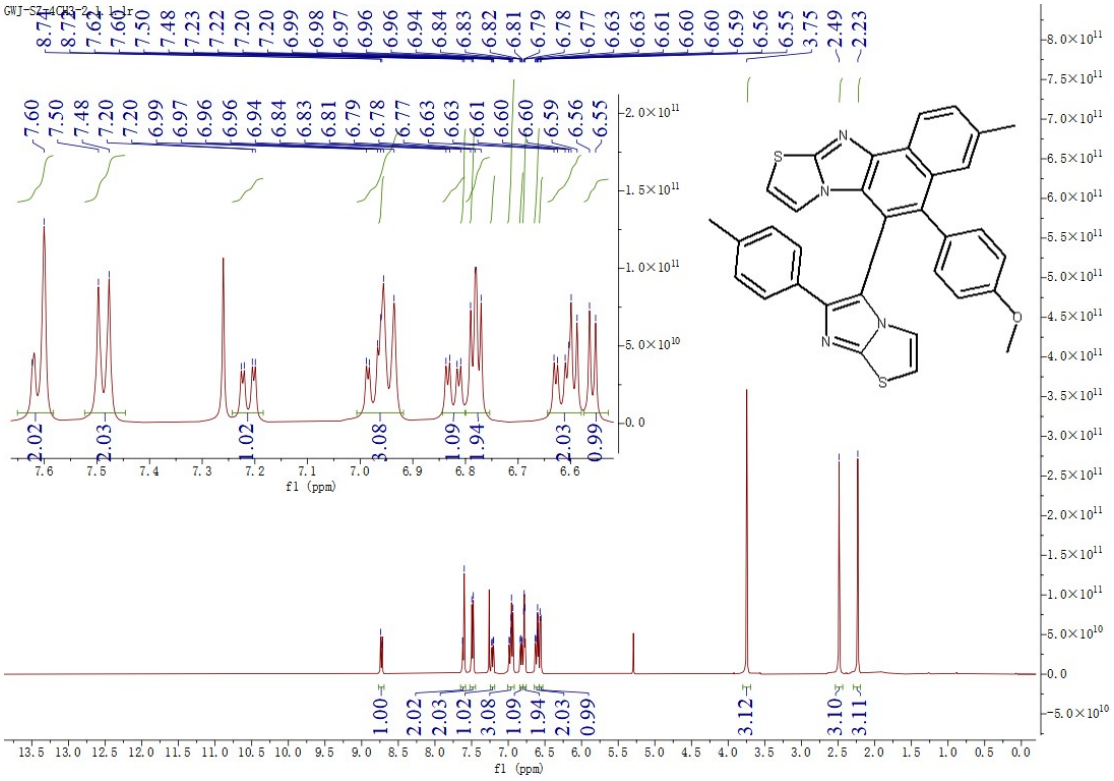
12、NMR Spectra for Electrolysis Products and 34-37, S5, S10

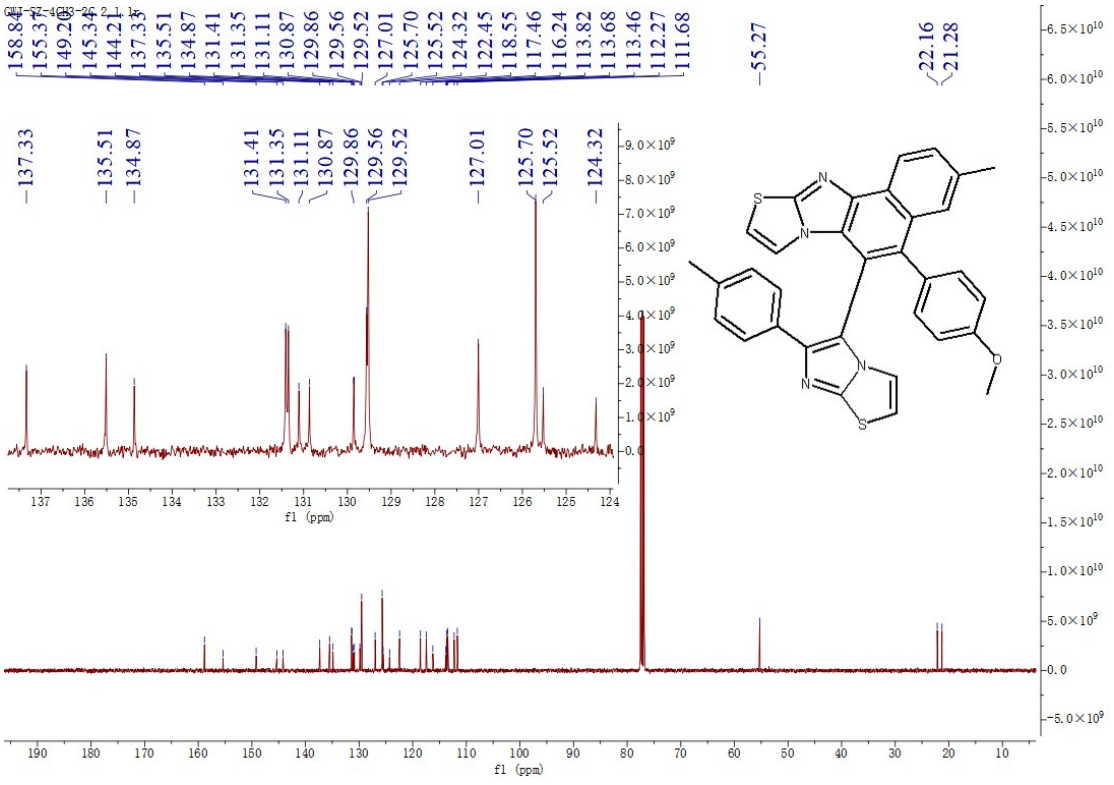
5-(4-methoxyphenyl)-6-(6-phenylimidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**3**):



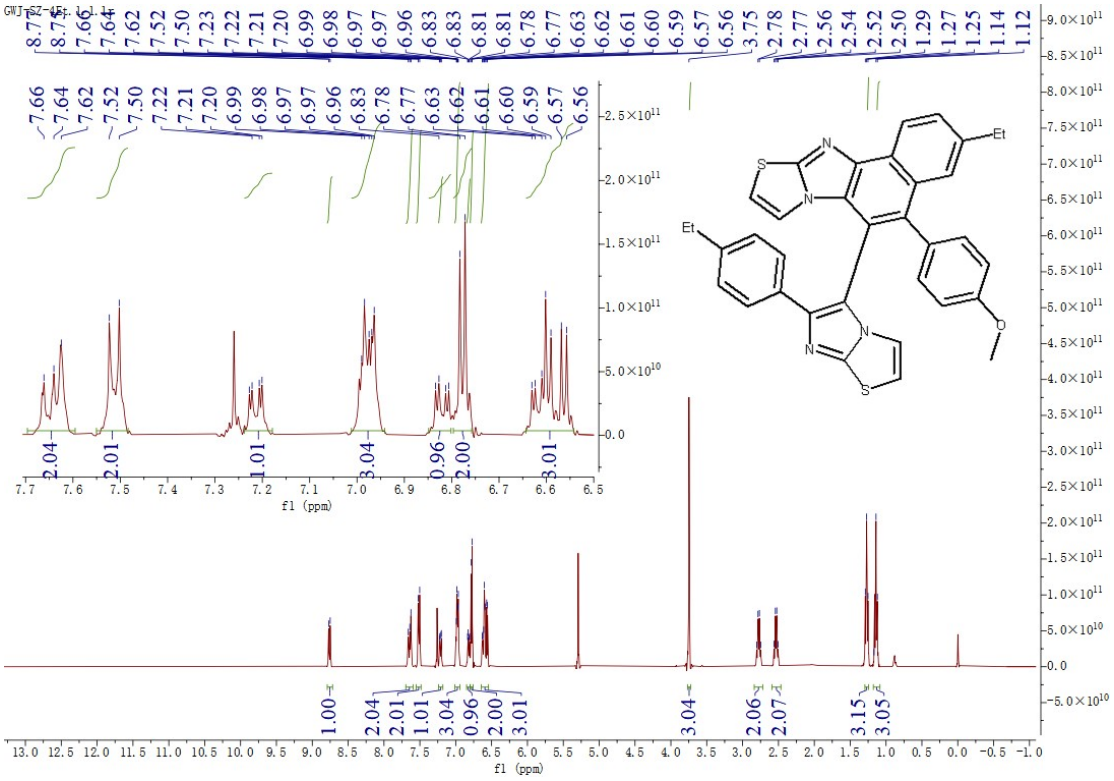


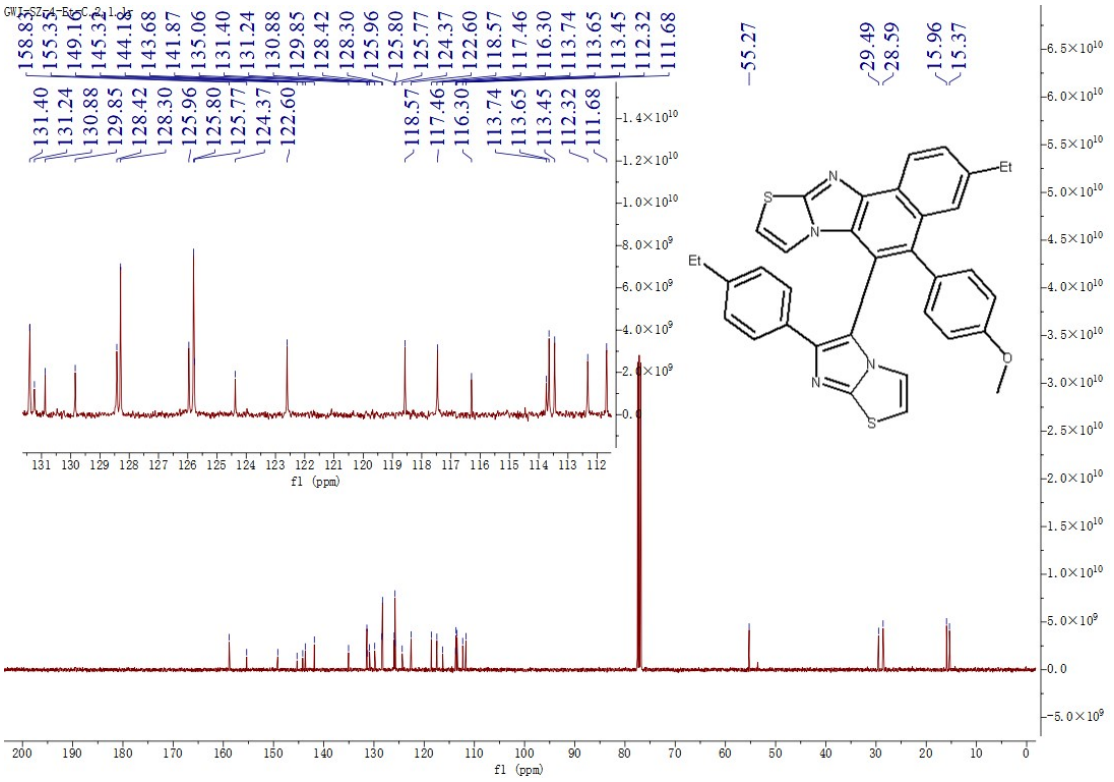
6-(4-methoxyphenyl)-3-methyl-6-(6-(*p*-tolyl)imidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**4**):



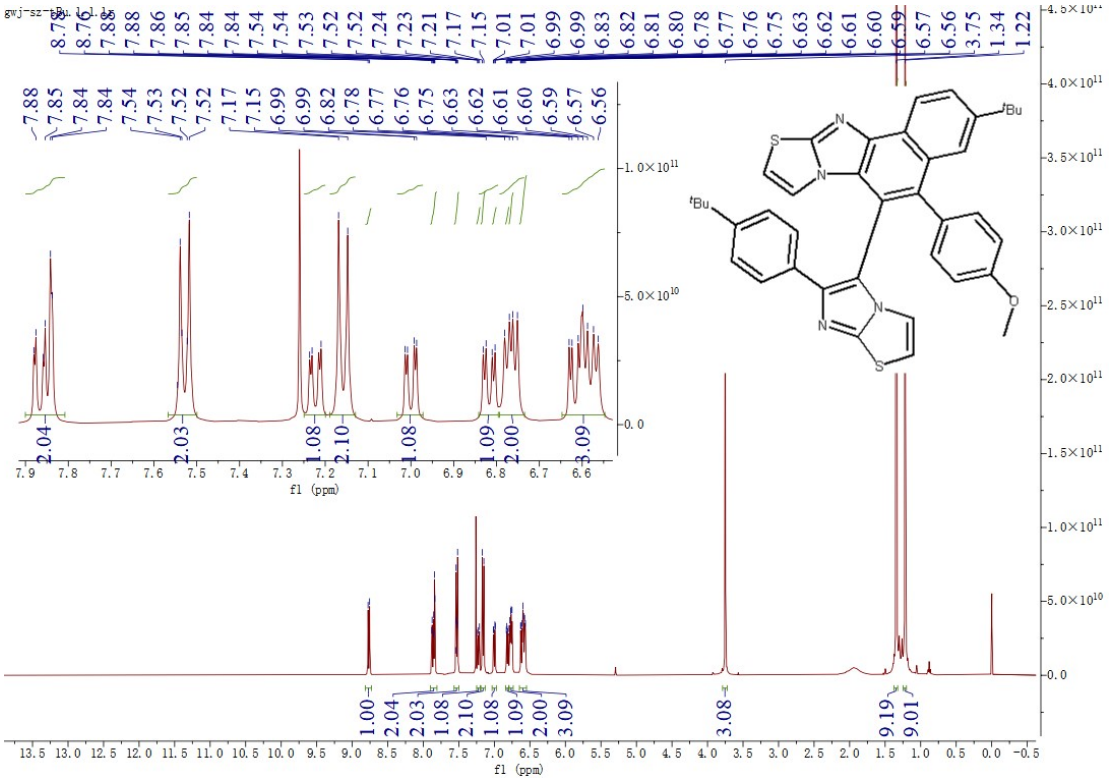


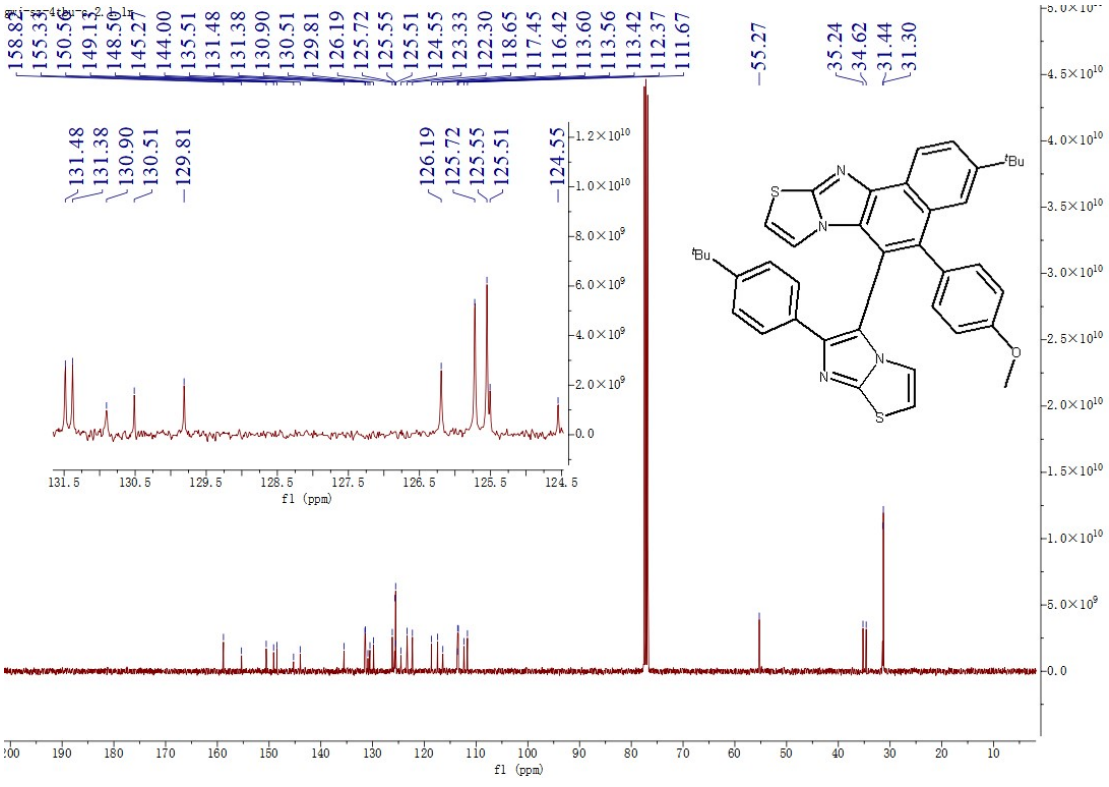
3-ethyl-6-(6-(4-ethylphenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**5**):



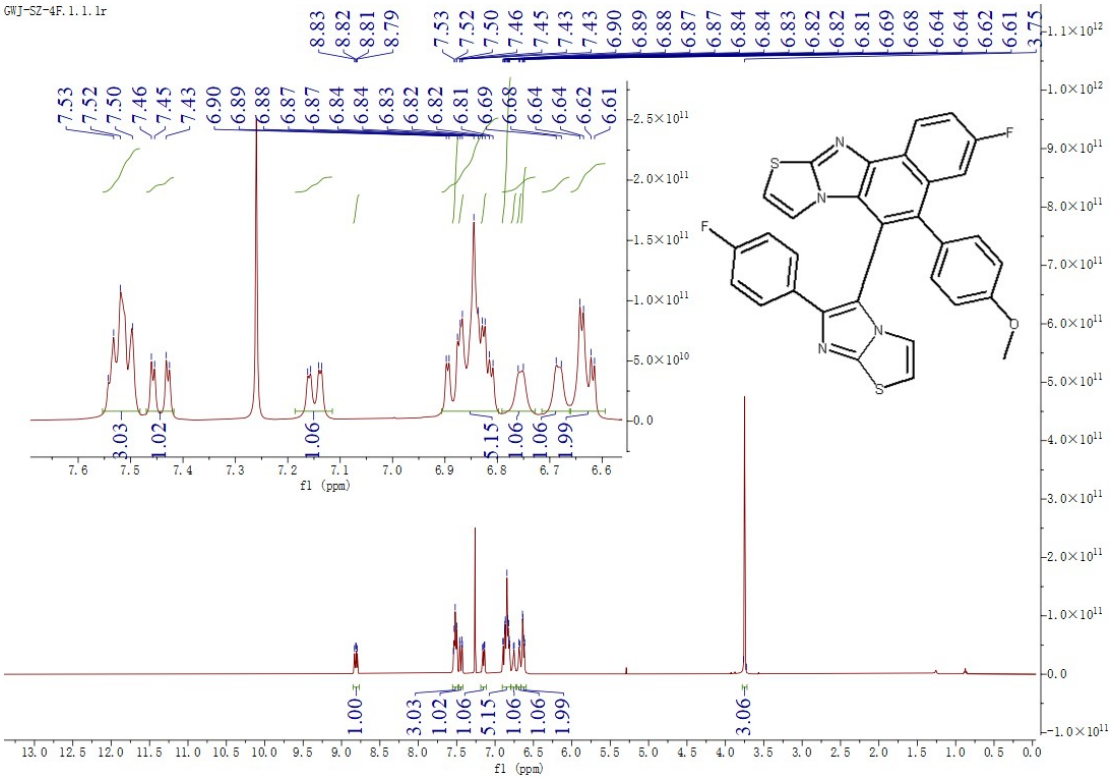


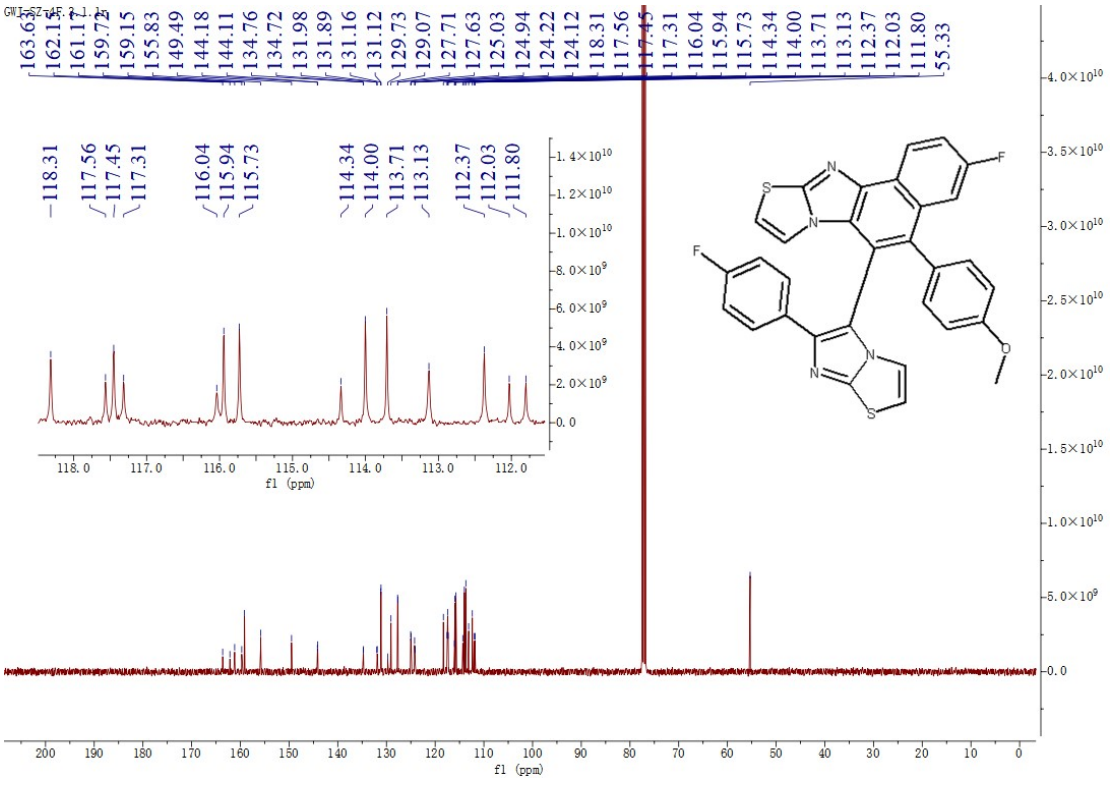
3-(*tert*-butyl)-6-(6-(4-(*tert*-butyl)phenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**6**):

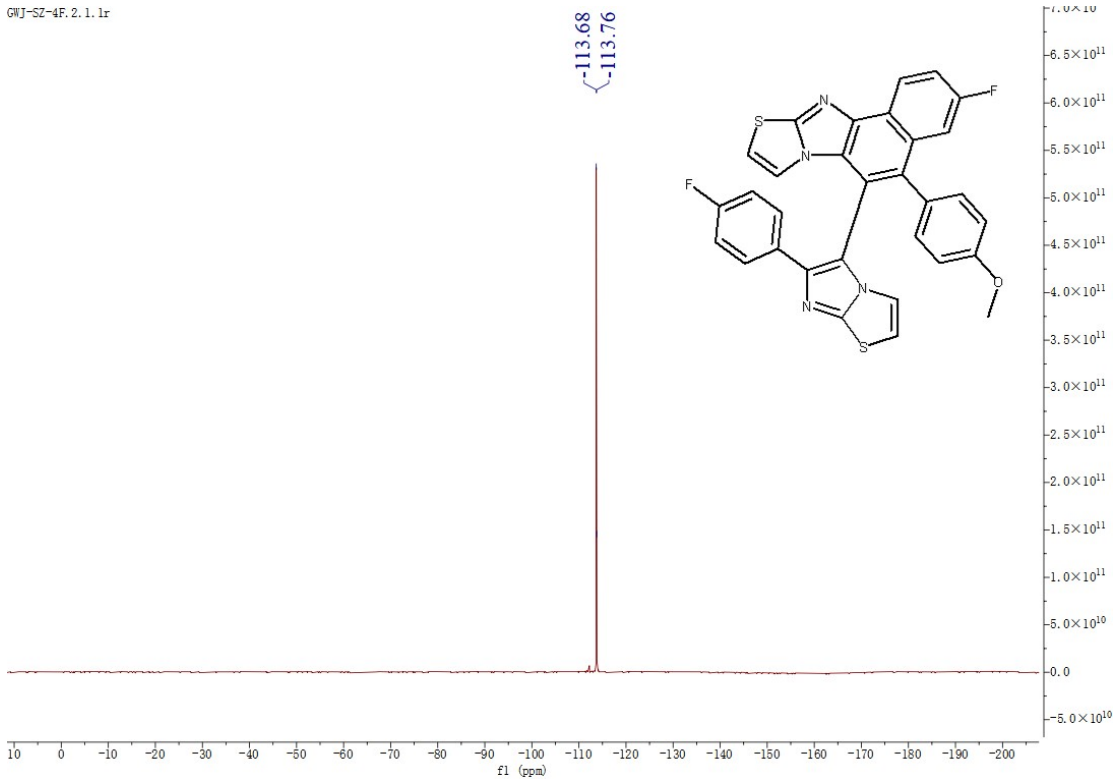




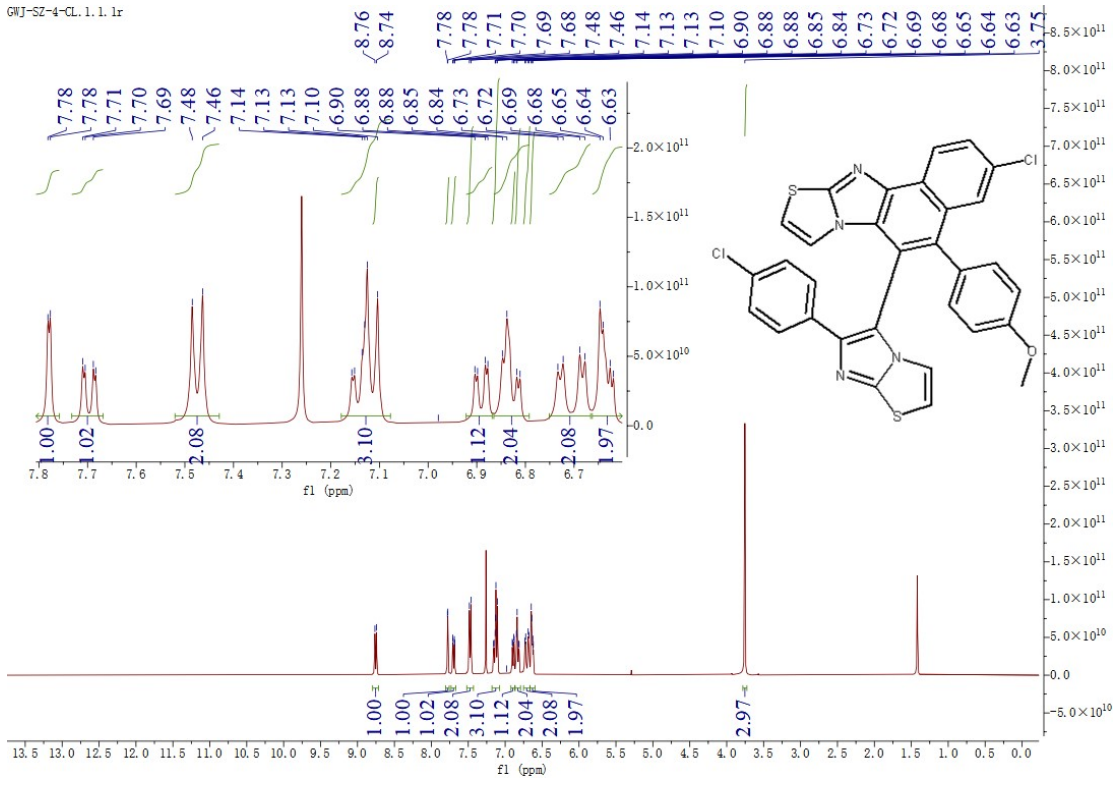
3-fluoro-6-(6-(4-fluorophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**7**):

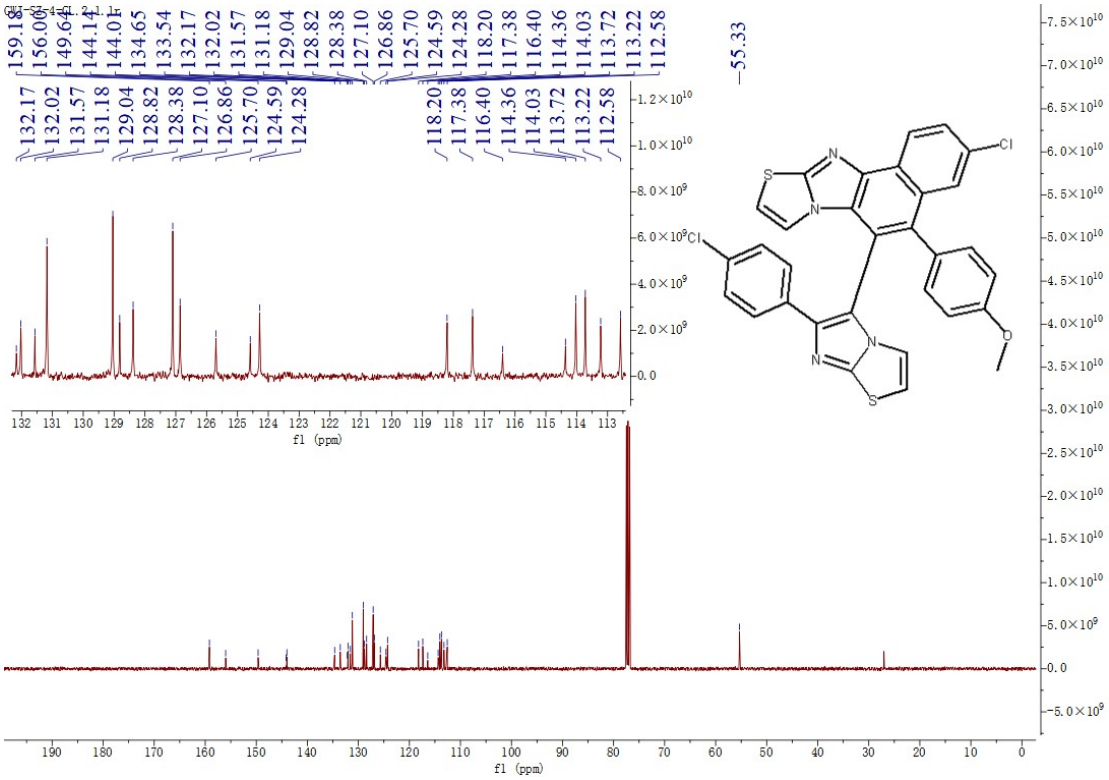




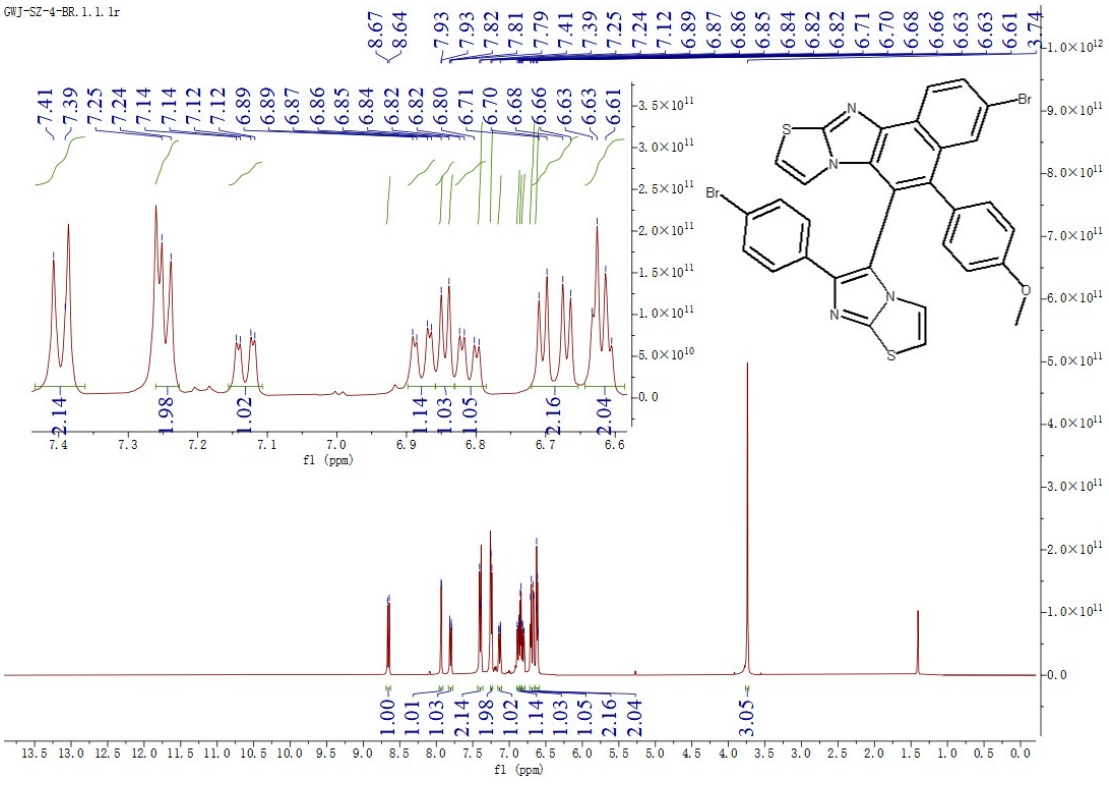


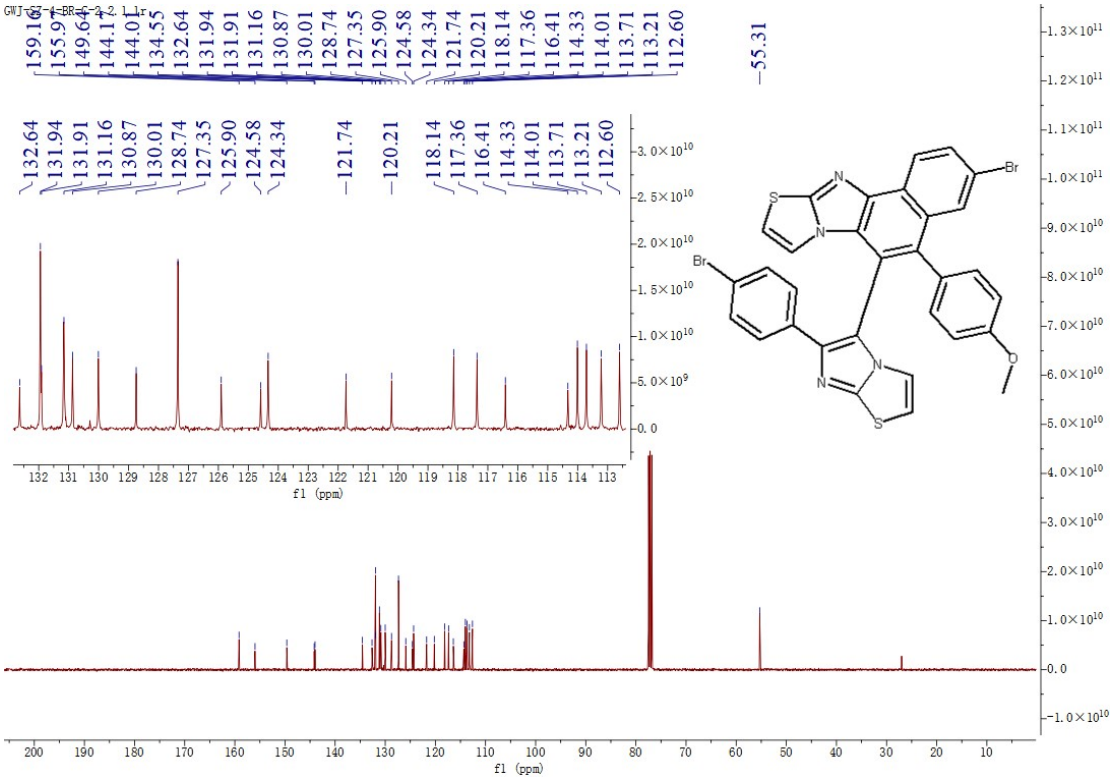
3-chloro-6-(6-(4-chlorophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**8**):



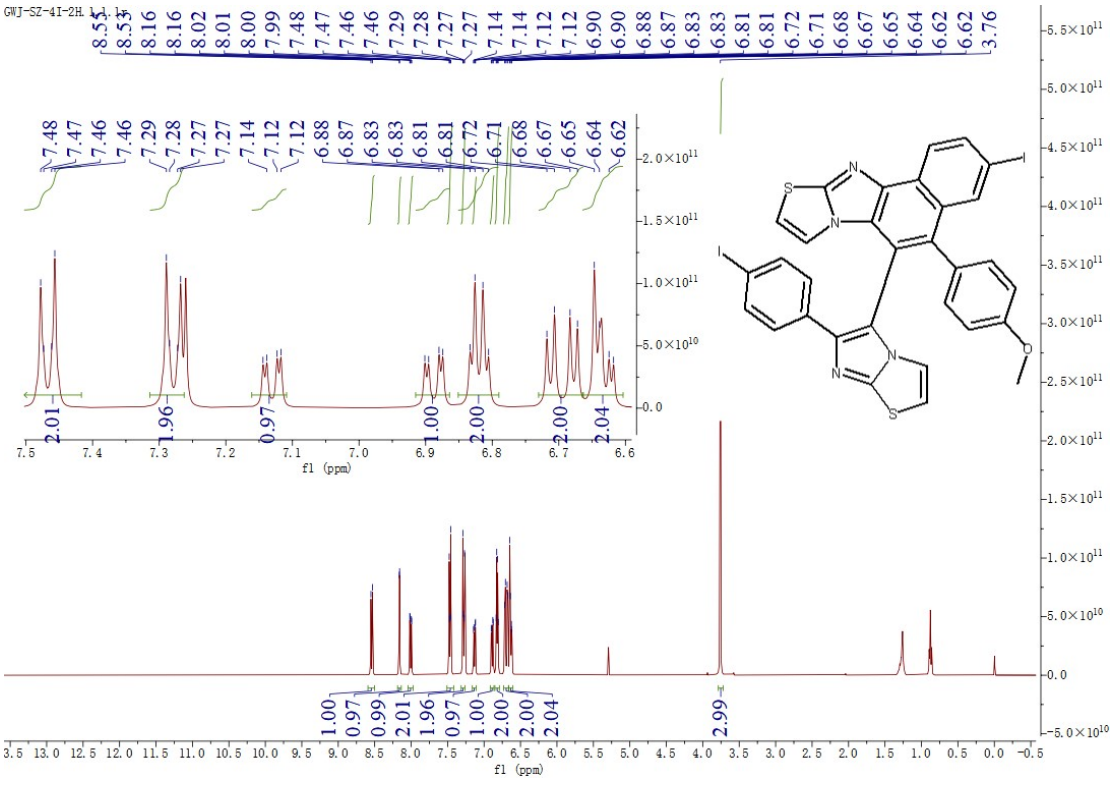


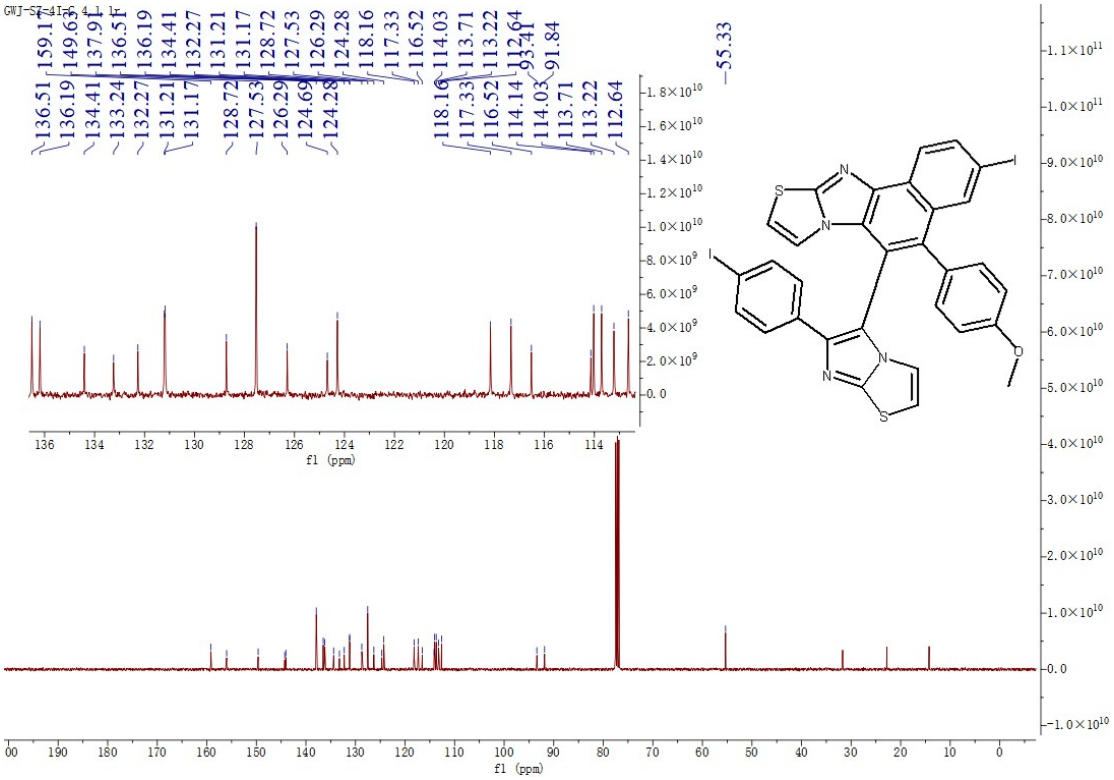
3-bromo-6-(6-(4-bromophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**9**):



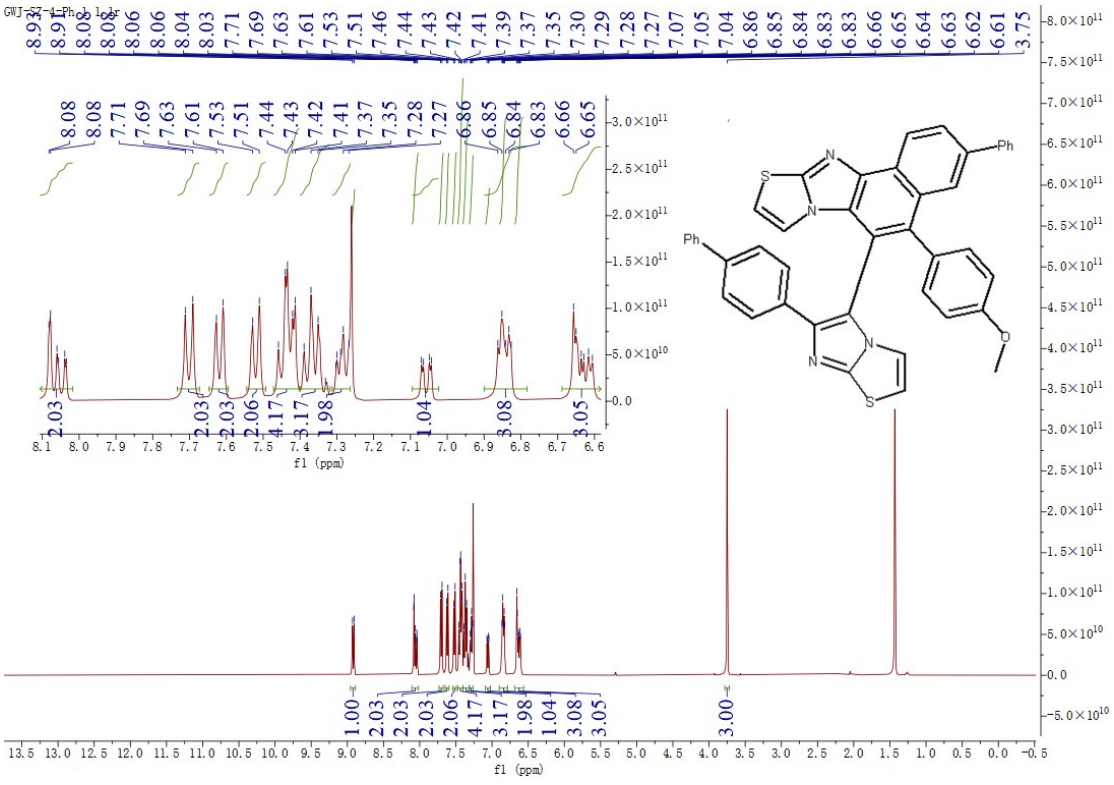


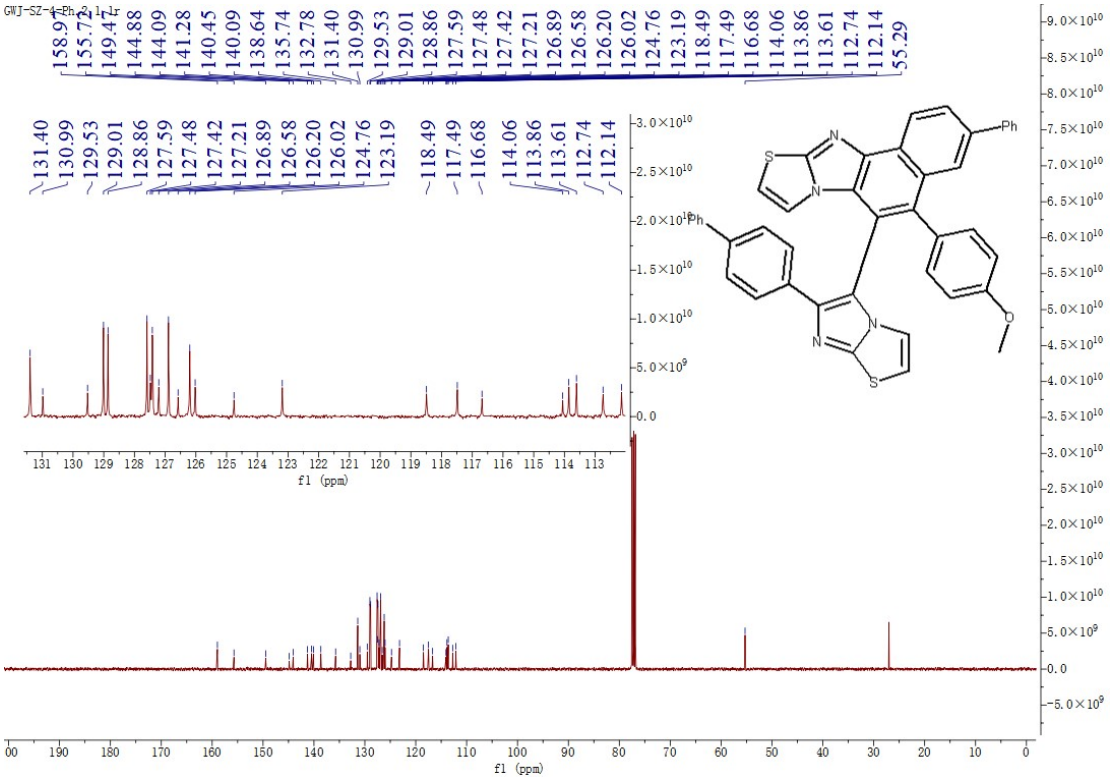
3-iodo-6-(6-(4-iodophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**10**):



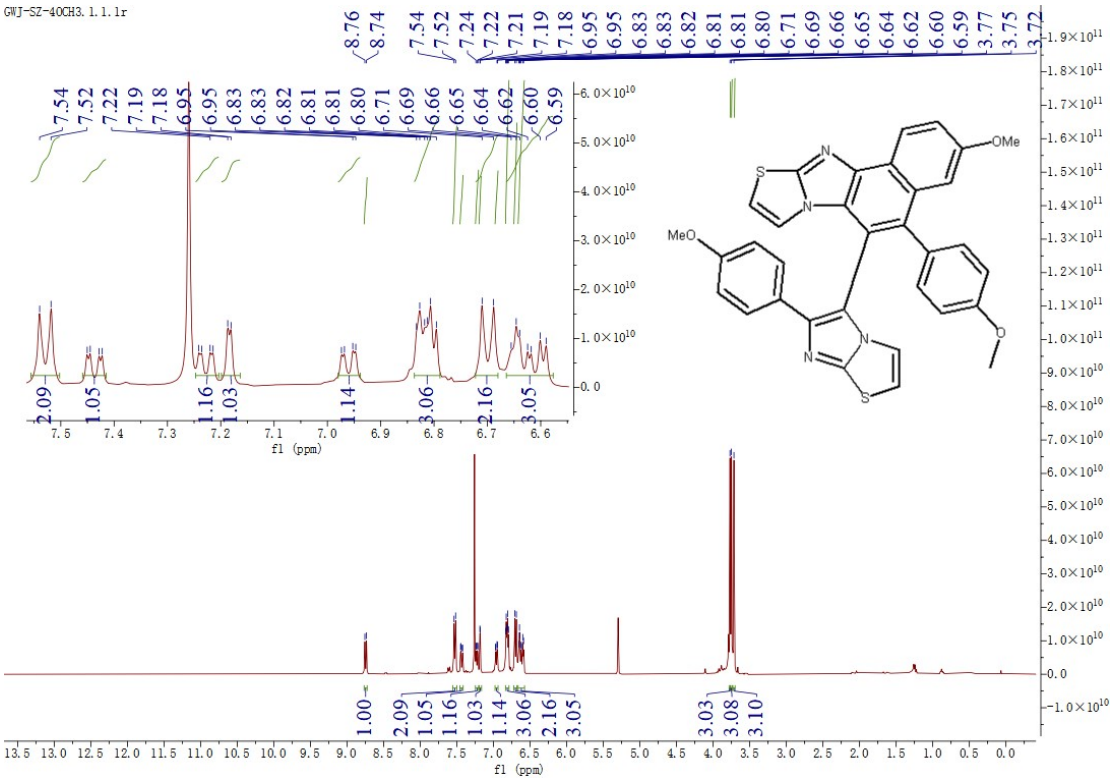


6-(6-([1,1'-biphenyl]-4-yl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)-3-phenylnaphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**11**):



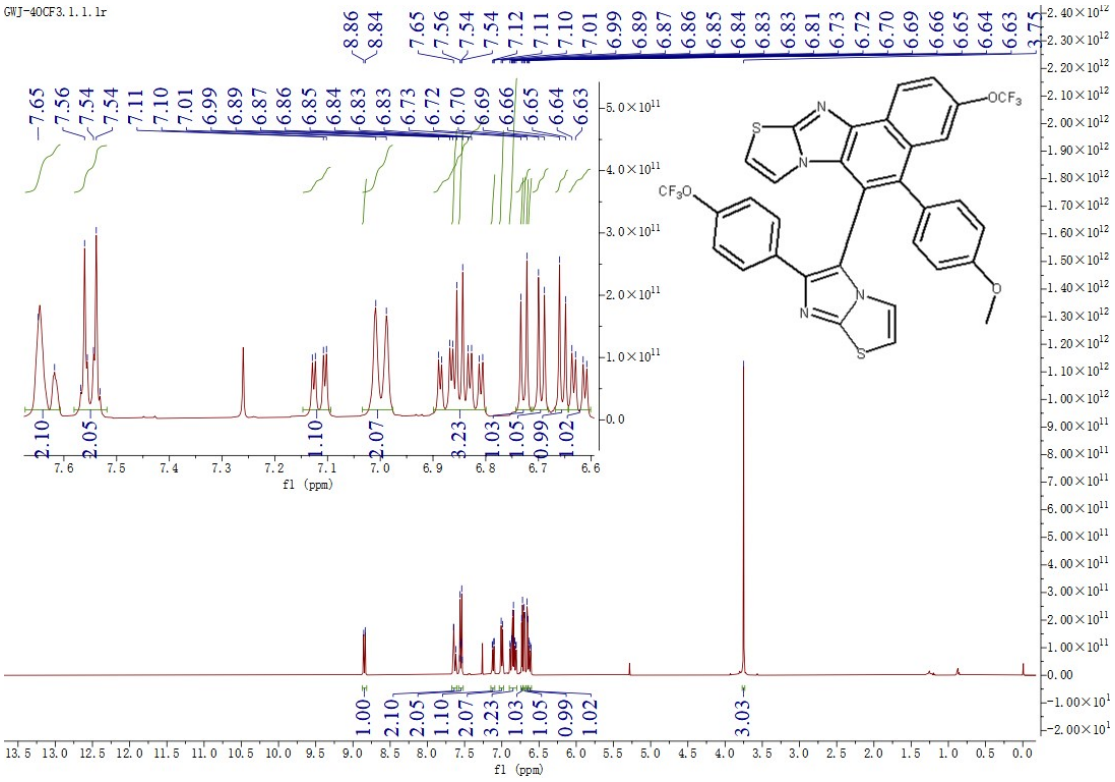


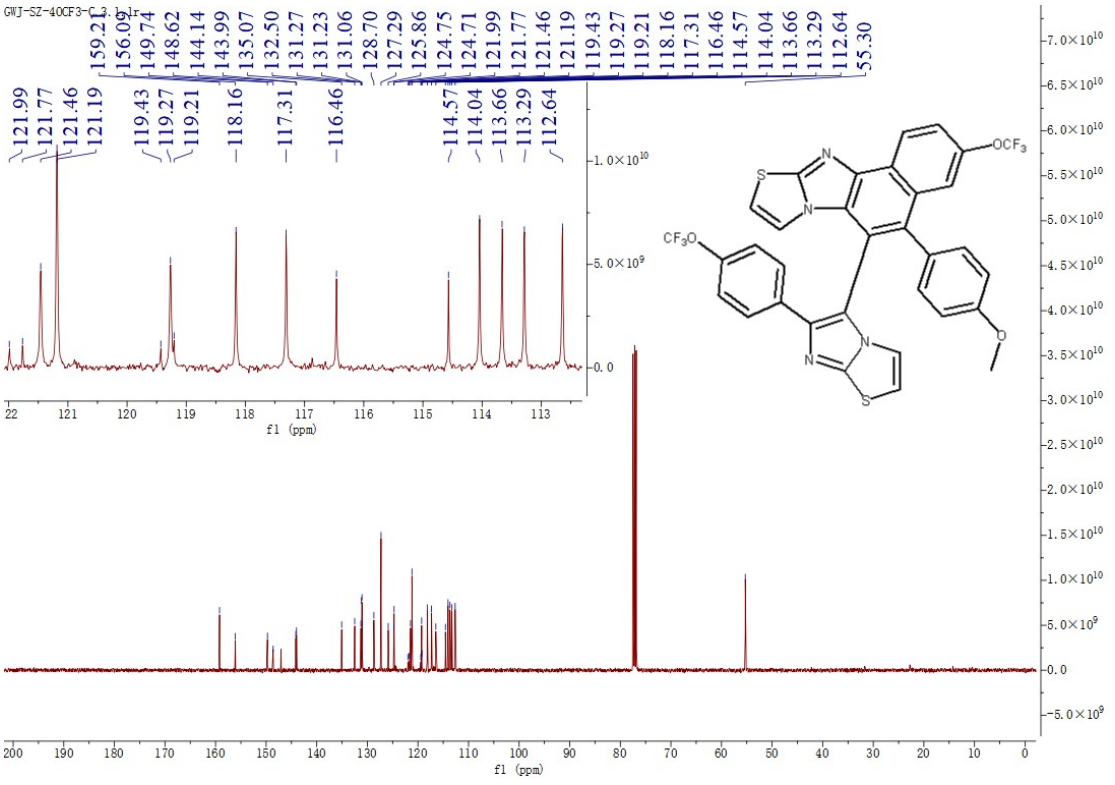
3-methoxy-5-(4-methoxyphenyl)-6-(6-(4-methoxyphenyl)imidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**12**):

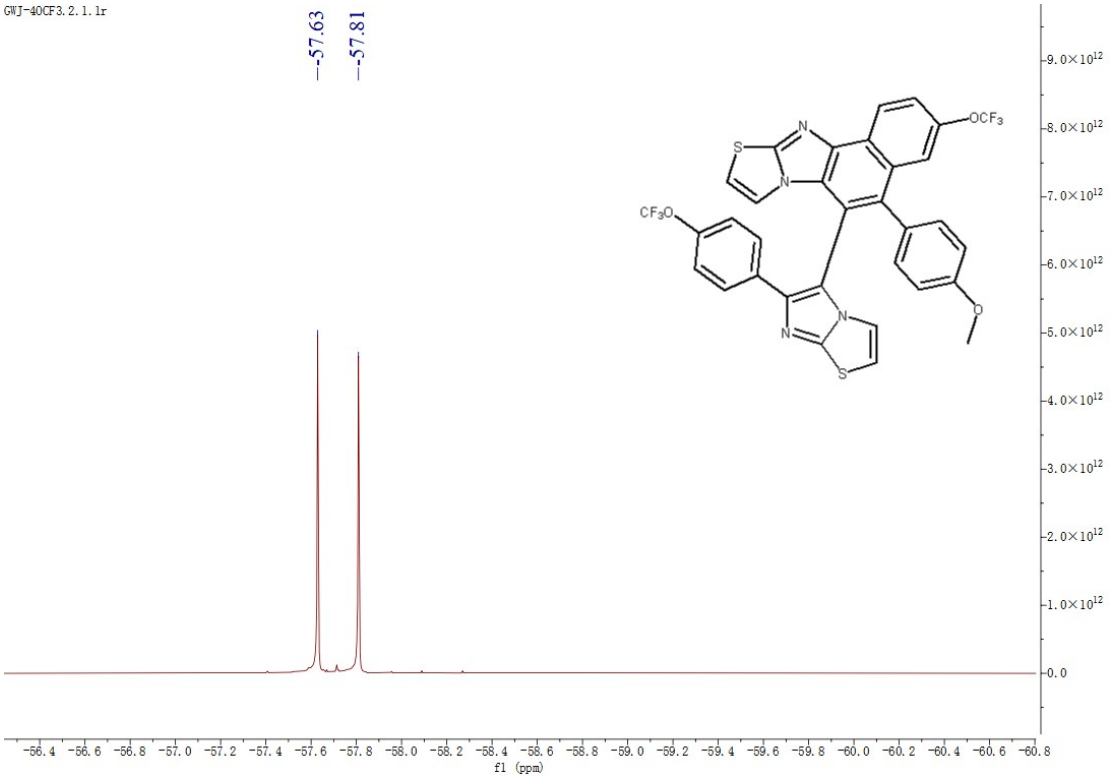


5-(4-methoxyphenyl)-3-(trifluoromethoxy)-6-(6-(4-

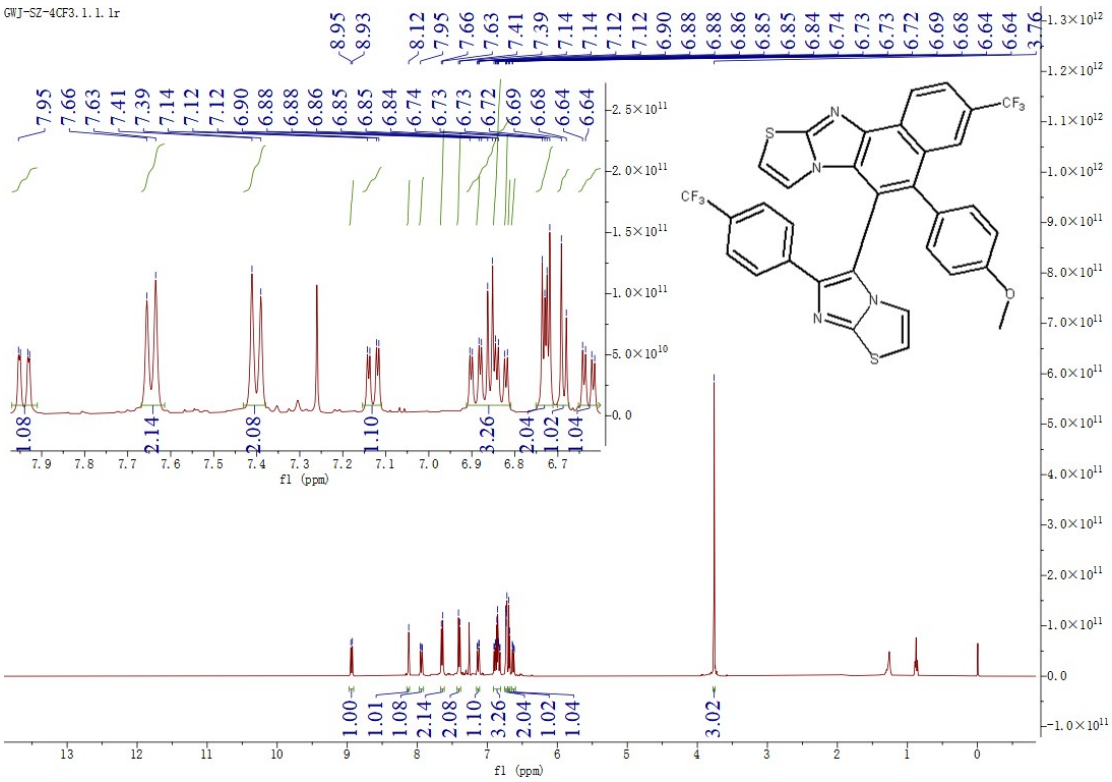
(trifluoromethoxy)phenyl)imidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**13**):

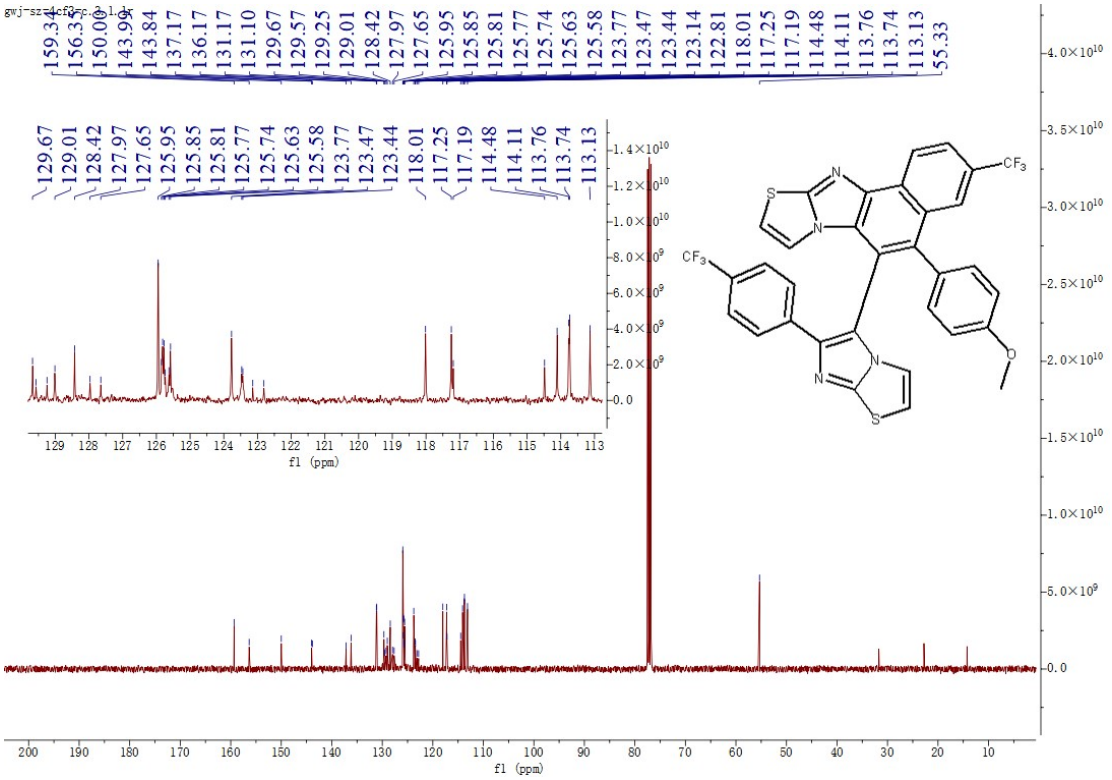




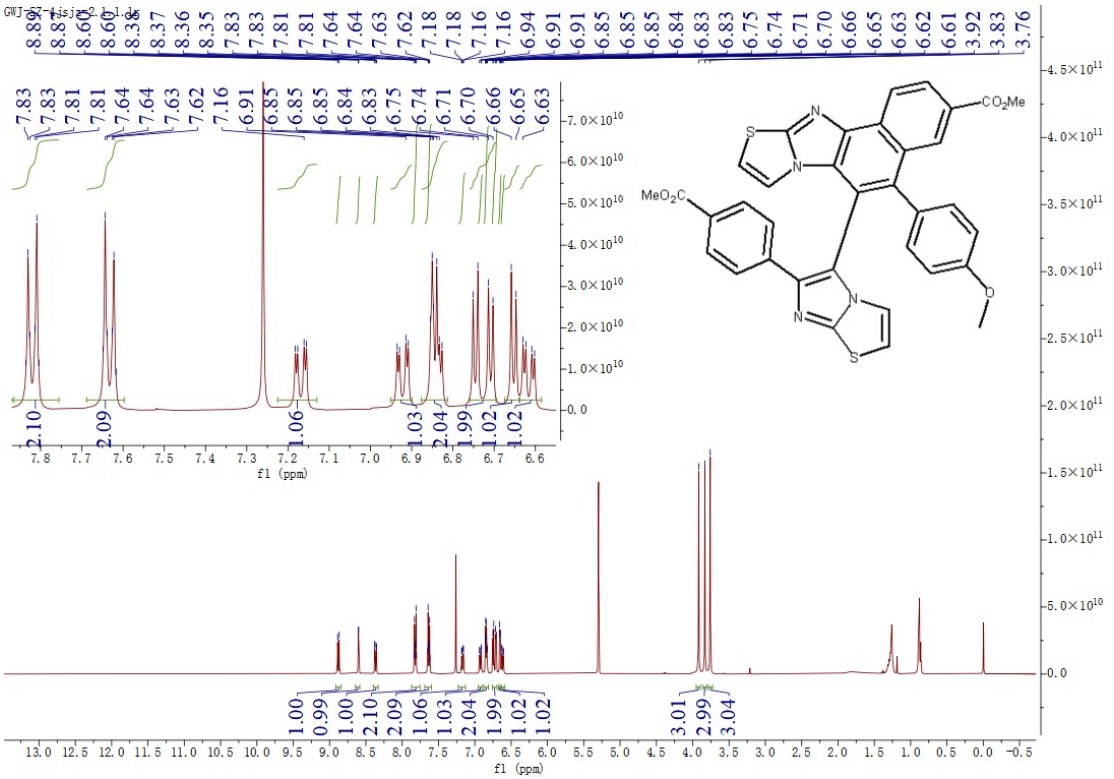


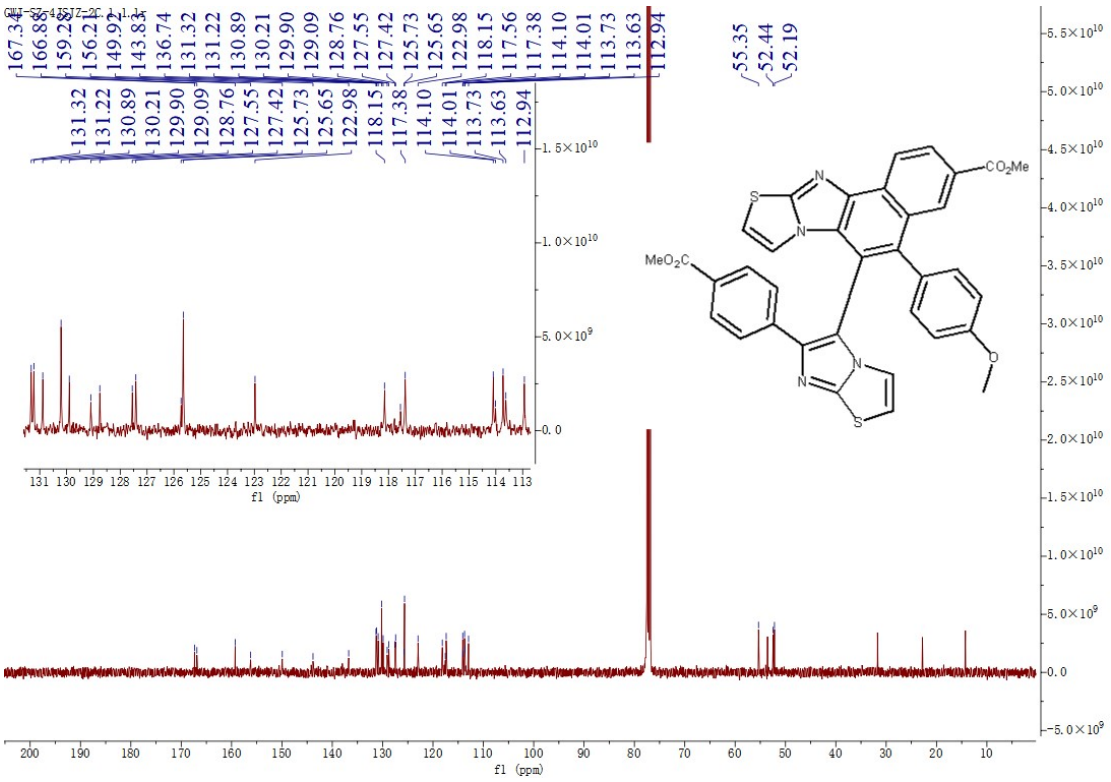
5-(4-methoxyphenyl)-3-(trifluoromethyl)-6-(6-(4-(trifluoromethyl)phenyl)imidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**14**):


GWJ-SZ-4CF3.1.1.1r
f1 (ppm)

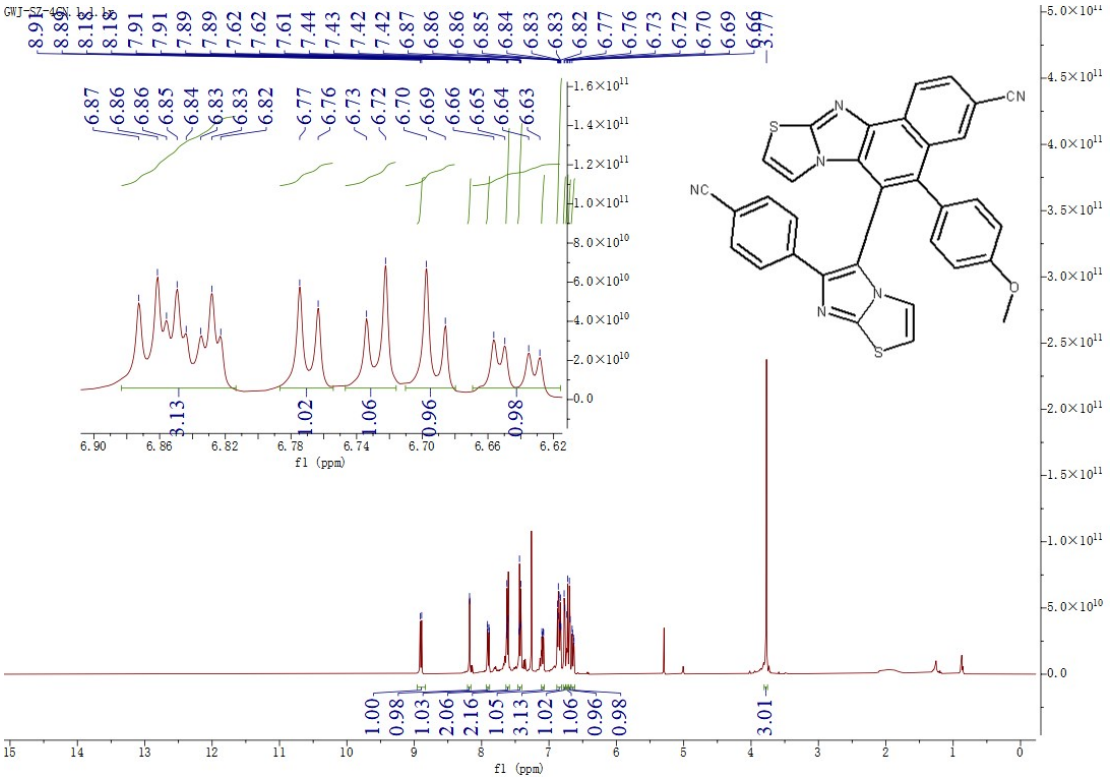

gwj-sz-4cf3c.1.1.1r
f1 (ppm)

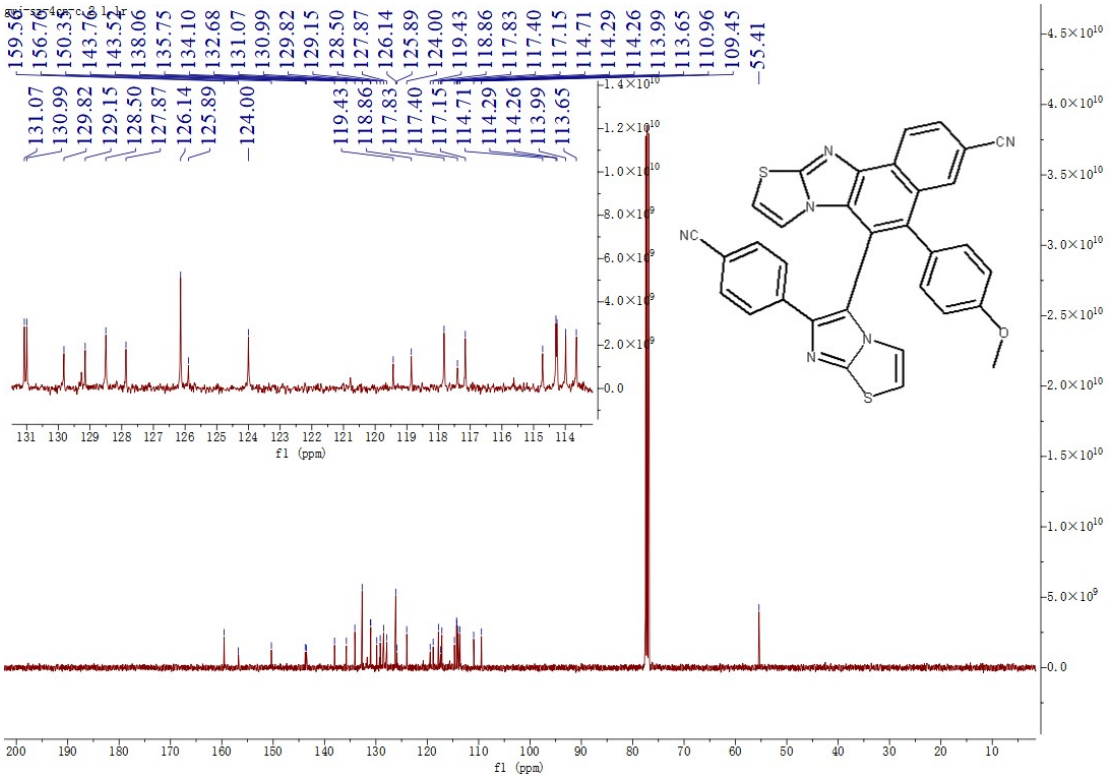
Methyl6-(6-(4-(methoxycarbonyl)phenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole-3-carboxylate (**15**):



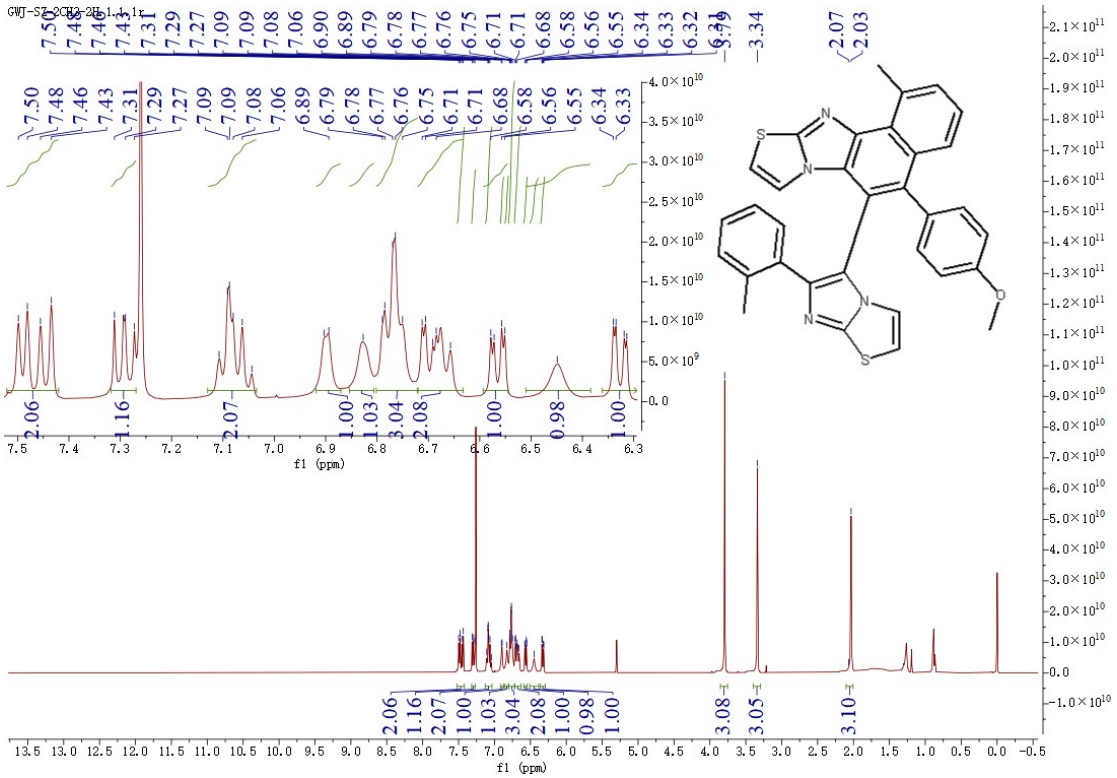


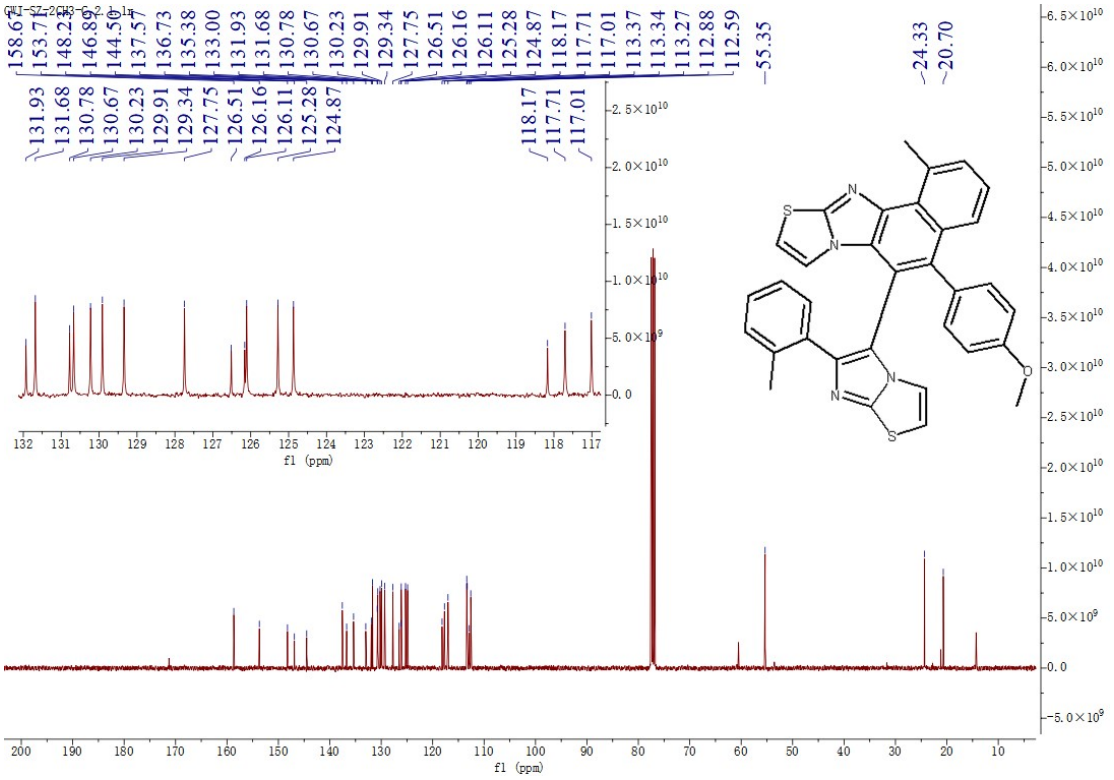
6-(6-(4-cyanophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole-3-carbonitrile (**16**):



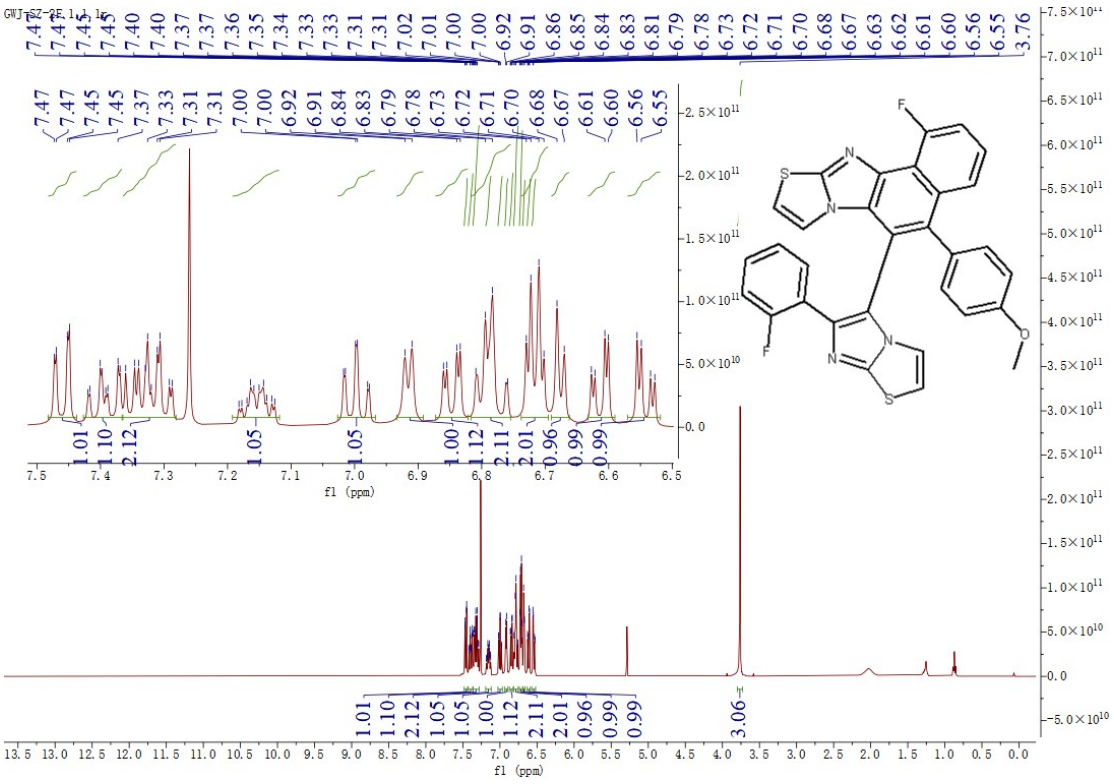


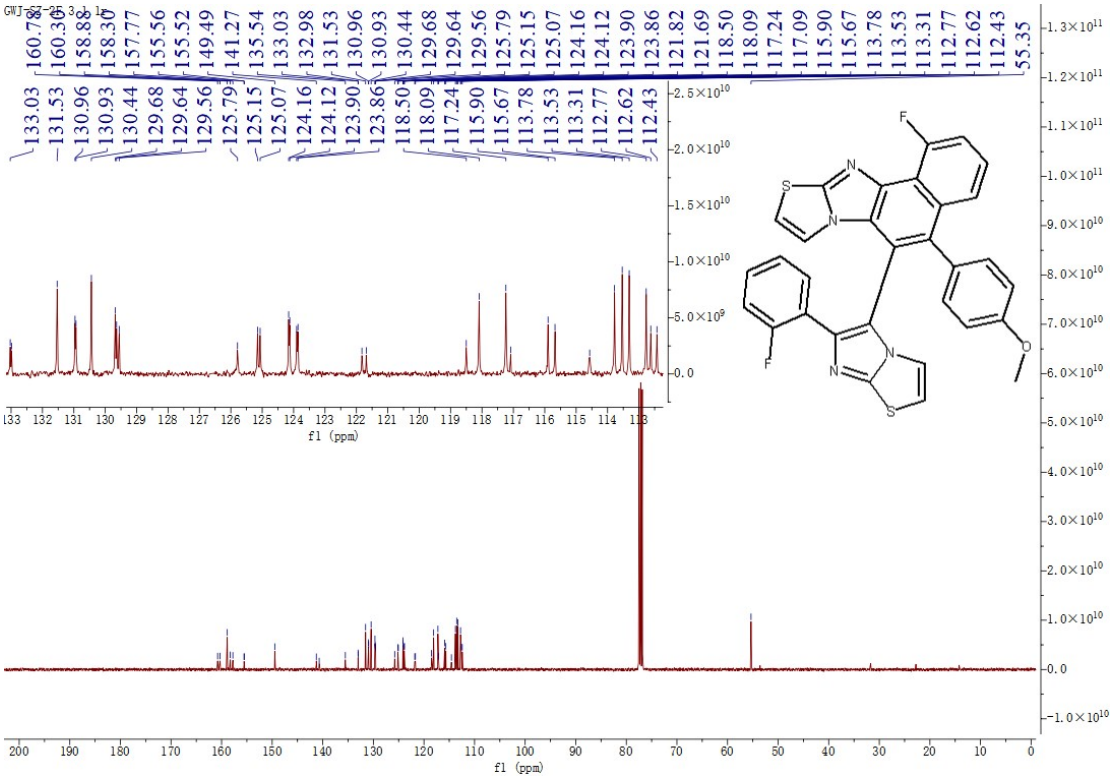
5-(4-methoxyphenyl)-1-methyl-6-(6-(*o*-tolyl)imidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**17**):

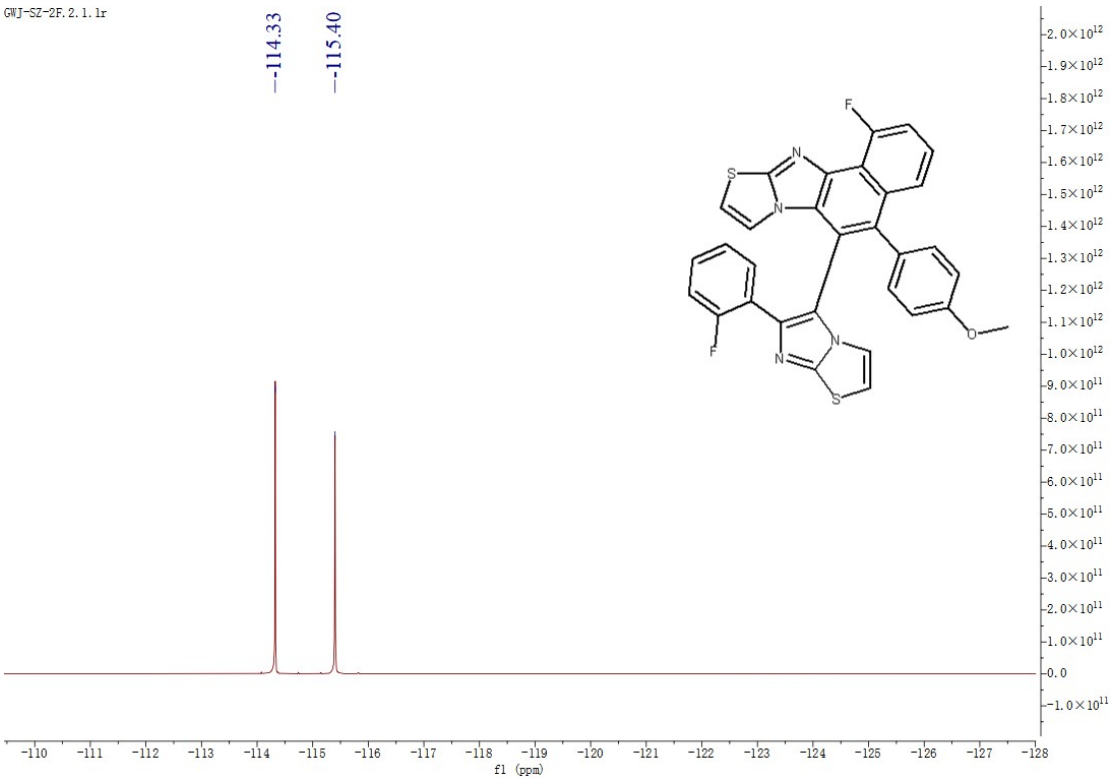




1-fluoro-6-(6-(2-fluorophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**18**):

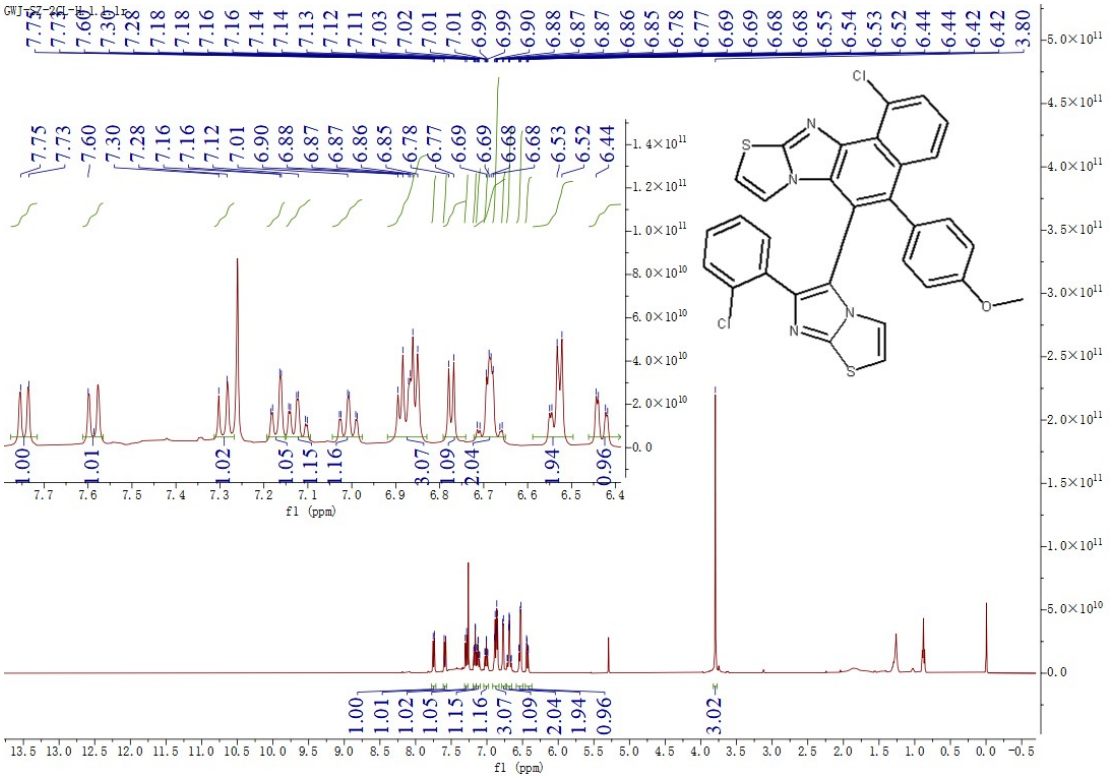


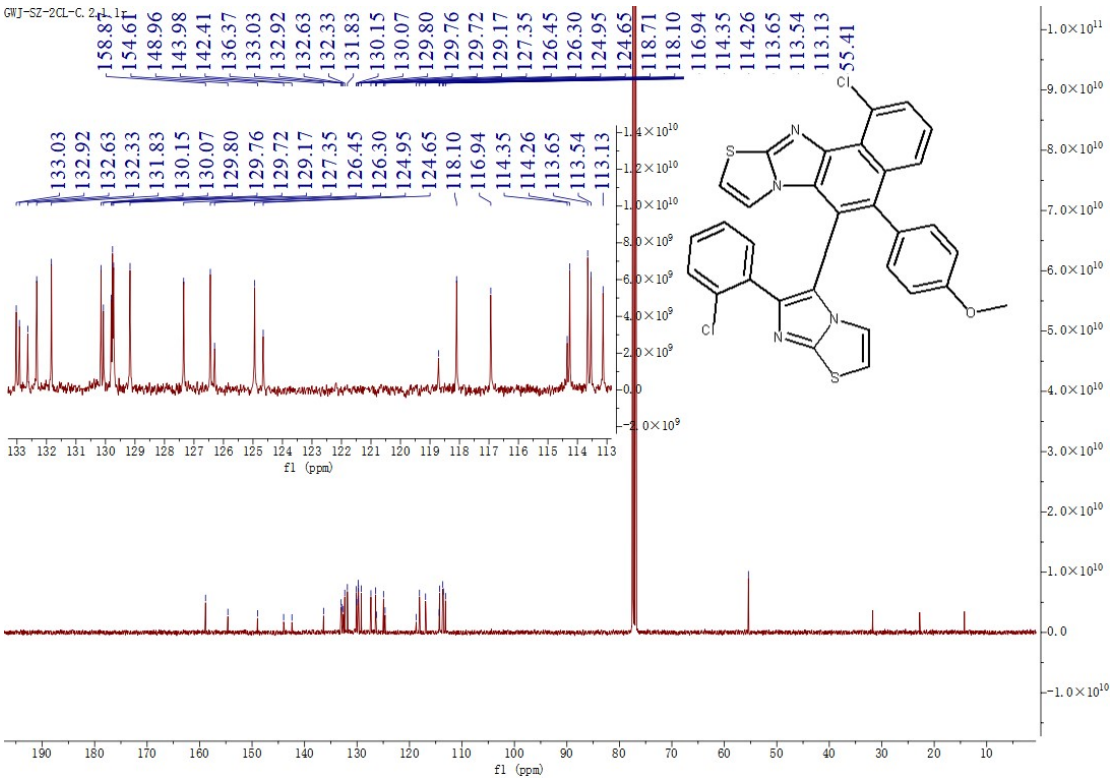




1-chloro-6-(6-(2-chlorophenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-

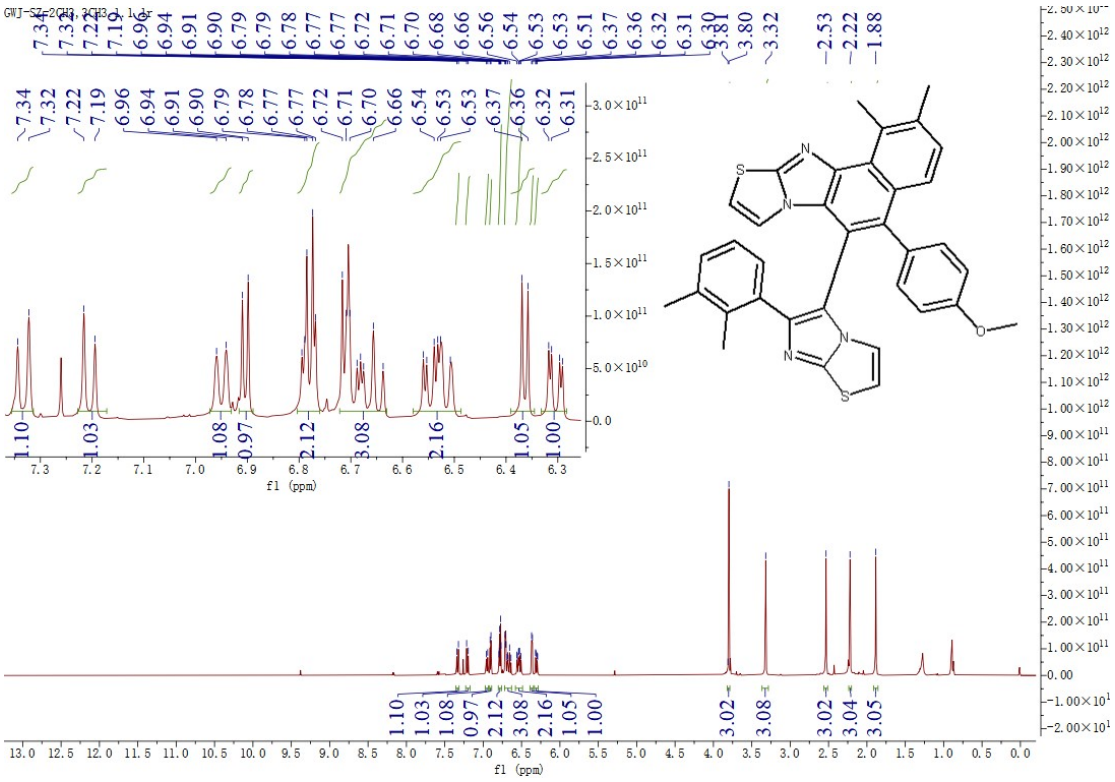
methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**19**):

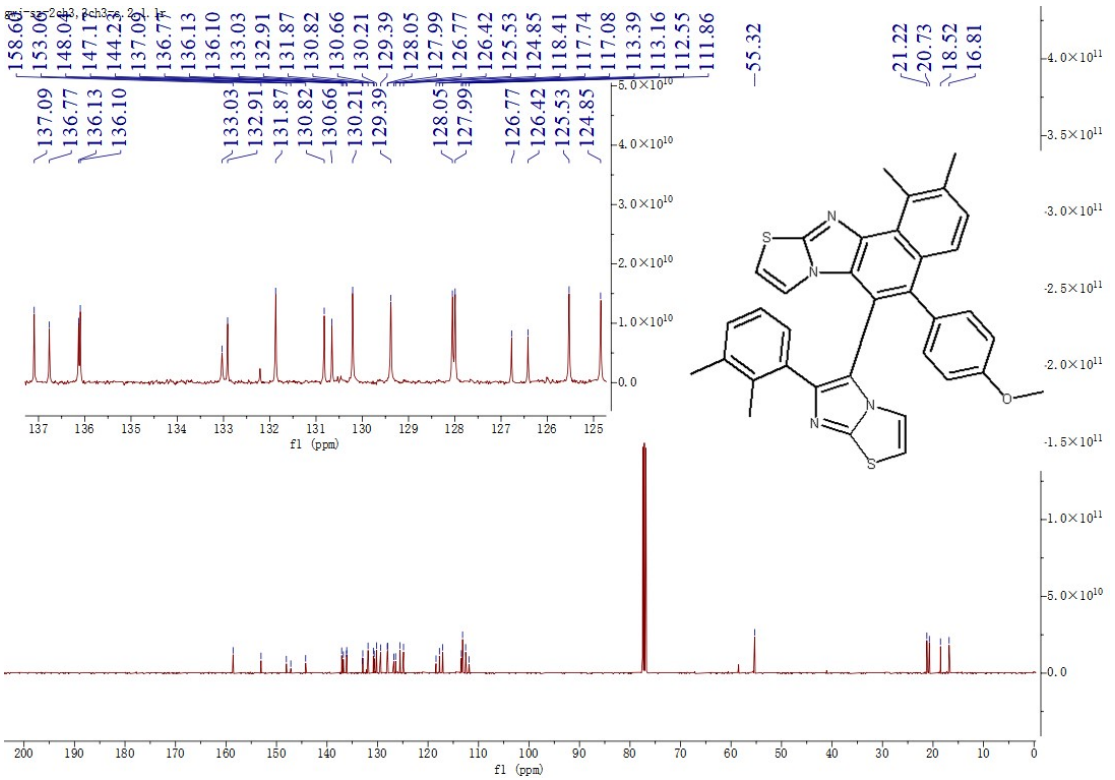




6-(6-(2,3-dimethylphenyl)imidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)-1,2-

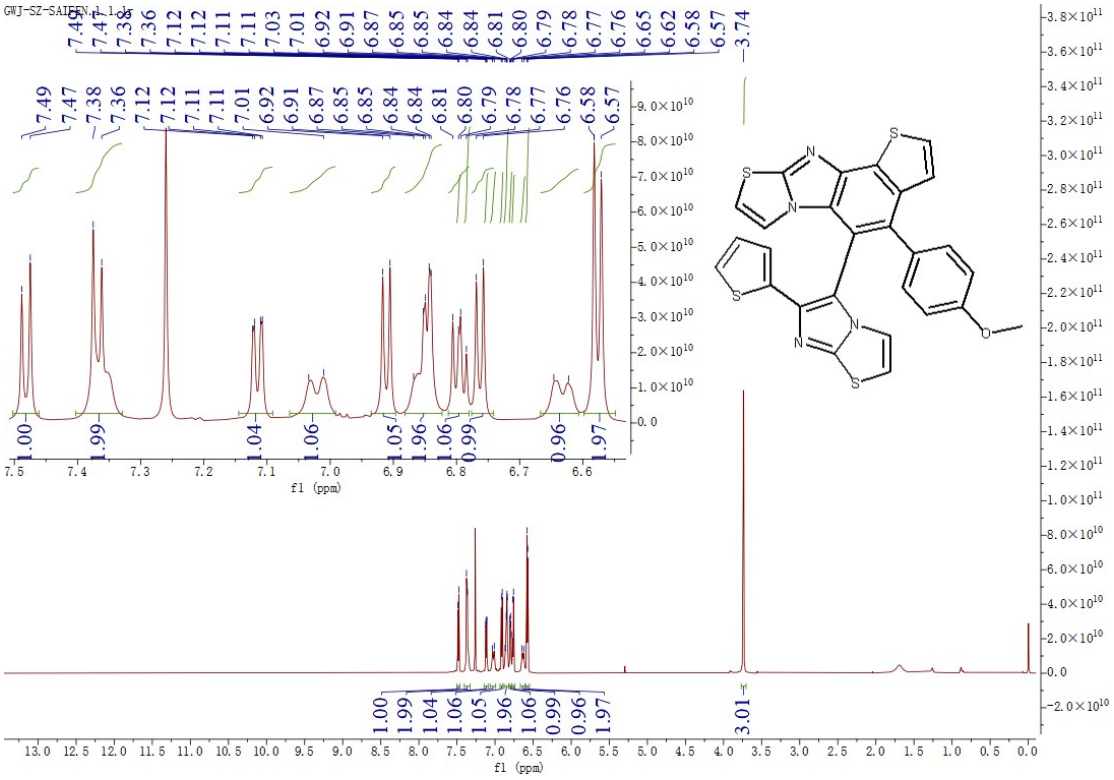
dimethylnaphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**20**):

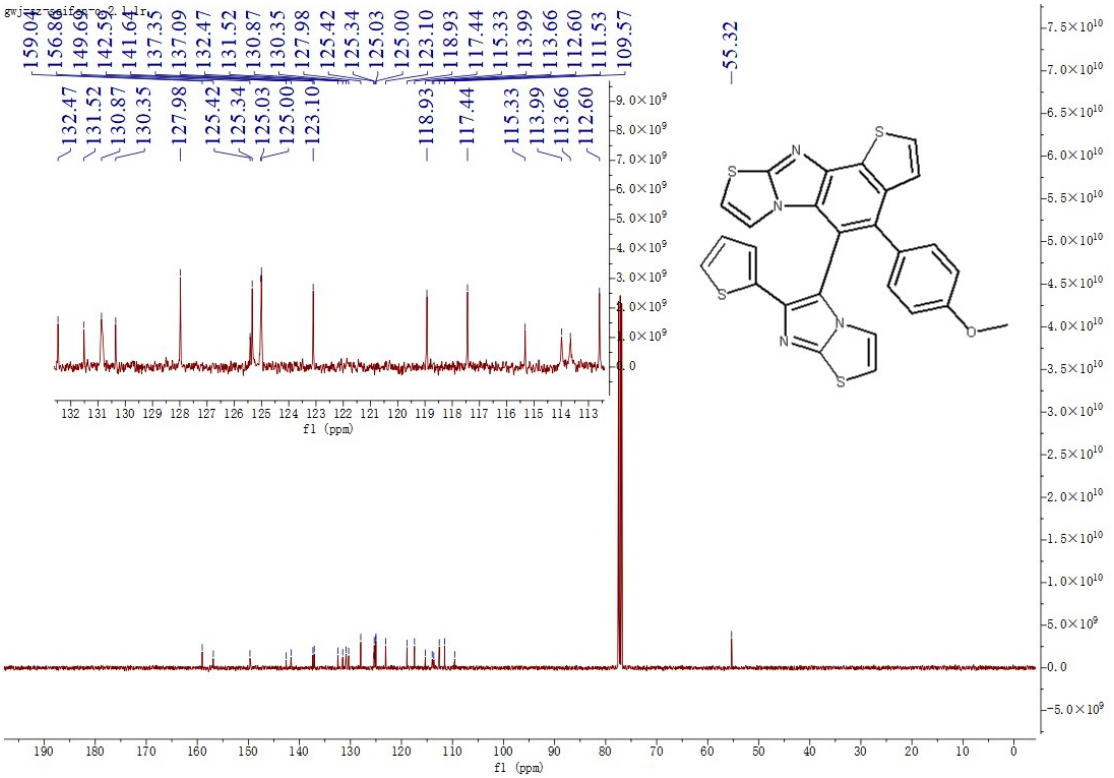




4-(4-methoxyphenyl)-5-(6-(thiophen-2-yl)imidazo[2,1-*b*]thiazol-5-

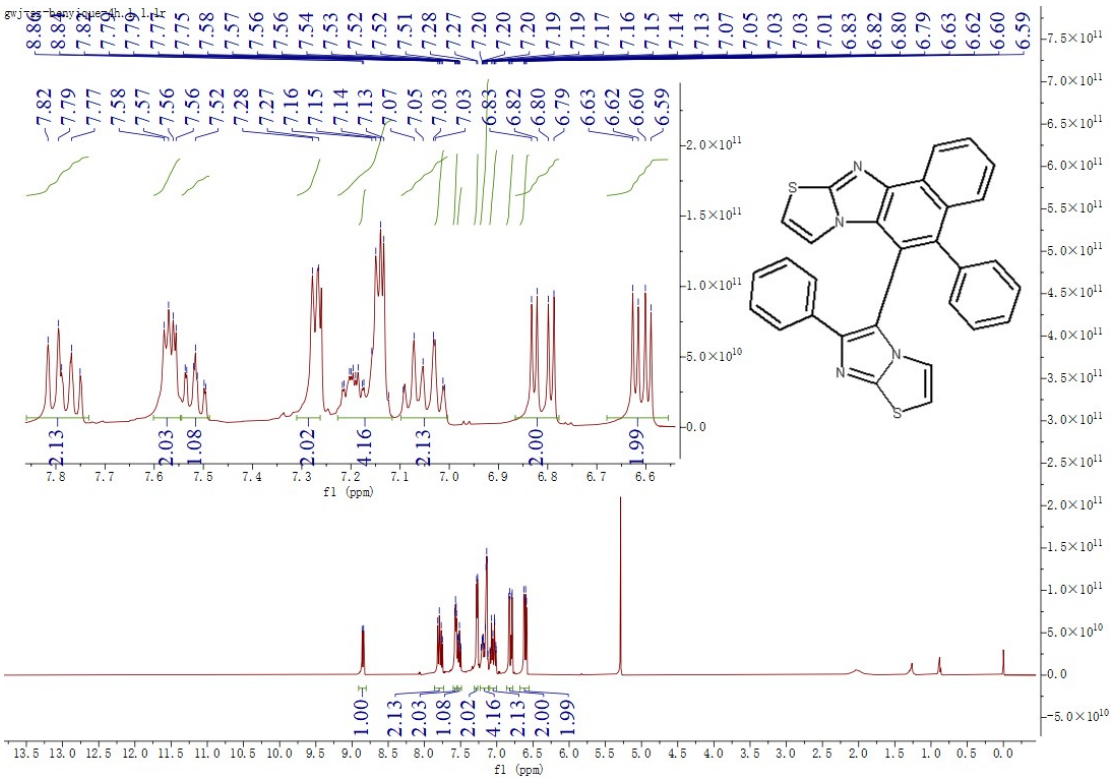
yl)thieno[3",2":5',6']benzo[1',2':4,5]imidazo[2,1-*b*]thiazole (**21**):

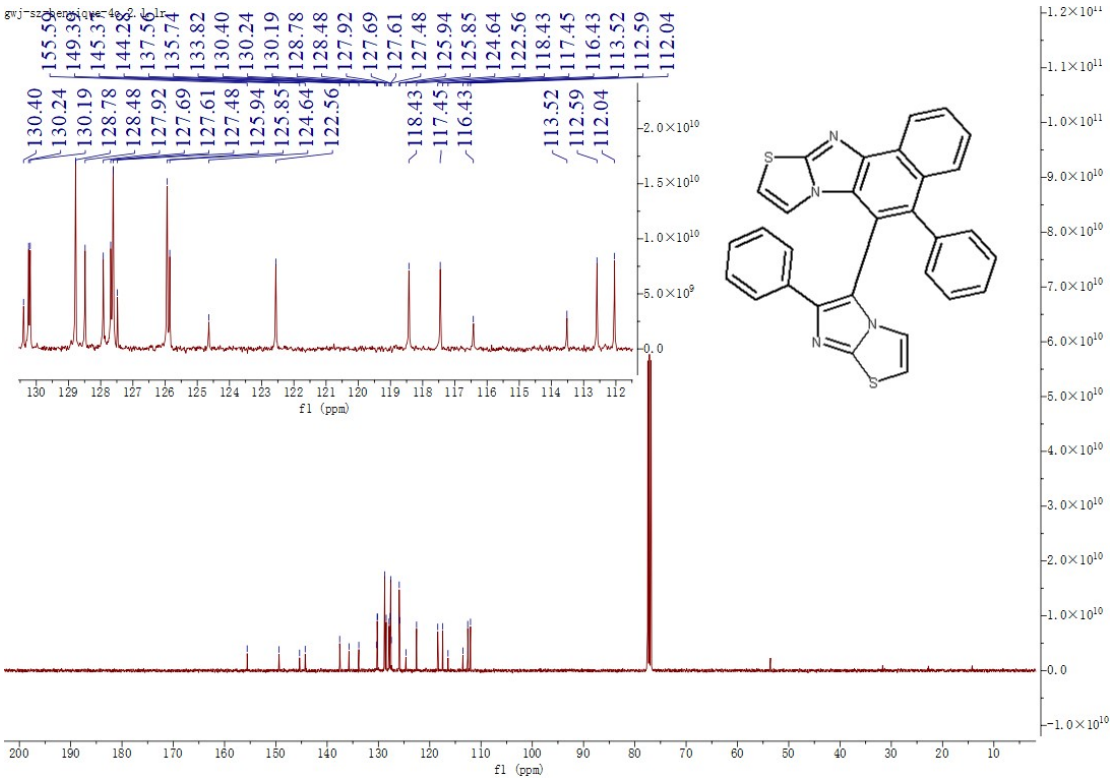




5-phenyl-6-(6-phenylimidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-

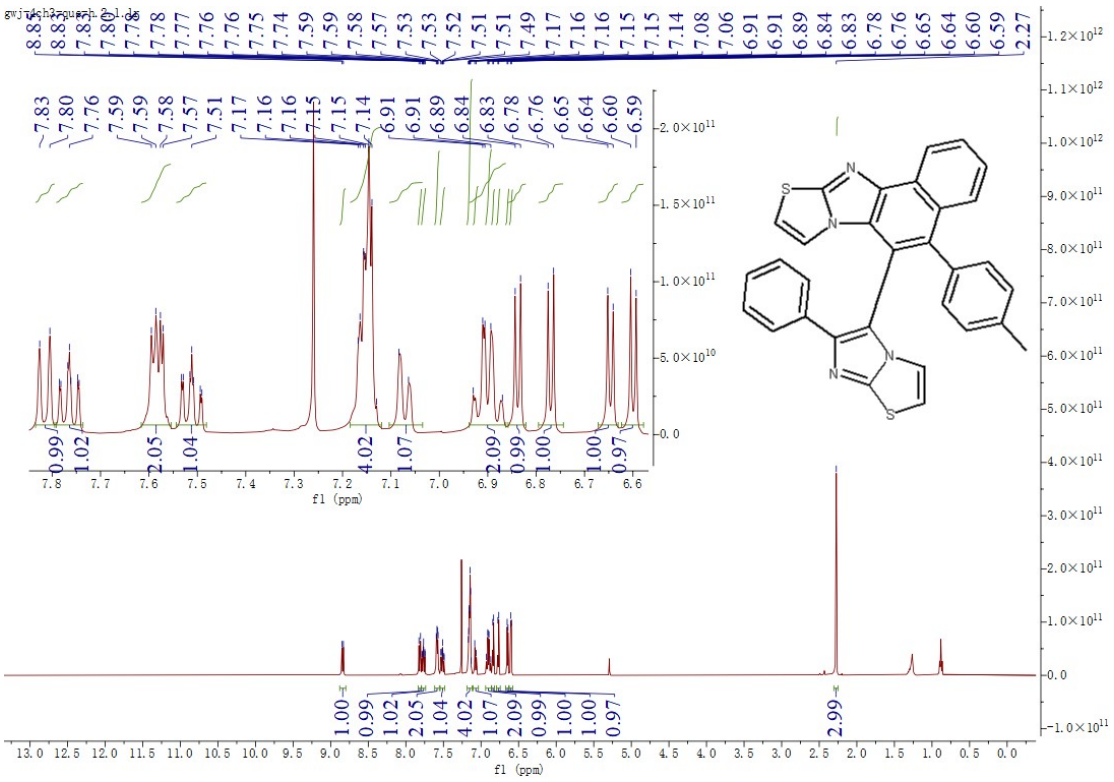
b]thiazole (**22**):

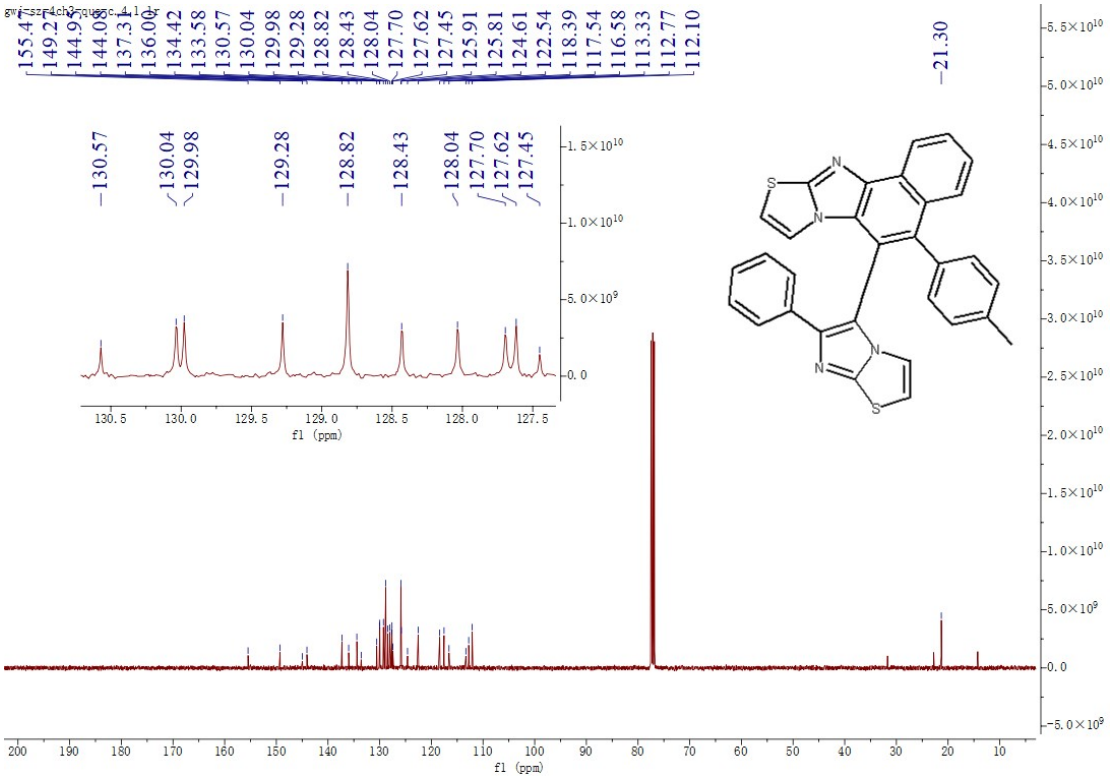




6-(6-phenylimidazo[2,1-*b*]thiazol-5-yl)-5-(*p*-tolyl)naphtho[1',2':4,5]imidazo[2,1-

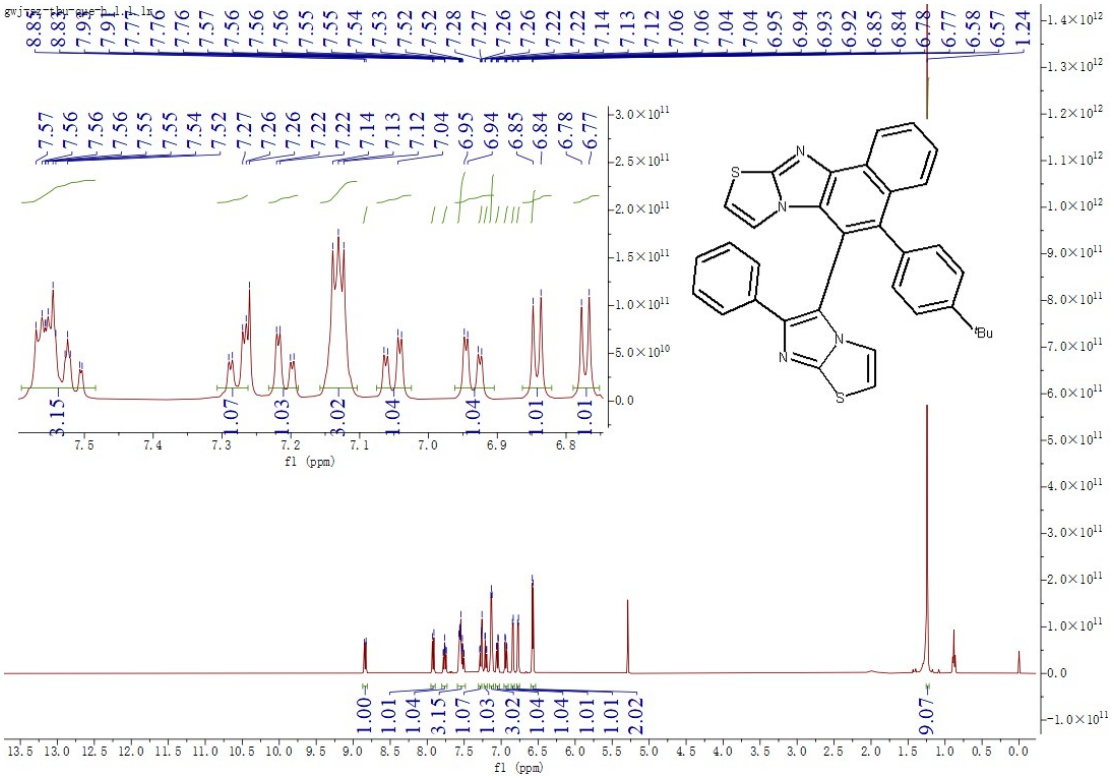
b]thiazole (**23**):

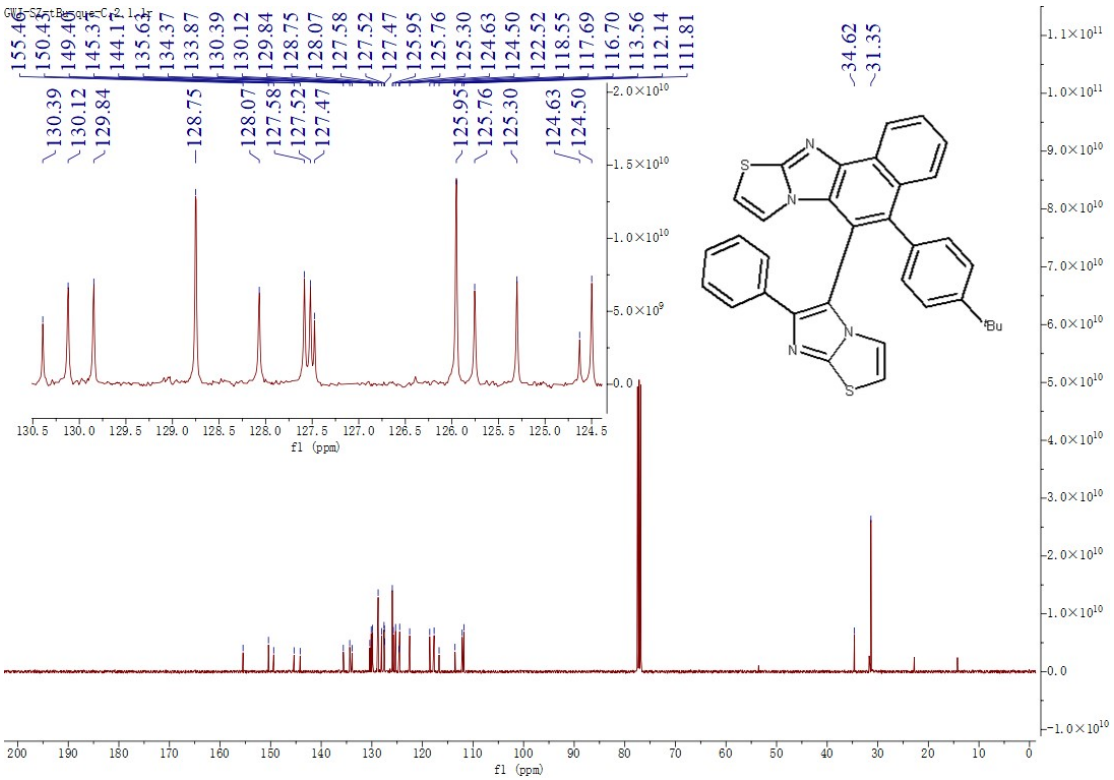




5-(4-(*tert*-butyl)phenyl)-6-(6-phenylimidazo[2,1-*b*]thiazol-5-

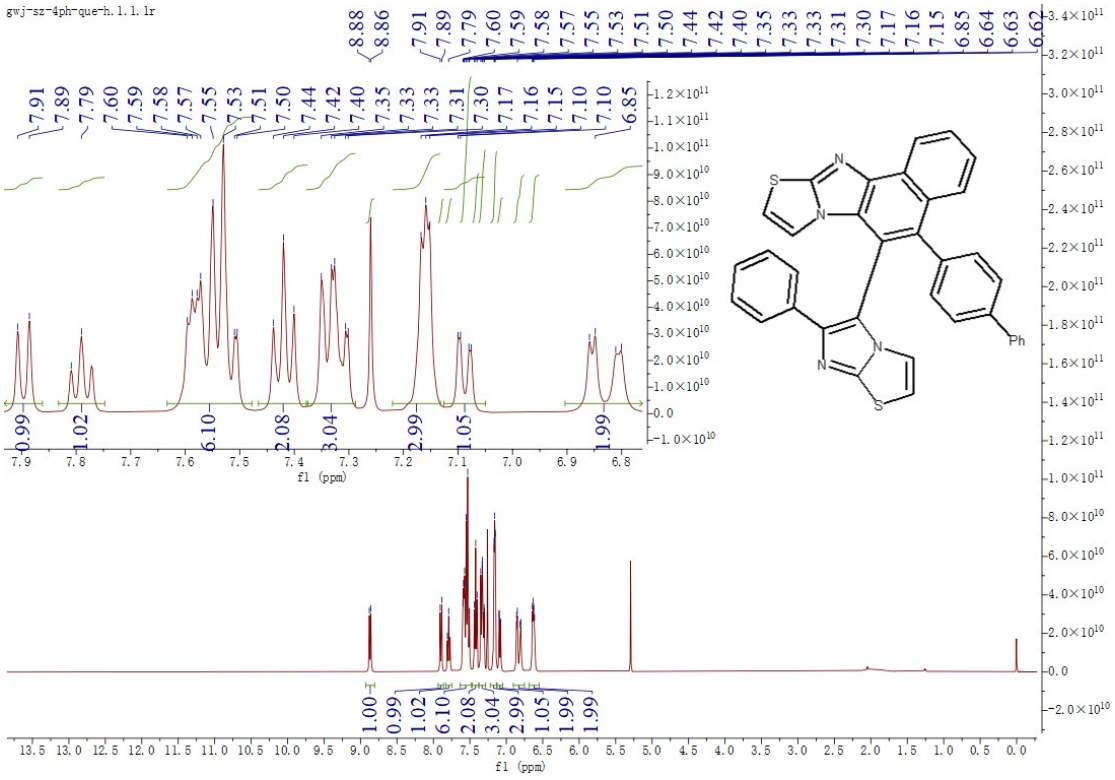
yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**24**):

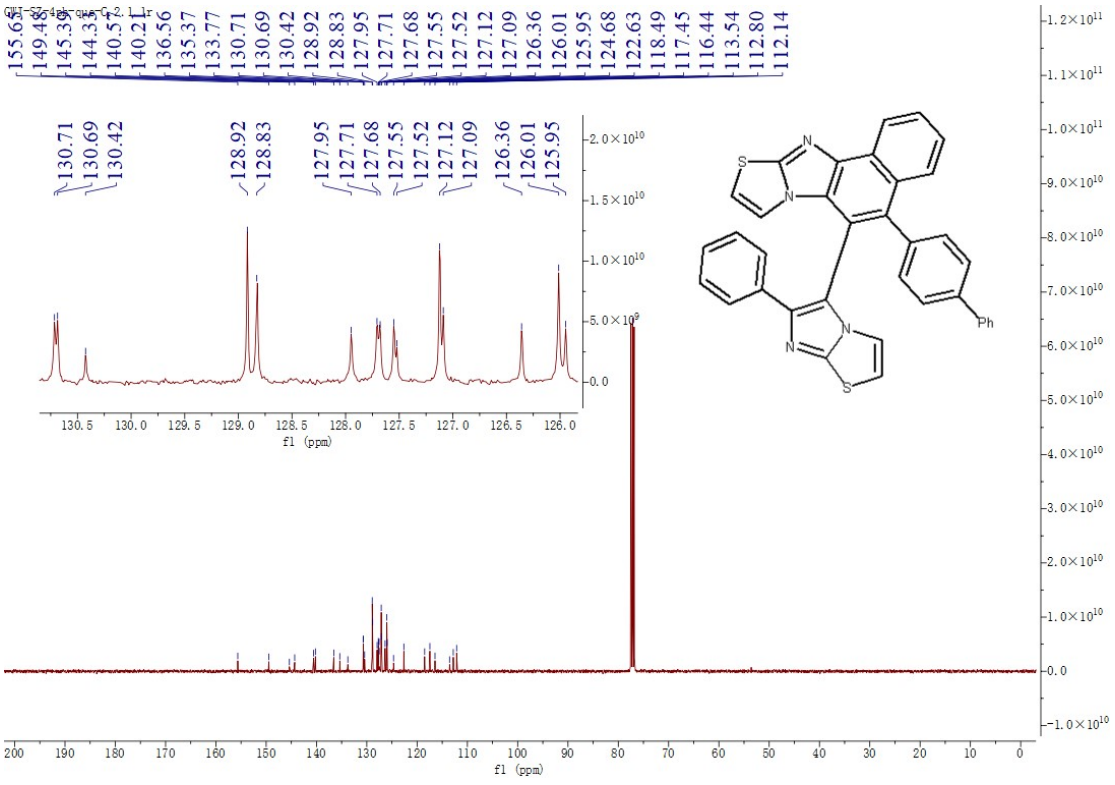




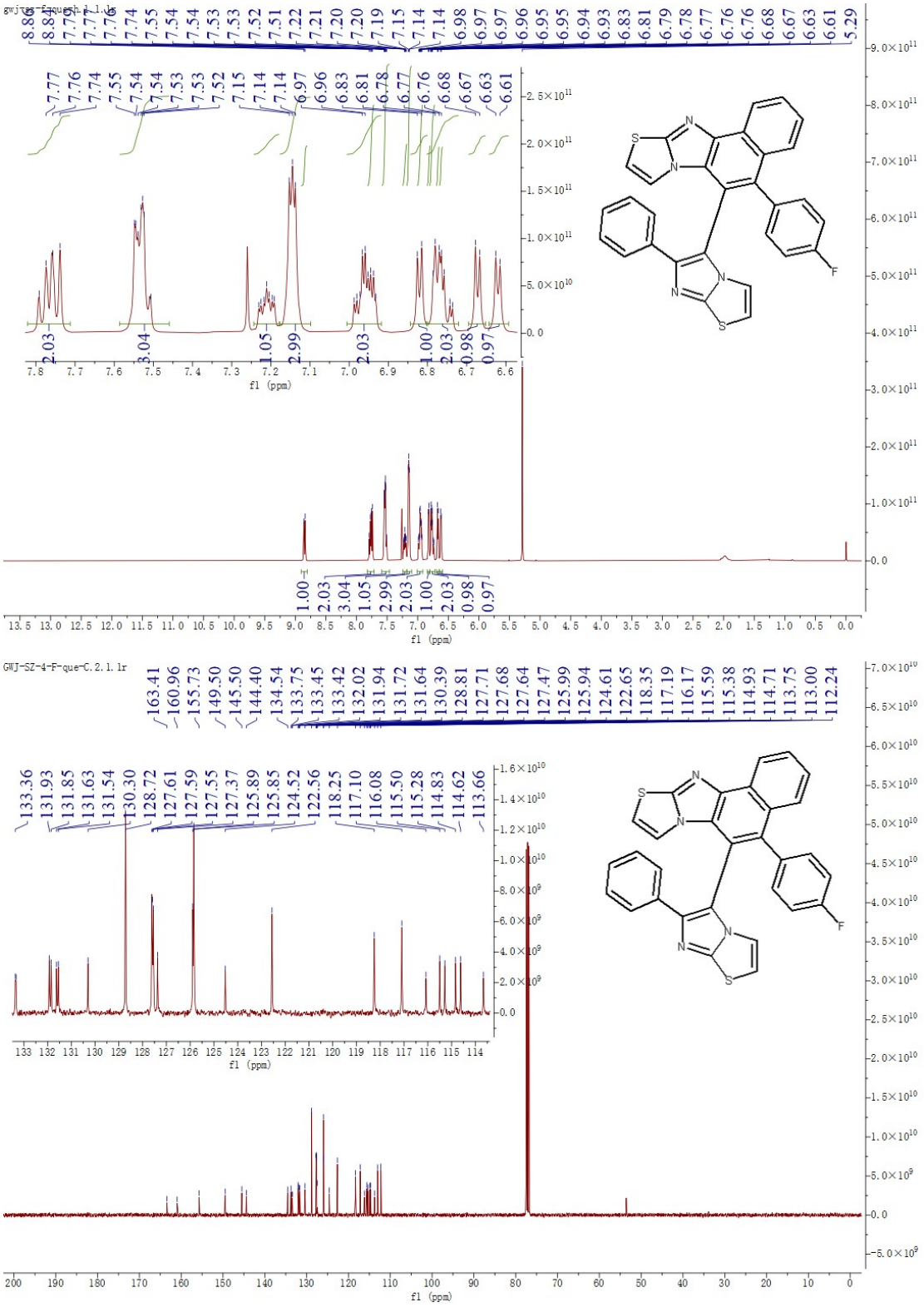
5-([1,1'-biphenyl]-4-yl)-6-(6-phenylimidazo[2,1-*b*]thiazol-5-

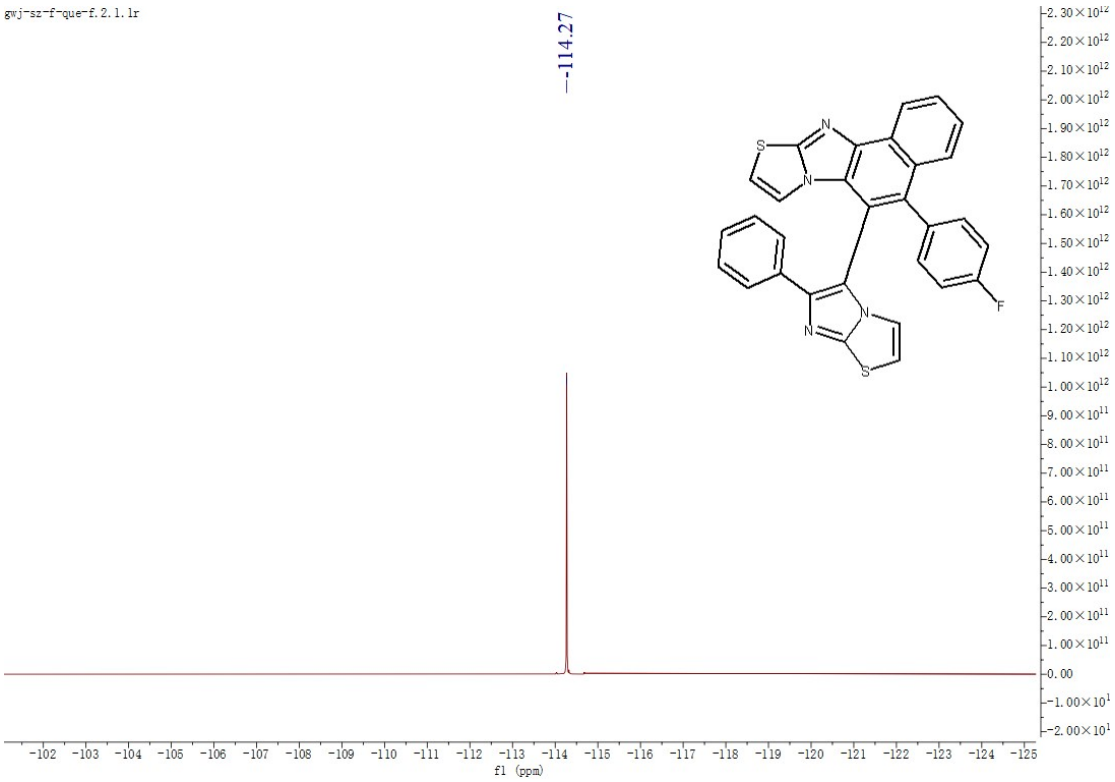
yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**25**):



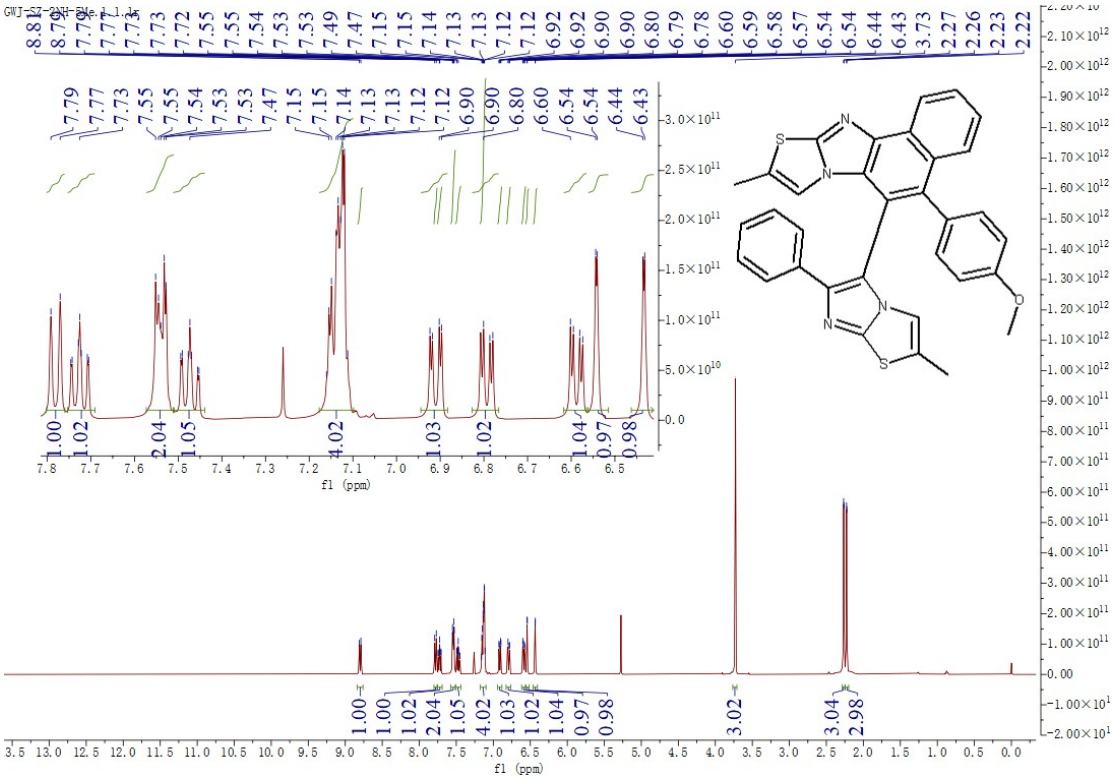


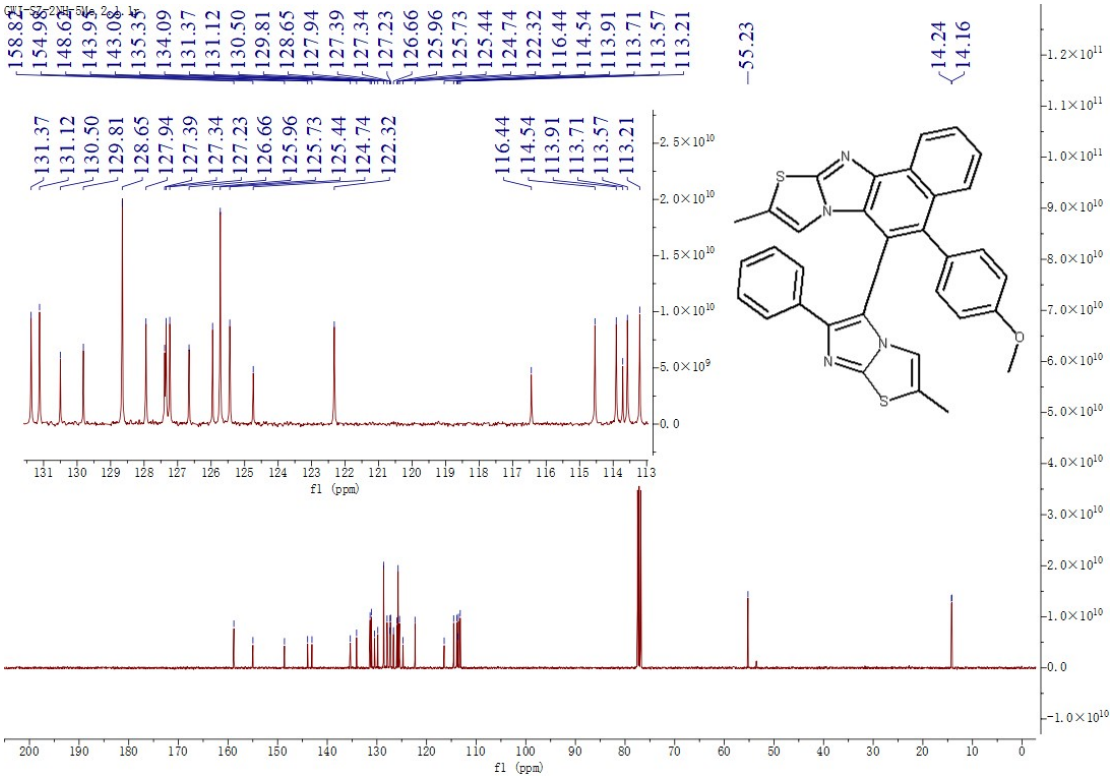
5-(4-fluorophenyl)-6-(6-phenylimidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**26**):



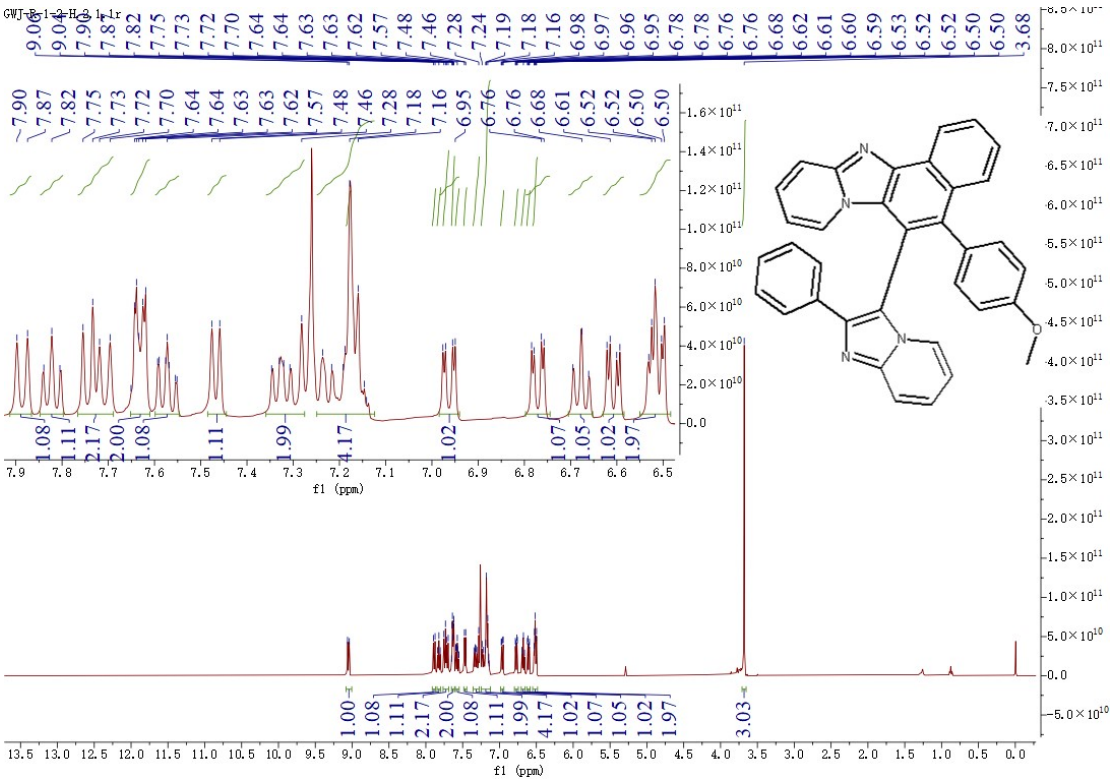


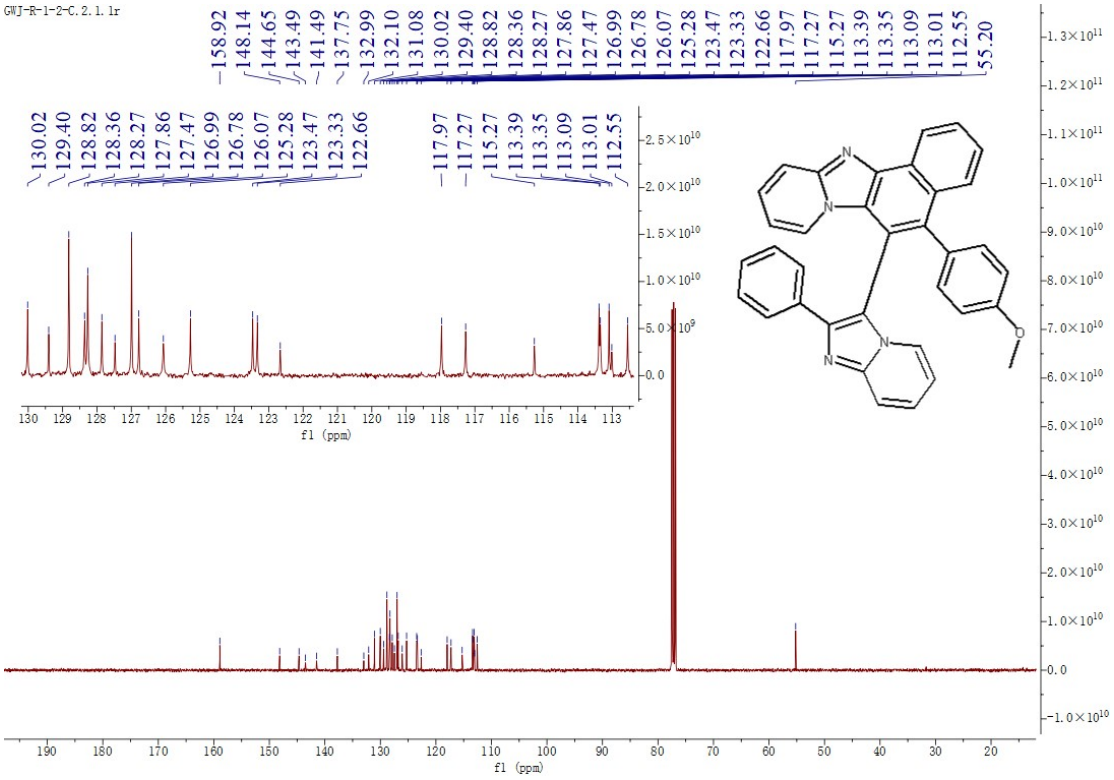
5-(4-methoxyphenyl)-9-methyl-6-(2-methyl-6-phenylimidazo[2,1-*b*]thiazol-5-yl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**27**):



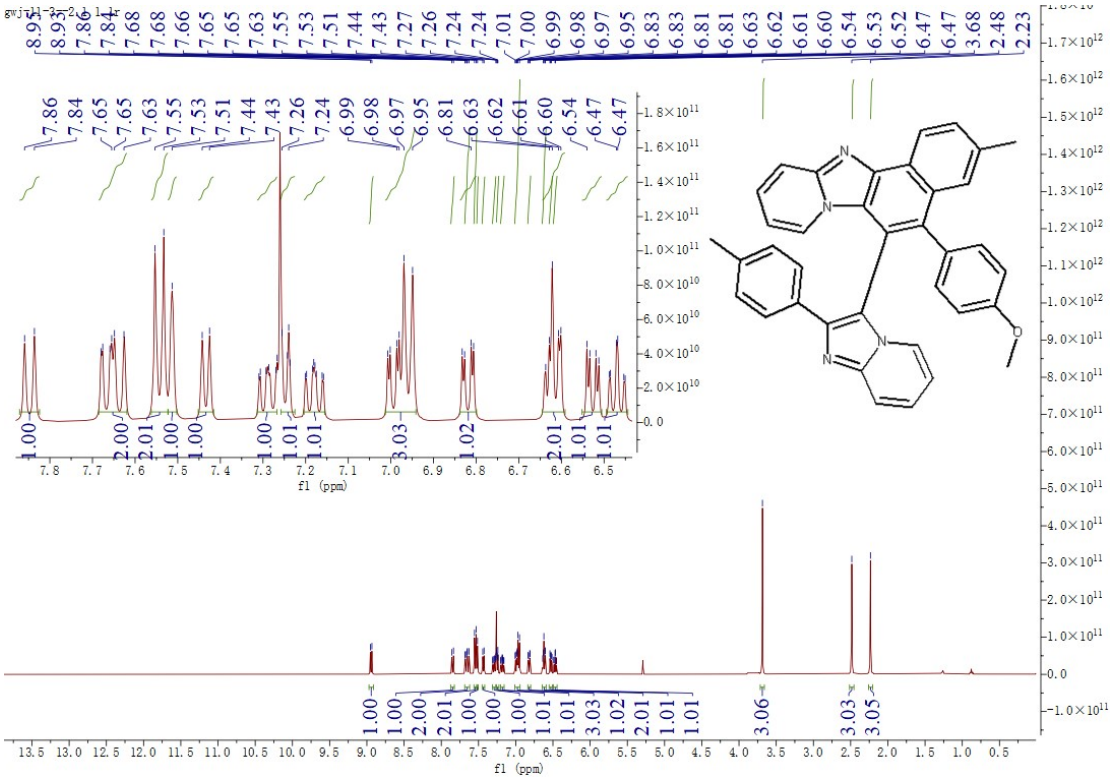


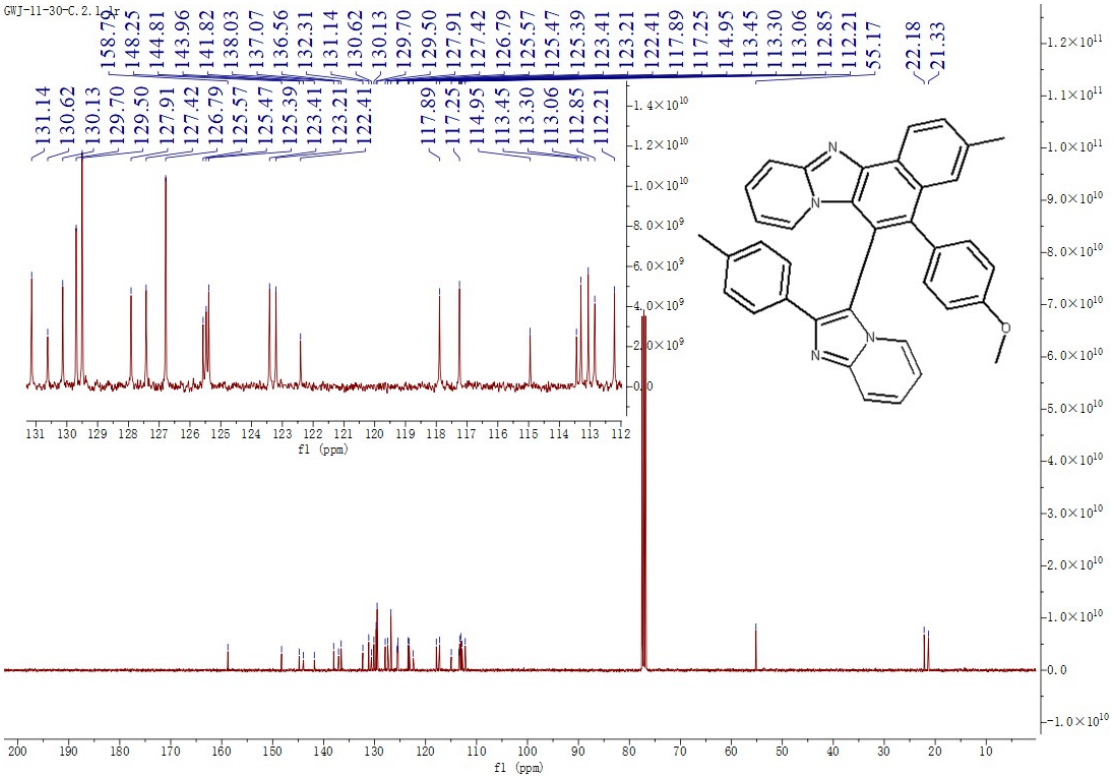
5-(4-methoxyphenyl)-6-(2-phenylimidazo[1,2-*a*]pyridin-3-yl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**28**):



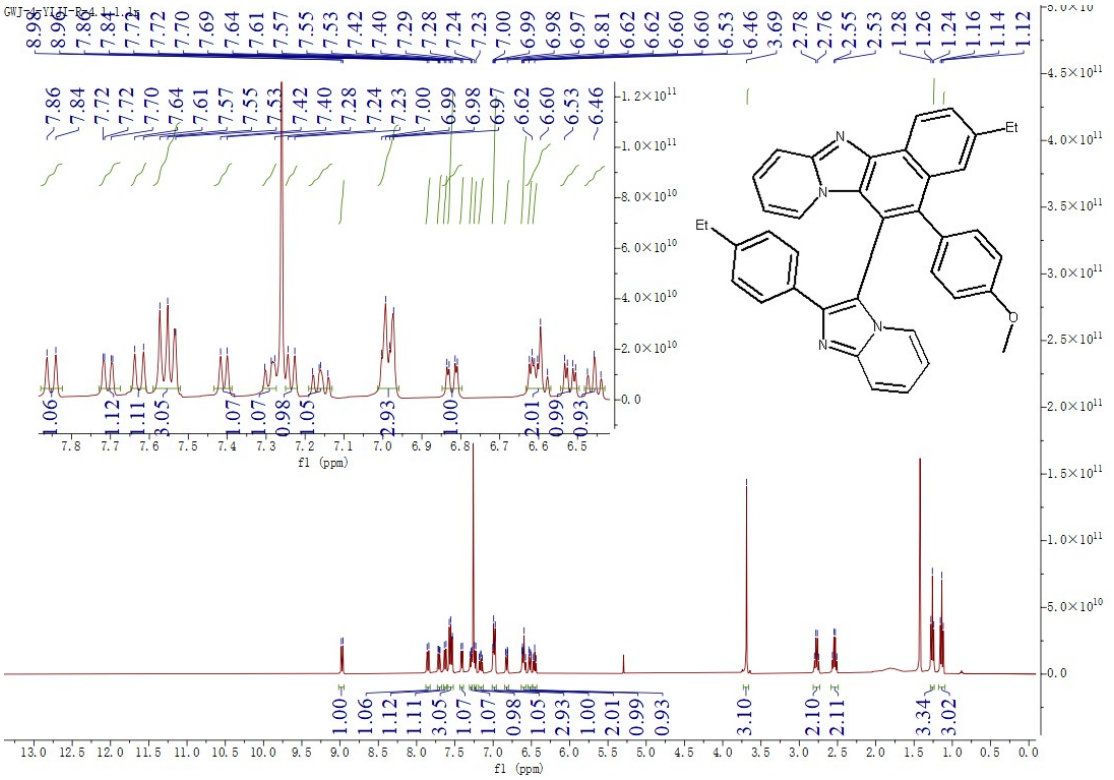


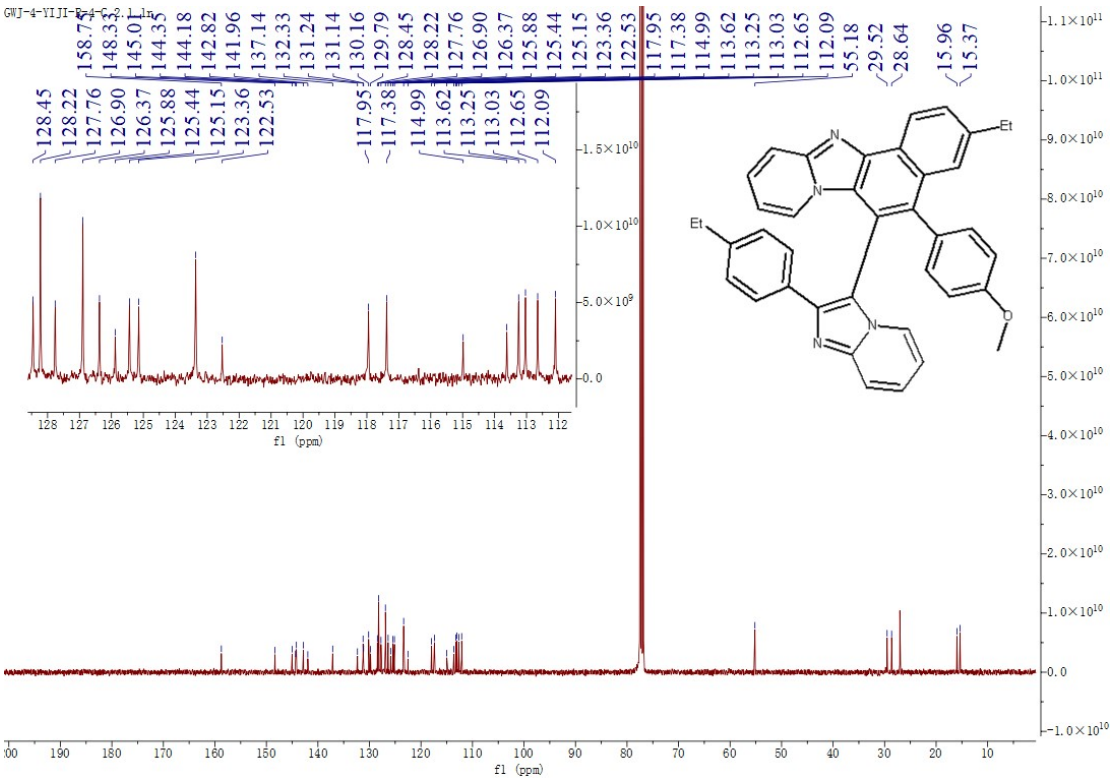
5-(4-methoxyphenyl)-3-methyl-6-(2-(*p*-tolyl)imidazo[1,2-*a*]pyridin-3-yl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**29**):



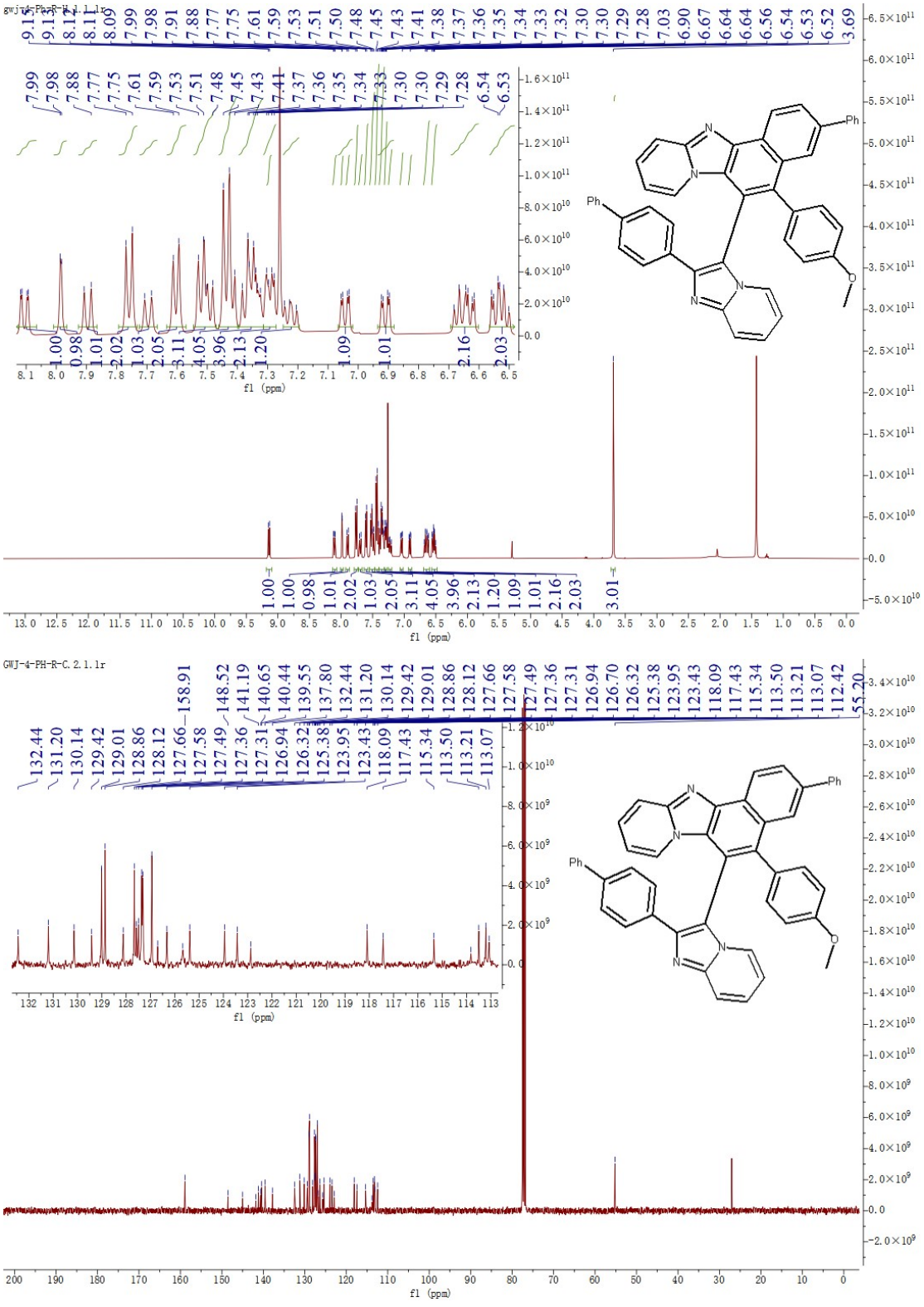


3-ethyl-6-(2-(4-ethylphenyl)imidazo[1,2-*a*]pyridin-3-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**30**):

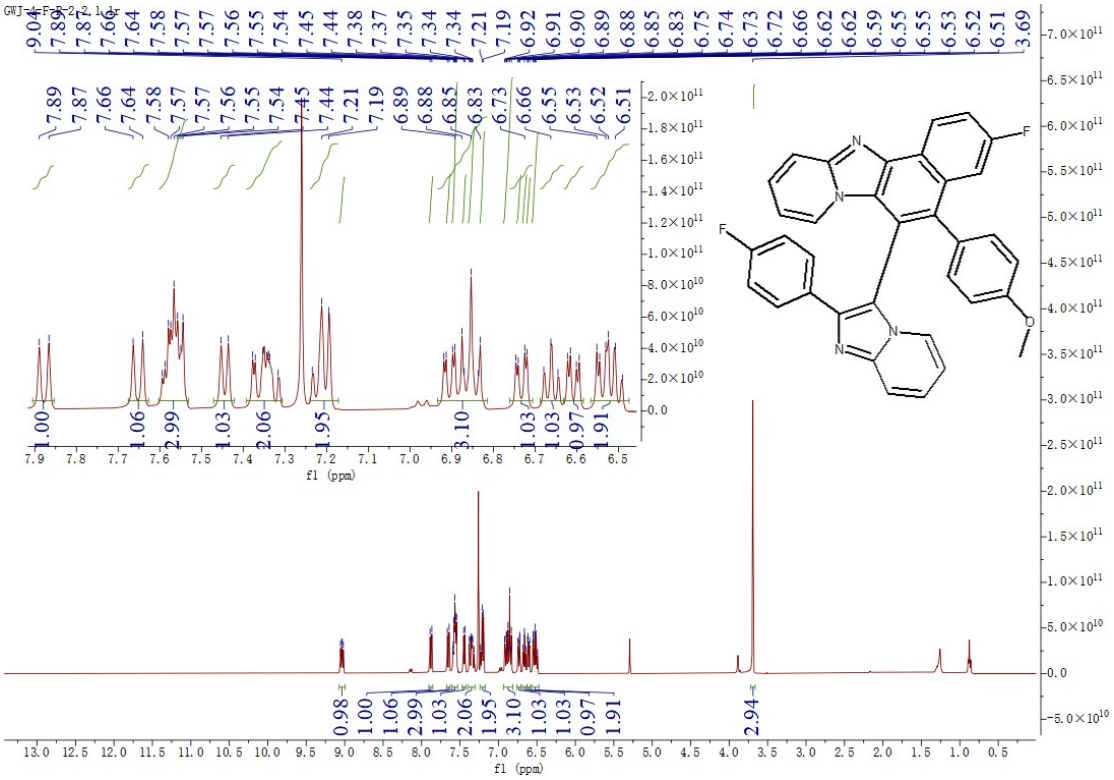


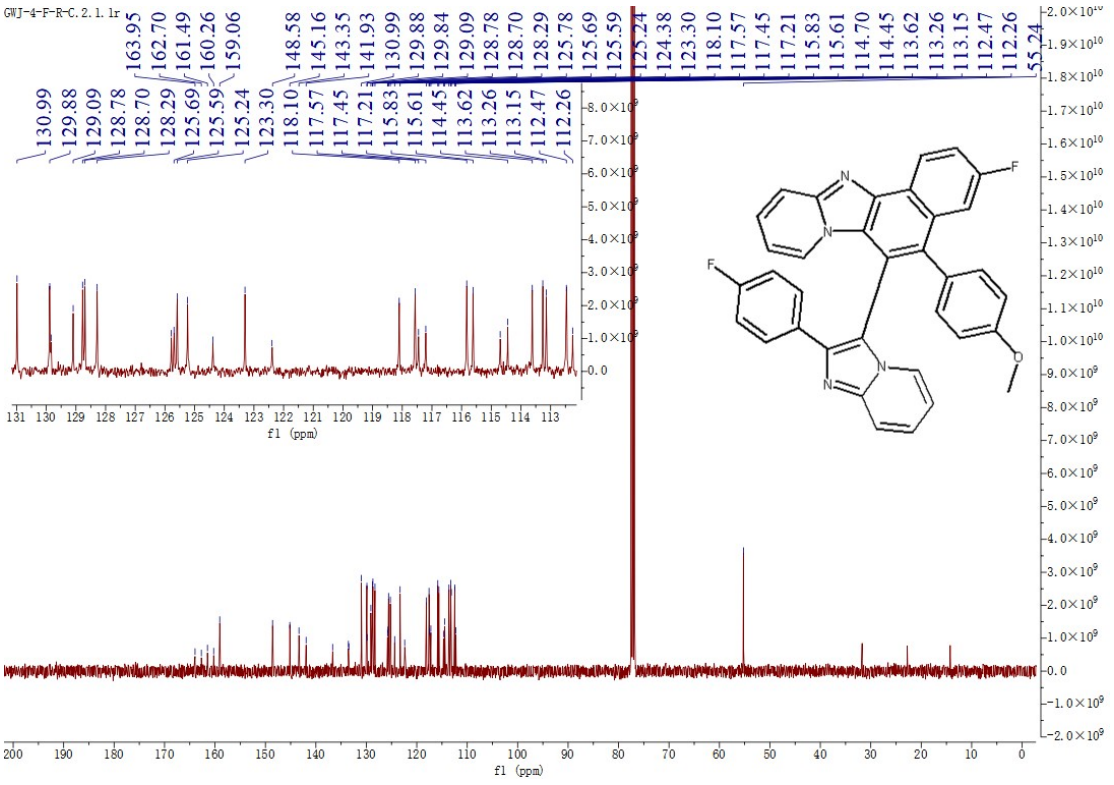


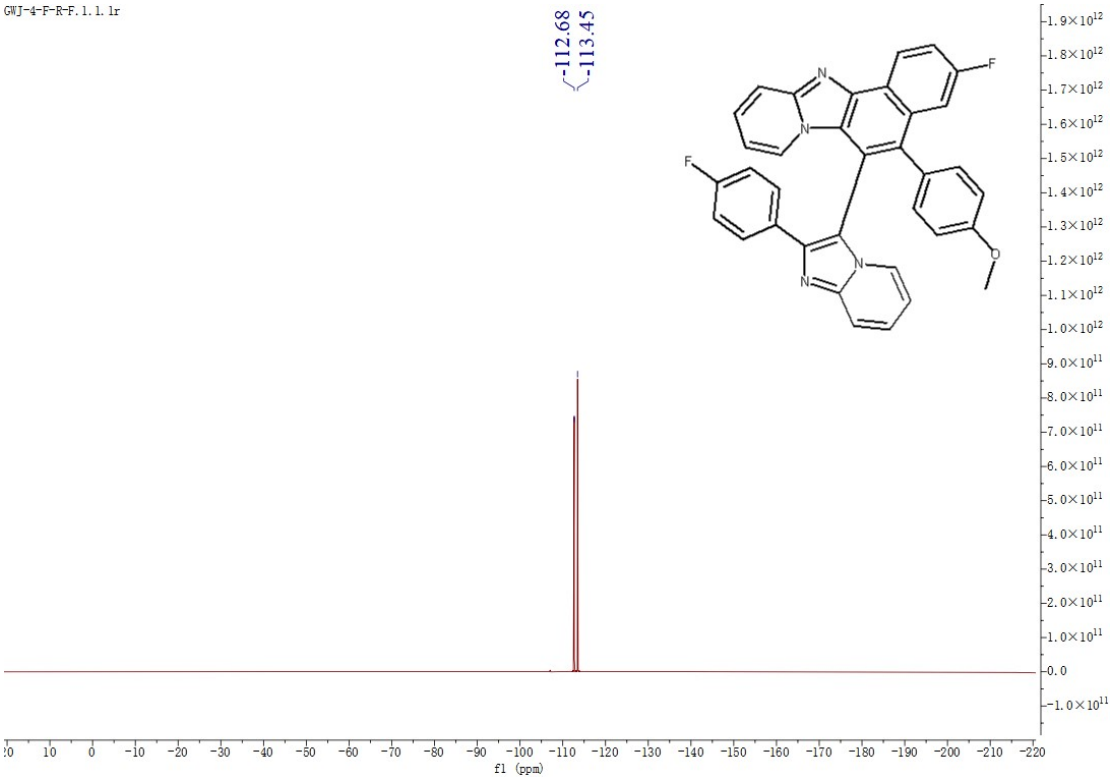
6-(2-([1,1'-biphenyl]-4-yl)imidazo[1,2-*a*]pyridin-3-yl)-5-(4-methoxyphenyl)-3-phenylnaphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**31**):



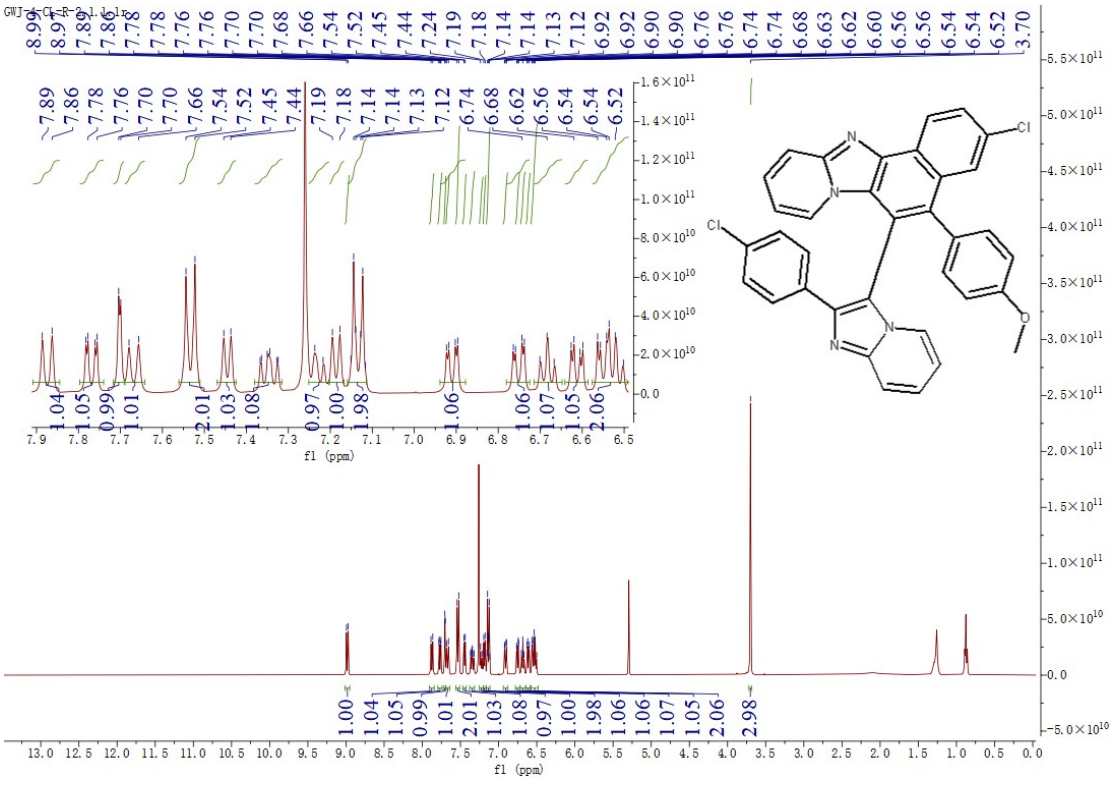
3-fluoro-6-(2-(4-fluorophenyl)imidazo[1,2-*a*]pyridin-3-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**32**):

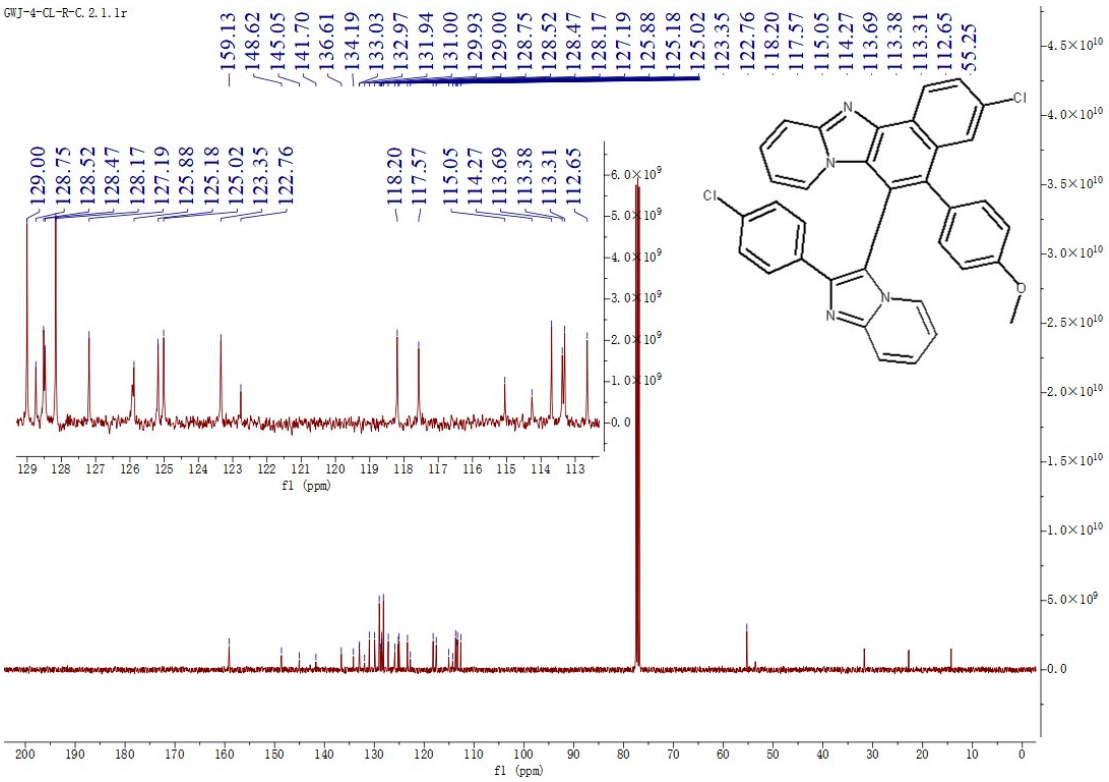




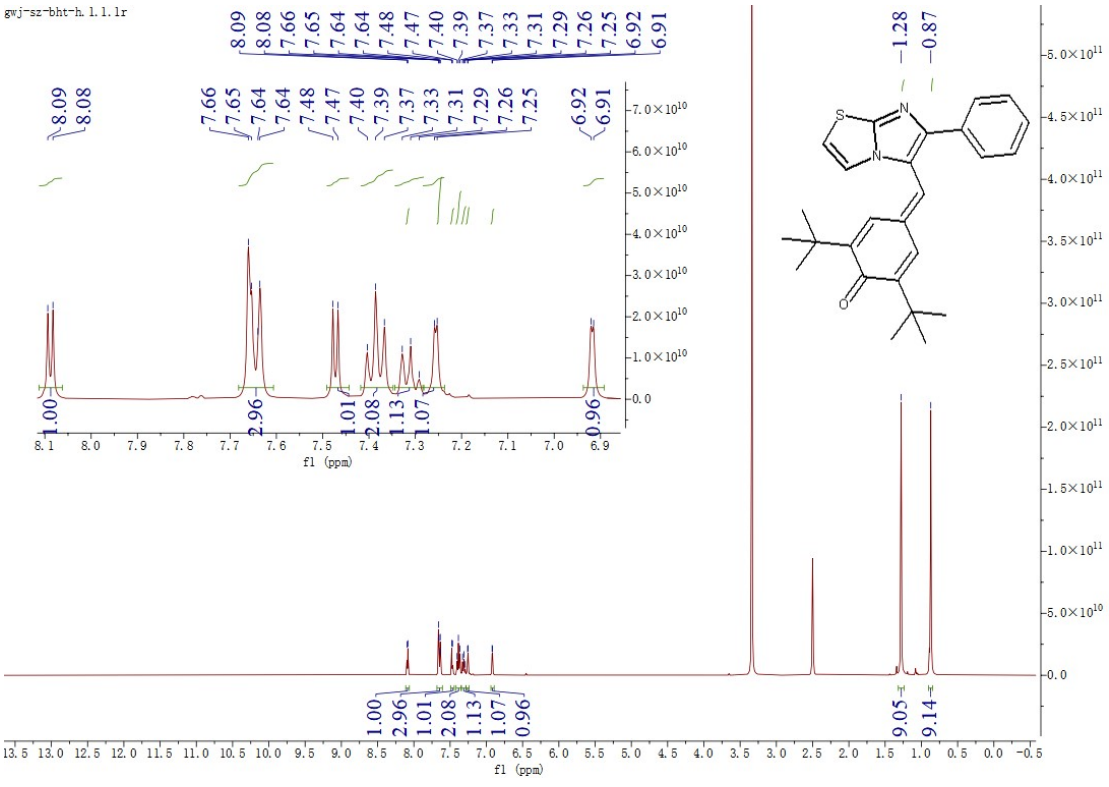


3-chloro-6-(2-(4-chlorophenyl)imidazo[1,2-*a*]pyridin-3-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**33**):

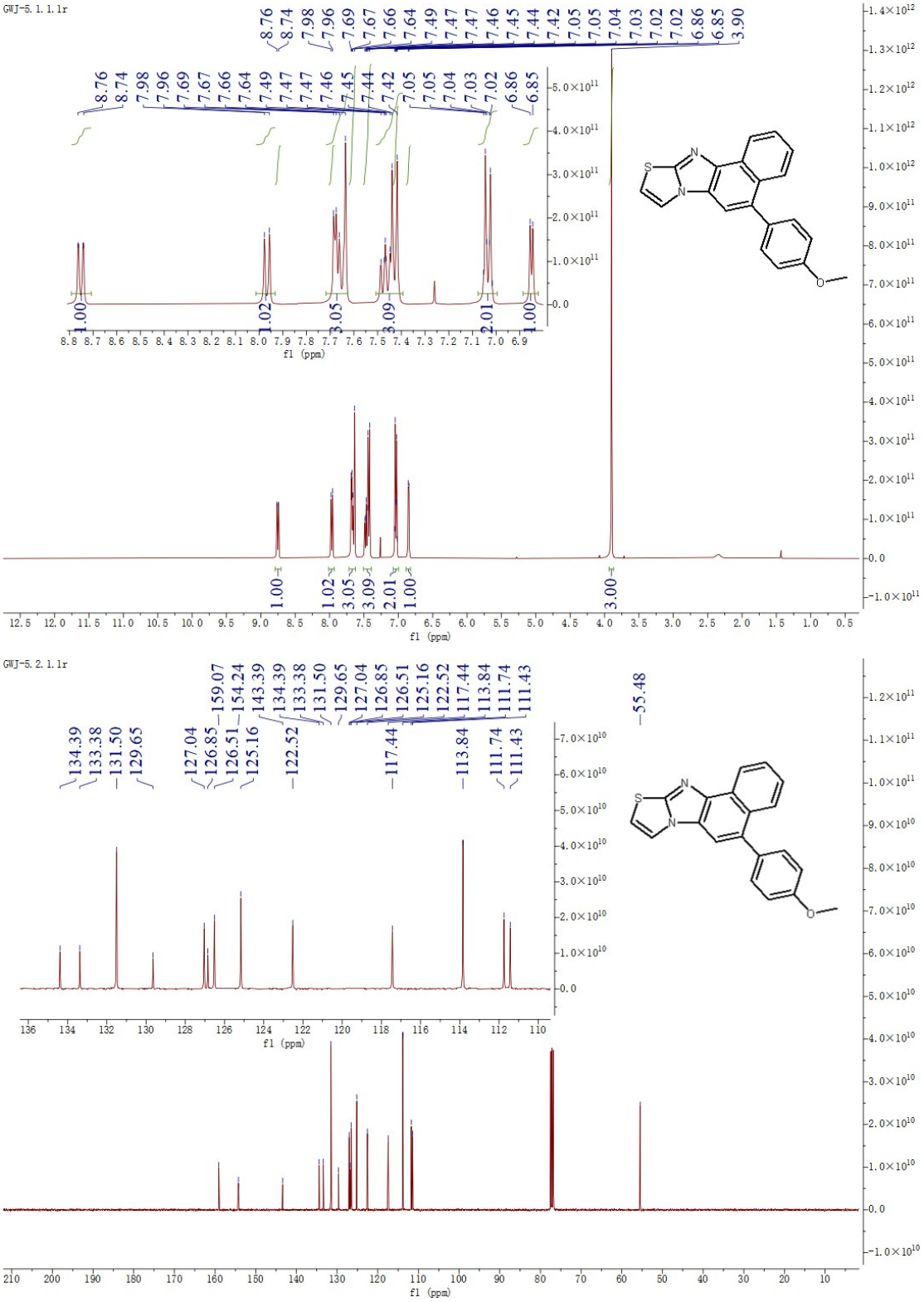




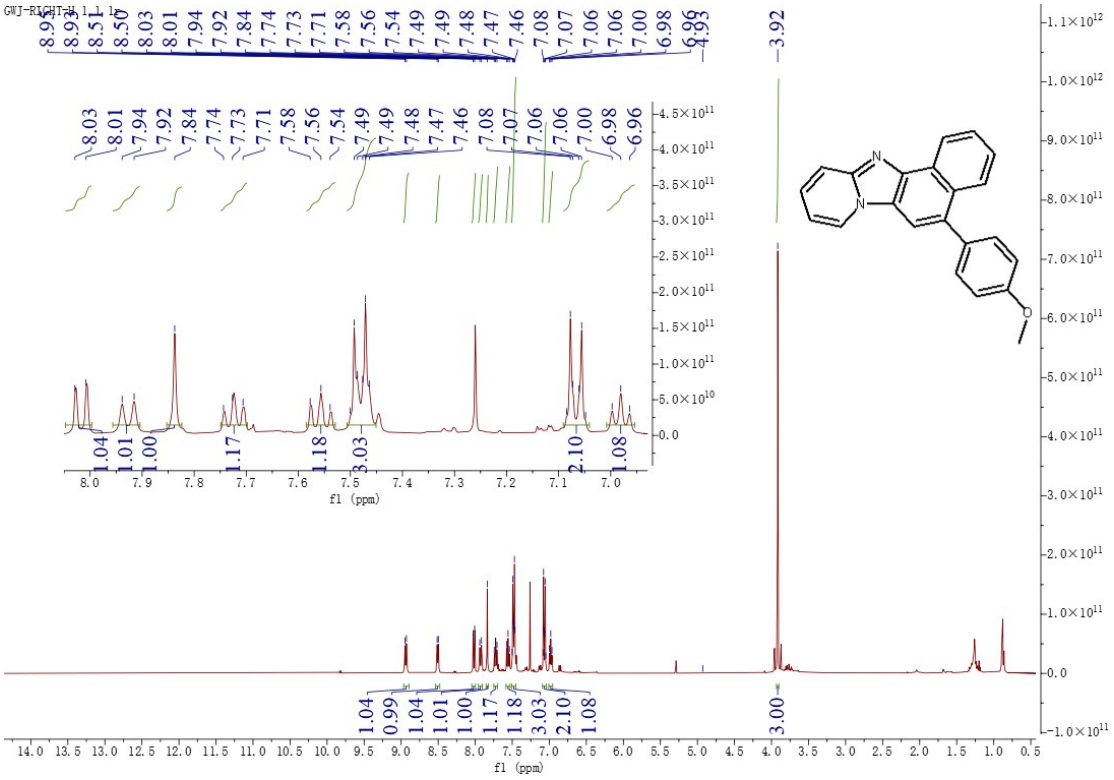
2,6-di-*tert*-butyl-4-((6-phenylimidazo[2,1-*b*]thiazol-5-yl)methylene)cyclohexa-2,5-dien-1-one (**34**):

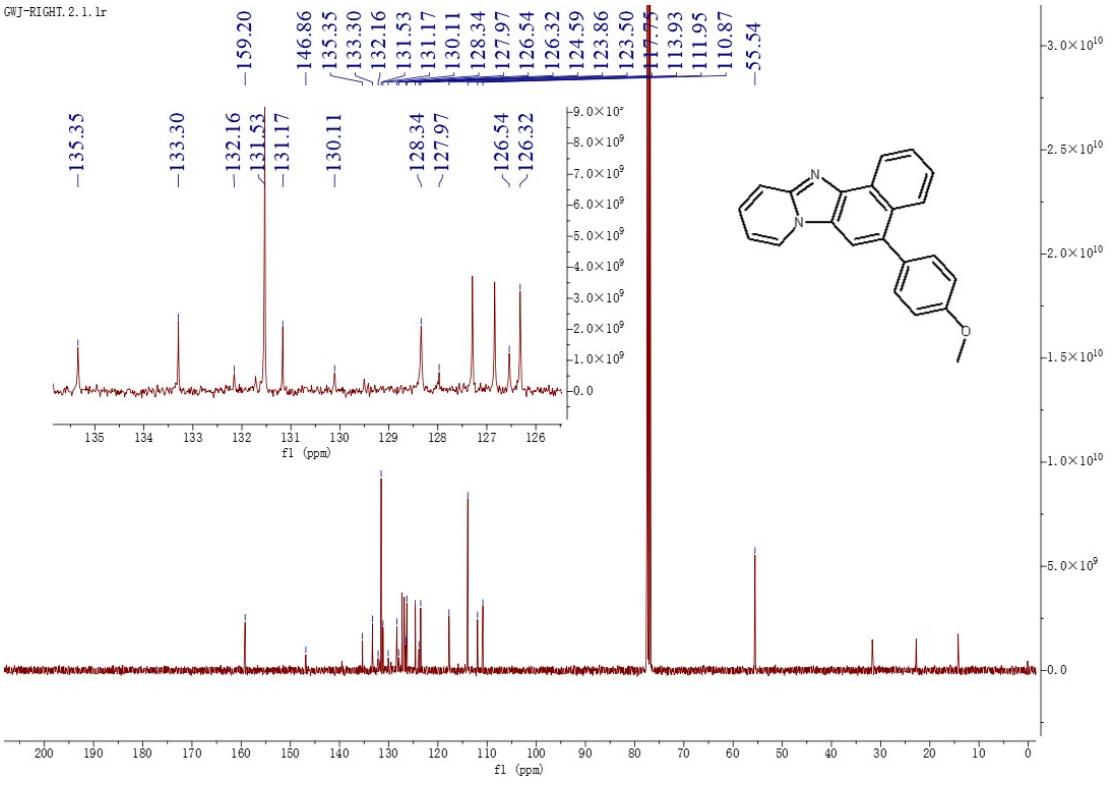


7-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**35**)

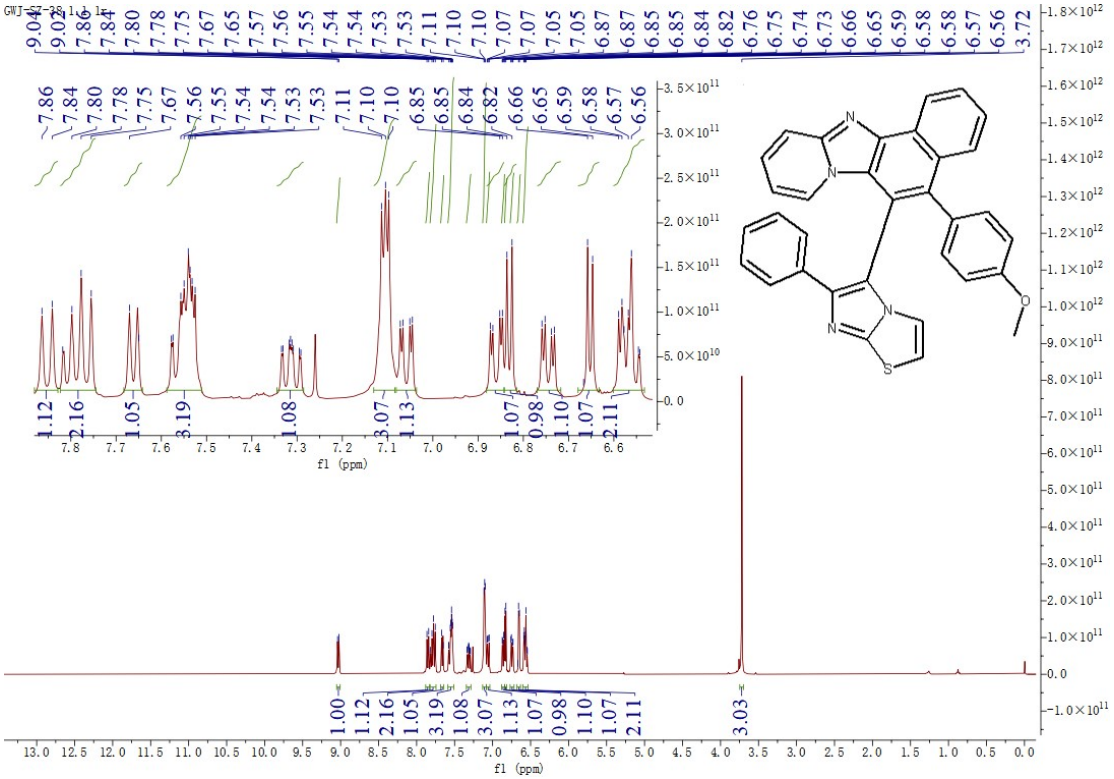


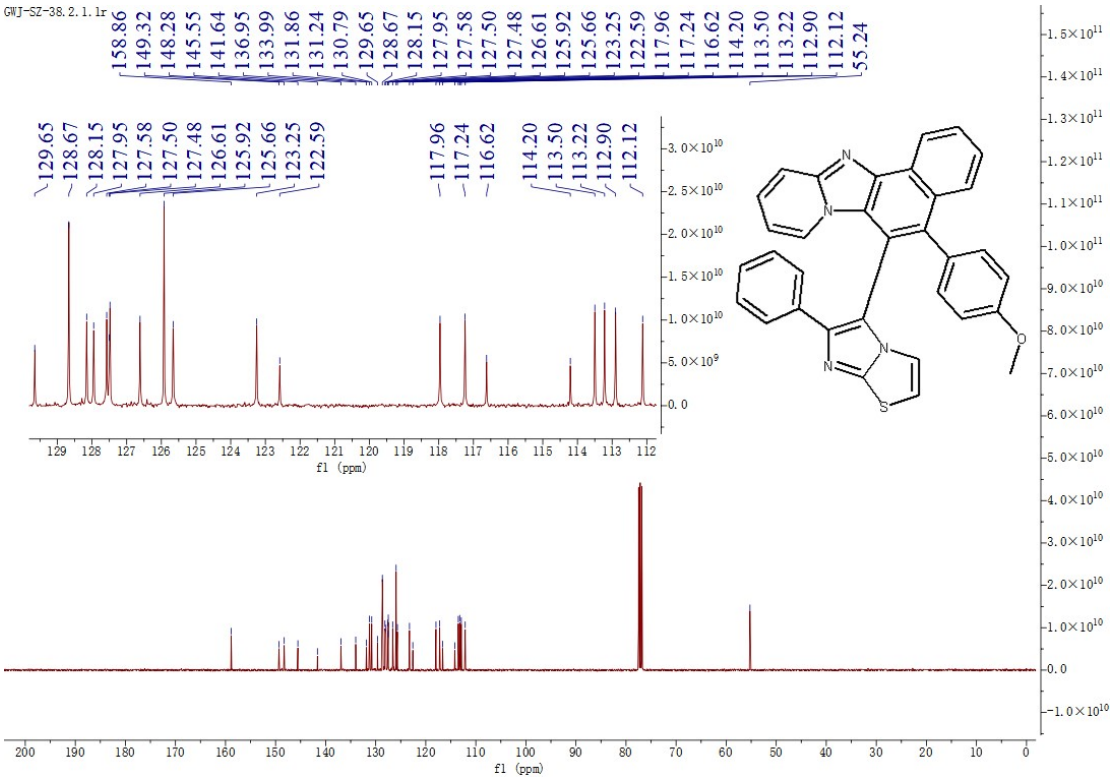
5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridine (**36**):



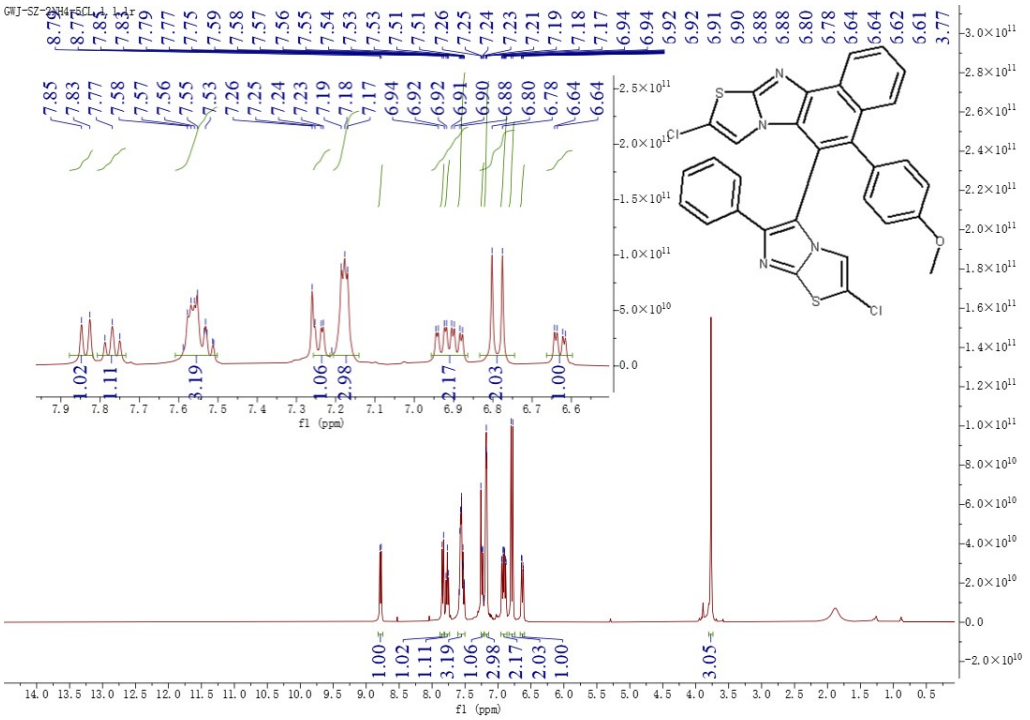


5-(5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[1,2-*a*]pyridin-6-yl)-6-phenylimidazo[2,1-*b*]thiazole (**37**):





9-chloro-6-(2-chloro-6-phenylimidazo[2,1-*b*]thiazol-5-yl)-5-(4-methoxyphenyl)naphtho[1',2':4,5]imidazo[2,1-*b*]thiazole (**S-5**):



6,6'-bis(4-(benzyloxy)phenyl)-5,5'-biimidazo[2,1-b]thiazole (**S-10**)